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Rodrigue Lelotte

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**THÈSE DE DOCTORAT**

**DE L'UNIVERSITÉ PSL**

Préparée à l'Université Paris-Dauphine

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multimarge**

Soutenue par

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<sup>1</sup>Que voulez-vous, mon expulsion du C606 m'avait laissé un goût amère.

<sup>2</sup>Voir note *infra*.

*signes sans faute, sans vérité, sans origine, offert à une interprétation active* »? Je divague, me diras-tu — encore une vieille astuce de philosophe, un enième faux-fuyant qui tire sa fuite fictive d'un arrière-monde tout aussi fictif. Ce serait cependant, à mon sens, mésentendre la vocation de la philosophie, qui ne tire pas sa vérité de *ses* productions — nous le savons, elles sont toutes fausses — mais de *sa* production, c'est-à-dire du seul fait qu'elle produit. Comme à remuer la langue pour lui faire avouer ses secrets, ses béances — comme à jouer avec un théorème mathématique pour le simple plaisir du *jeu*. En somme, comme le disait si bien Heidegger, nous ne pouvons rien faire de la philosophie, car c'est *elle* qui fait quelque chose de nous. Pure passivité — temps inerte. Mais, laissons-ça de côté, mon temps est ici très limité<sup>3</sup>. Maintenant, je dirais autrement — et par-là, j'en dirais plus. Ce qui seul peut faire *effraction*, au sens de cet inédit qui raccommode la maille d'un temps en déliquescence, d'un temps inerte déjà joué, en somme d'un temps sans histoire et sans durée propre, c'est l'*Autre*. Tu connais bien mes diverses obédiences philosophiques — ceci est pour moi une évidence : le temps, c'est l'*Autre*. Tu n'auras aucun mal à comprendre, lévinassien aguerri que tu es devenu. *Au comptoir donc...* où deux amis tordent le cou à Cronos. Que de beaux souvenirs, où le temps jaillissait de nous — où le sens naissait de nous.

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<sup>3</sup>Suivant ton précieux conseil, j'avais commencé la rédaction de ces remerciement bien en amont de la rédaction du manuscrit *per se*. Ayant malencontreusement perdu le document correspondant, je me trouve à les réécrire à la hâte. La dureté du réel, que veux-tu...

<sup>4</sup>Langue inconnue.

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# INTRODUCTION (EN FRANÇAIS)

*Dans ce chapitre introductif, nous commençons par introduire un problème générique de physique statistique, celui de la détermination de l'énergie fondamentale d'un système de particules classiques en interaction, et nous montrons comme ce problème peut être exprimé au travers du prisme de la théorie du transport optimal multimarge. Nous rappelons quelques résultats importants concernant le transport optimal multimarge pour les coûts répulsifs issus de la physique. Nous introduisons ensuite une discrétisation de ce problème dans le cas du coût de Coulomb, basée sur l'interprétation physique de la dualité de Kantorovich, et qui consiste à écrire le potentiel de Kantorovich comme un potentiel électrostatique généré par une distribution de charge. Nous énonçons les propriétés de cette charge qui seront démontrées au Chapitre 1. Nous énonçons ensuite les résultats du Chapitre 2 concernant le comportement asymptotique du potentiel de Kantorovich pour les coûts répulsifs. Pour finir, dans la dernière partie de cette introduction, nous présentons les résultats obtenus au Chapitre 3 sur les transitions de phases pour les gaz de Riesz en une dimension d'espace avec interaction à très longue portée.*

## Sommaire du présent chapitre

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## §1. Un problème générique de physique statistique

Nous allons partir d'un problème très général en physique, celui de la détermination de l'*énergie fondamentale* d'un système de particules classiques en interaction. Nous montrons comment ce problème, dont la résolution numérique est *a priori* difficile, peut être réécrit de façon à faire apparaître la théorie du *transport optimal*.

### §1.1. Le problème de l'énergie fondamentale

On se donne un système de  $N$  particules classiques et ponctuelles situées en  $x_1, \dots, x_N \in \mathbb{R}^d$ . Ces particules interagissent entre elles par le biais d'un potentiel d'interaction à deux corps, que nous noterons  $w(x, x')$ , et que nous supposons toujours par la suite être symétrique, c'est-à-dire telle que  $w(x, x') = w(x', x)$ . De surcroît, les particules sont soumises à un potentiel extérieur  $V(x)$ . Suivant les lois de la physique, nous savons qu'à l'équilibre ces particules vont se disposer de telle façon à minimiser l'énergie totale du système. Il s'agit donc de résoudre le problème de minimisation

$$E_{N,w}(V) := \inf_{x_1, \dots, x_N \in \mathbb{R}^d} \left\{ \sum_{1 \leq i < j \leq N} w(x_i, x_j) + \sum_{i=1}^N V(x_i) \right\}. \quad (\text{E})$$

Nous avons mis en indice le potentiel d'interaction  $w$  dans la notation de l'*énergie fondamentale*  $E_{N,w}$ . Autre qu'une simple convention stylistique, la raison en est la suivante. En effet, il faut se rappeler que le potentiel  $w$  est typiquement fixé par la nature des particules que l'on considère. Quand bien même il puisse être artificiellement modifié dans les limites du raisonnable en laboratoire, il faudra garder à l'esprit que ce dernier est habituellement donné comme une prémisses qui échappe *a priori* au bon-vouloir de l'expérimentateur ou du théoricien. Quant au potentiel extérieur  $V$ , il décrit l'action que l'expérimentateur applique au système et il peut donc être plus facilement modifié — toujours dans les limites du raisonnable. Cependant, si nous laissons entendre par notre notation que  $V$  puisse varier, la raison en est différente, comme cela deviendra bientôt apparent.

Le **Problème E** est un problème tout à fait général, qui peut s'écrire autant pour des systèmes moléculaires ou atomiques que pour des systèmes gravitationnels. Si dans cette première partie nous restons à un certain niveau de généralité en ne spécifiant pas la forme du potentiel d'interaction  $w$ , nous nous

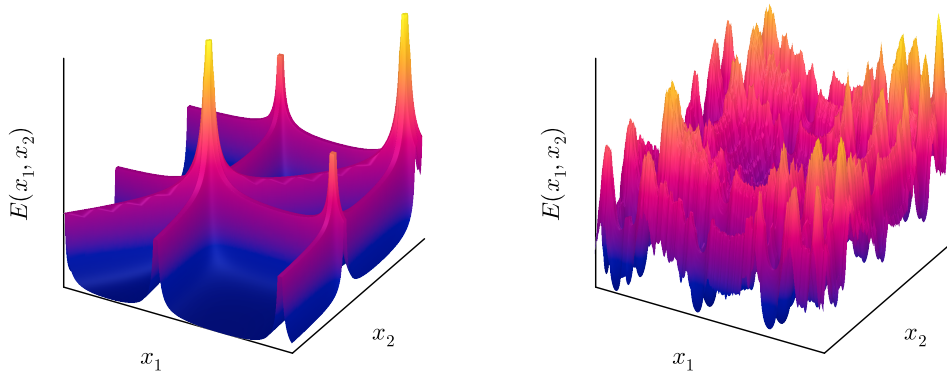


FIGURE 1 : On considère des particules  $x_1, \dots, x_N \in \mathbb{R}$  qui interagissent par le biais du potentiel d'interaction  $|x - x'|^{-1}$ . À gauche, on suppose que  $N = 4$  et que sont fixées arbitrairement deux particules  $x_3$  et  $x_4$ . La surface représente alors l'énergie  $E(x_1, x_2)$  des deux autres particules. *Idem* à droite pour  $N = 50$ , où sont fixées les particules  $x_3, \dots, x_{50}$ . On remarque que l'énergie  $E(x_1, x_2)$  présente un nombre important de *minima* locaux.

intéresserons par la suite principalement aux systèmes de particules chargées en interaction. On peut par exemple penser à des ions qui interagissent entre eux au travers le *potentiel de Coulomb*, comme cela est le cas dans l'étude du coeur des *naines blanches* en astrophysique [VH19].

Il serait tentant de s'essayer à résoudre le **Problème E** par la voie analytique. À l'exception de quelques cas particuliers et du reste marginaux, une telle entreprise devra bien vite se confronter à l'illusion qui l'a fait naître. On peut affirmer — quoique que l'inventivité de certains esprits peut parfois nous surprendre — que la résolution analytique d'un tel problème est en toute généralité impossible, du moins sans information sur la nature des minimiseurs. Si le mathématicien s'en trouvera alors peut-être contrarié, le physicien se suffira d'une résolution approchée par la voie d'un calcul numérique. Cependant, les choses ne sont pas forcément plus simple numériquement. Notons tout d'abord que, dans le cadre de la physique statistique du moins, le nombre de particules  $N$  est typiquement espéré de l'ordre du *nombre d'Avogadro*. Espérance immédiatement déçue par les limitations de nos ordinateurs modernes et *a priori* futurs. On devra donc se suffire de quelques centaines ou milliers de particules. La difficulté d'un tel problème réside dans le fait bien connu qu'en grande dimension l'énergie totale que l'on cherche à minimiser présentera généralement un nombre important de *minima* locaux, bêtes noires des algorithmes d'optimisation (voir Figure 1). Si la chose n'est pas du tout impossible — en l'occurrence, elle est tout à fait possible et réalisée quotidiennement par la communauté physique — elle n'est pas tout à fait aisée.

Si nous pourrions en rester là, nous allons cependant prendre un chemin différent. On notera tout d'abord que  $E_{N,w}(V)$  est une fonction concave du

potentiel extérieur, en tant que minimisation d'une fonction linéaire. On peut alors considérer la *transformée de Legendre*<sup>5</sup> de l'énergie fondamentale  $E_{N,w}(V)$  vue comme une fonctionnelle du potentiel extérieur  $V$ . S'il peut sembler de prime abord qu'une telle idée est quelque peu arbitraire, il faudra se rappeler qu'il s'agit là d'une technique assez habituelle en physique — *e.g.* en mécanique classique, où elle permet de passer du formalisme *Lagrangien* à partir du formalisme *Hamiltonien* et *vice-versa*, ou encore en physique statistique, où l'entropie est la transformée de Legendre de l'énergie libre. Pour une discussion sur la transformée de Legendre en physique, on pourra se référer à [ZRM09]. Nous considérons donc  $F_{N,w}$  la transformée de Legendre de  $E_{N,w}$ , donnée par

$$F_{N,w}(\rho) := \sup_{V \in \mathcal{V}} \left\{ E_{N,w}(V) - \int_{\mathbb{R}^d} V(x)\rho(x)dx \right\}. \quad (\text{F})$$

Ici le *supremum* est pris sur un espace fonctionnel  $\mathcal{V}$  qui caractérise l'ensemble des potentiels extérieurs  $V$  admissibles, et  $\rho$  est un élément de son dual topologique  $\mathcal{V}'$ , typiquement une mesure. On pourra supposer, pour fixer les idées, que  $\mathcal{V}$  est l'ensemble des fonctions continues et bornées, et par conséquent que son dual  $\mathcal{V}'$  est bien connu et donné par l'ensemble des mesures de Radon. Une fois la transformée de Legendre  $F_{N,w}$  de l'énergie fondamentale  $E_{N,w}$  définie, l'astuce consiste à reprendre la transformée de  $F_{N,w}$  elle-même. Il est connu, par un théorème classique d'analyse convexe<sup>6</sup> [Sim11], que sous de bonnes hypothèses — typiquement vérifiées pour des potentiels d'interaction  $w$  semicontinus inférieurement — la transformée de Legendre est une involution. Par conséquent, on obtient

$$E_{N,w}(V) = \sup_{\rho \in \mathcal{V}'} \left\{ F_{N,w}(\rho) - \int_{\mathbb{R}^d} V(r)\rho(r)dr \right\}. \quad (\text{D})$$

Nous avons donc obtenu une réécriture du **Problème E** de la détermination de l'énergie fondamentale. *Ce problème est-il plus simple? A priori*, il semblerait que ce nouveau problème soit tout aussi, si ce n'est plus compliqué que notre problème originel. Après tout, nous sommes passés d'un problème de minimisation sur l'ensemble  $(\mathbb{R}^d)^N$  des configurations à un problème variationnel sur un espace de mesure plus gros — et qui de surcroît doit être discrétisé lors de son traitement numérique. Cependant, le **Problème D** présente quelques avantages. On notera d'abord qu'il s'agit d'un problème variationnel convexe. De façon plus intéressante, la fonctionnelle  $F_{N,w}$  étant indépendante du potentiel extérieur  $V$ , on peut s'imaginer qu'une connaissance parfaite de  $F_{N,w}$  pourrait éventuellement nous fournir une procédure de résolution du **Problème E** qui serait en

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<sup>5</sup>On rappelle que, étant donné un espace de Banach  $X$ , la *transformée de Legendre* d'une fonction  $F : X \rightarrow \mathbb{R}$  est définie comme la fonction  $F^* : X^* \rightarrow \mathbb{R}$  définie sur le dual topologique  $X^*$  de  $X$  munit de son crochet de dualité  $\langle \cdot, \cdot \rangle$  par l'égalité (selon la convention que cette transformée est convexe)

$$F^*(v) = \sup_{u \in X} \{F(u) - \langle u, v \rangle\}.$$

<sup>6</sup>Le dit théorème est le *théorème de Fenchel–Moreau*. Sous l'hypothèse que  $F$  est une fonction propre, convexe, et semicontinue inférieurement, alors  $(F^*)^* = F$ .

quelque sorte *universelle* en le potentiel extérieur  $V$ , ce dernier n'apparaissant que linéairement dans **Problème D**. Il nous reste donc à en savoir un peu plus sur la fonctionnelle  $F_{N,w}$ . En pratique, on cherchera ensuite à approcher  $F_{N,w}$  par une fonctionnelle plus simple.

*Remarque.* Dans le langage de la théorie de l'optimisation et de l'analyse convexe, on remarquera que le **Problème D** correspond exactement à un *principe de dualité* pour l'énergie fondamentale  $E_{N,w}$ . On rappelle qu'un tel principe énonce en substance qu'à chaque problème de minimisation convexe on peut associer un problème de maximisation équivalent, dit *problème dual*. Par abus de langage, on pourra dire que  $F_{N,w}$  est la fonctionnelle *duale* de l'énergie fondamentale  $E_{N,w}$ . Du reste, cette remarque n'a rien d'étonnant, étant pensé à fond que la transformée de Legendre est exactement le pendant analytique de la dualité telle que définie en optimisation [Sim11].

### Détermination de la fonctionnelle $F_{N,w}$

*Qui est donc la fonctionnelle  $F_{N,w}$  ?* Il se trouve que nous pouvons donner une formulation beaucoup plus explicite de  $F_{N,w}$ , et qui nous mènera très vite au cœur de notre propos, c'est-à-dire à la théorie du transport optimal. Nous faisons d'abord remarquer que, dans la définition de l'énergie fondamentale  $E_{N,w}$ , il est toujours possible de substituer à la minimisation sur l'espace des configurations  $(x_1, \dots, x_N) \in \mathbb{R}^{dN}$  une minimisation sur l'ensemble des mesures de probabilités  $\mathbb{P}(x_1, \dots, x_N)$  sur  $\mathbb{R}^{dN}$ . En d'autres termes, nous passons à une vision probabiliste de nos particules — ce qui n'a non seulement rien d'étonnant du point de vue de la physique statistique, mais ce qui sera de surcroît obligatoire dans le cas de la température positive, comme nous le verrons plus tard. Comme nous avons supposé que le potentiel d'interaction  $w$  était symétrique, nous pouvons toujours nous restreindre aux distributions  $\mathbb{P}$  symétriques par rapport aux particules, c'est-à-dire telle que  $\mathbb{P}(x_1, \dots, x_N) = \mathbb{P}(x_{\sigma(1)}, \dots, x_{\sigma(N)})$  pour toute permutation d'indices  $\sigma \in \mathfrak{S}_N$ . On trouve alors que

$$E_{N,w}(V) = \inf_{\mathbb{P}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) + \int_{\mathbb{R}^d} V(x) \rho_{\mathbb{P}}(x) \right\}.$$

Nous avons utilisé ici que le potentiel extérieur  $V$  est un potentiel à un corps. En particulier, l'énergie potentielle d'une distribution de particules  $\mathbb{P}$  peut se réécrire comme

$$\int_{\mathbb{R}^{dN}} \sum_{i=1}^N V(x_i) d\mathbb{P}(x_1, \dots, x_N) = \int_{\mathbb{R}^d} V(x) \rho_{\mathbb{P}}(x) \quad (1)$$

où  $\rho_{\mathbb{P}} \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  est la mesure positive définie par

$$\rho_{\mathbb{P}}(x) = N \int_{\mathbb{R}^{d(N-1)}} \mathbb{P}(x, x_2, \dots, x_N).$$

*Remarque.* La fonction  $\rho_{\mathbb{P}}$  est appelée *densité électronique* dans le cas des systèmes de particules chargées à l'exemple des électrons. On l'appellera plus

simplement la *densité* ("de notre système"). On remarquera que cette densité est normalisée telle que  $\int_{\mathbb{R}^d} \rho_{\mathbb{P}} = N$ . Si les particules sont distribuées selon la mesure de probabilité  $\mathbb{P}$ , la fonction  $\rho_{\mathbb{P}}$  correspond à la densité moyenne de charge en espace. On pourra penser à elle comme d'une représentation moyenne de l'état de notre système à la manière d'un *champ moyen*. Ainsi, une particule située très loin du système ne "verra" pas toutes les configurations géométriques particulières dans lesquelles peut se trouver le dit système, mais plutôt une version moyennée correspondant exactement à  $\rho_{\mathbb{P}}$ .

Une fois l'énergie fondamentale  $E_{N,w}$  réécrite comme un *infimum* sur l'ensemble des distributions  $\mathbb{P}$  sur l'espace des configurations  $(\mathbb{R}^d)^N$ , nous allons séparer cette minimisation en deux étapes. Dans un premier temps, nous allons *fixer* une densité  $\rho$ , et nous allons considérer la minimisation de l'énergie sur l'ensemble des distributions  $\mathbb{P}$  qui vérifient la contrainte  $\rho_{\mathbb{P}} = \rho$ . Dans un second temps, nous optimisons sur la densité  $\rho$  précédemment fixée. On obtient alors

$$E_{N,w}(V) = \inf_{\rho} \left\{ \int_{\mathbb{R}^d} V(x) \rho(x) + \inf_{\rho_{\mathbb{P}}=\rho} \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) \right\}, \quad (2)$$

où le premier *infimum* est pris sur l'ensemble des mesures positives  $\rho$  telle que  $\int_{\mathbb{R}^d} \rho = N$ . Une telle réécriture fait écho au *problème variationnel de Lieb* et à la *fonctionnelle de Levy-Lieb* [Lev79; Lie83; HT22], comme nous le verrons plus bas lorsque nous parlerons de la *théorie de la fonctionnelle de la densité* (DFT). Nous insérons maintenant cette réécriture dans la définition de  $F_{N,w}$  comme transformée de Legendre de l'énergie  $E_{N,w}$  — qui est d'ailleurs équivalente au *problème variationnel de Hohenberg-Kohn* en DFT [HT22; HK64]. Nous pouvons alors *formellement* intervertir le *supremum* de la transformée de Legendre et les *infimums*, de façon à obtenir l'égalité

$$F_{N,w}(\rho) = \inf_{\mathbb{P}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) + \sup_{V \in \mathcal{V}} \int_{\mathbb{R}^d} V(x) (\rho_{\mathbb{P}}(x) - \rho(x)) \right\}.$$

On remarque ensuite que le *supremum* sur l'ensemble des potentiels  $V \in \mathcal{V}$  est nul si et seulement si  $\rho_{\mathbb{P}} = \rho$ , et infini dans le cas contraire. Nous sommes donc par conséquent implicitement forcés à ne considérer que l'ensemble des distributions  $\mathbb{P}$  dont la densité électronique est  $\rho$ , c'est-à-dire telles que  $\rho_{\mathbb{P}} = \rho$ . Nous obtenons alors finalement

$$F_{N,w}(\rho) = \inf_{\rho_{\mathbb{P}}=\rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) \right\}. \quad (F)$$

Lorsque  $\rho$  n'est pas une mesure positive telle que  $\int_{\mathbb{R}^d} \rho = N$ , on peut poser  $F_{N,w}(\rho) = \infty$ . Nous sommes arrivés à une expression satisfaisante de la transformée de Legendre  $F_{N,w}$  de l'énergie fondamentale. Nous voyons directement qu'étant donnée une densité  $\rho$ , la quantité  $F_{N,w}(\rho)$  correspond à l'énergie d'interaction minimale qui puisse être atteinte par un système de  $N$  particules sous contrainte que sa densité électronique soit égale à  $\rho$ . Nous allons maintenant pouvoir faire apparaître la théorie du transport optimal — si le lecteur averti ne l'a pas déjà démasqué.

## La fonctionnelle $F_{N,w}$ et le transport optimal

Nous pouvons maintenant affirmer que la fonctionnelle  $F_{N,w}$ , que nous avons définie comme la transformée de Legendre de l'énergie fondamentale  $E_{N,w}$  vue comme une fonctionnelle du potentiel extérieur  $V$ , s'exprime — dans le cas des potentiels d'interactions  $w$  symétriques — comme un problème de *transport optimal multimarges* [BDPGG12; CFK13; DMGN15; FG22] où le nombre de marges est donné par le nombre de particules  $N$ , où toutes les marges sont contraintes à la même densité  $\rho$ , et où pour finir le coût de transport  $c(x_1, \dots, x_N)$  est donnée par l'énergie d'interaction

$$c(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} w(x_i, x_j). \quad (3)$$

Nous rappelons que nous avons introduit le **Problème F** et par conséquent la fonctionnelle  $F_{N,w}$  non par simple goût de l'abstraction, mais bien pour obtenir une réécriture du **Problème E** de la détermination de l'énergie fondamentale  $E_{N,w}(V)$  pour un certain potentiel extérieur  $V$  *a priori* fixé. Nous avons l'espoir que ce nouveau point de vue puisse nous offrir des outils pour résoudre ce problème. Le **Problème F** ne nous semblait pas *a priori* plus simple que le problème initial — mais il présentait quelques avantages par rapport à celui-ci. Maintenant que nous avons une expression tout à fait explicite de la fonctionnelle  $F_{N,w}(\rho)$  comme un problème de transport optimal multimarges, nous pouvons confirmer notre intuition. Il est en effet connu que le transport optimal est un problème compliqué, tant d'un point de vue théorique que numérique. Nous y reviendrons plus en détails par la suite. Cependant, dans une perspective physique où l'efficacité d'un calcul numérique supplante l'idéalité mathématique d'une résolution exacte, rien ne nous empêche de faire appel à des *approximations* de  $F_{N,w}$ . Du reste, c'est là un des cœurs de la *théorie de la fonctionnelle de la densité* (DFT). Cette théorie s'écrit aussi bien en classique qu'en quantique. Dans la partie suivante, nous introduisons brièvement la DFT dans le cas quantique, et nous reviendrons un peu plus tard sur le cas classique.

## §1.2. Théorie de la fonctionnelle de la densité en chimie quantique

La *Théorie de la fonctionnelle de la densité* [ED11; LLS19b; BW13] — en anglais *Density Functional Theory* (DFT) — est un ensemble de méthodes numériques très populaires en chimie quantique. Nous allons expliquer l'idée maitresse de cette théorie, sans rentrer dans ses détails techniques qui nécessite le formalisme mathématique de la mécanique quantique [Lew22b]. Nous notons par ailleurs que ce que nous avons esquissé jusqu'à présent peut être vu comme une version *classique* de cette théorie — et nous reviendrons plus en détails sur cette question lorsqu'il sera question de la température positive.

On considère un système de  $N$  particules *quantiques* qui interagissent au travers un potentiel d'interaction à deux corps  $w$  et qui sont soumises à un potentiel extérieur  $V$ . L'énergie du système est cette fois donnée par la forme quadratique

$$\begin{aligned}
Q_{N,w,V}(\Psi) &= \hbar^2 \int_{\mathbb{R}^{dN}} \sum_{i=1}^N |\nabla_{x_i} \Psi(x_1, \dots, x_N)|^2 \\
&+ \int_{\mathbb{R}^{dN}} \sum_{i=1}^N V(x_i) |\Psi|^2(x_1, \dots, x_N) + \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) |\Psi|^2(x_1, \dots, x_N),
\end{aligned} \tag{4}$$

pour toute *fonction d'onde*  $\Psi$ . On rappelle que  $\Psi$  est par définition une fonction de carré intégrable  $\Psi \in L^2(\mathbb{R}^{dN})$  normalisée telle que  $\int_{\mathbb{R}^{dN}} |\Psi|^2 = 1$ , et qui vérifie de surcroît une contrainte de symétrie de telle façon à ce que la fonction  $|\Psi|^2$  soit invariante par changement des indices des particules. La condition de normalisation assure que cette fonction  $|\Psi|^2$  est une densité de probabilité sur l'espace des configurations  $(\mathbb{R}^d)^N$ . Cette quantité est l'équivalent de notre  $\mathbb{P}$  précédent. Le premier terme dans (4) correspond à l'énergie cinétique. On note que ce terme ne peut pas s'exprimer directement en fonction  $\mathbb{P}$  — sauf dans le cas particulier où la fonction  $\Psi$  est de signe constant. Le second (resp. troisième) correspond à l'énergie potentielle (resp. d'interaction). La constante  $\hbar$  est à la *constante de Planck*. Le **Problème E** de la détermination de l'énergie fondamentale que nous avons introduit précédemment se réécrit maintenant dans sa version quantique [Lew22b, Chap. 1] comme

$$E_{N,w}(V) := \inf_{\substack{\Psi \in L^2(\mathbb{R}^{dN}) \\ \int_{\mathbb{R}^{dN}} |\Psi|^2 = 1}} Q_{N,w,V}(\Psi). \tag{5}$$

Ce problème variationnel est équivalent à déterminer la plus petite valeur propre  $E = E_{N,w}(V)$  (et son vecteur propre  $\Psi$  correspondant) de l'opérateur de Schrödinger associé,

$$\left( -\hbar^2 \sum_{i=1}^N \Delta_{x_i} + \sum_{i=1}^N V(x_i) + \sum_{1 \leq i < j \leq N} w(x_i, x_j) \right) \Psi = E\Psi. \tag{6}$$

À l'image du cas classique, la résolution analytique de ce problème est impossible, à l'exception de quelques cas d'école comme l'atome d'hydrogène [Lew22b, Sec. 1.3]. Quant à sa résolution numérique, l'histoire se répète en pire, car il est nécessaire de discrétiser un espace  $L^2$ . On trouve donc que la complexité algorithmique est typiquement exponentielle en le nombre de particules et en la taille de la discrétisation. L'idée principale de la DFT, que nous avons déjà rencontrée précédemment, consiste à séparer la minimisation sur l'ensemble des fonctions d'onde par une minimisation en deux étapes. La première sur l'ensemble des densités électroniques  $\rho$ , et la deuxième sur l'ensemble des fonctions d'ondes telles que la distribution des particules  $|\Psi|^2$  vérifie  $\rho_{|\Psi|^2} = \rho$ . On trouve alors

$$E_{N,w}(V) = \inf_{\rho} \left\{ F_{\hbar}(\rho) - \int_{\mathbb{R}^d} V(r)\rho(r)dr \right\} \tag{7}$$



où la *fonctionnelle de Levy–Lieb*  $F_{\hbar}$  [Lev79; Lie83; HT22] est définie par

$$F_{\hbar}(\rho) := \inf_{\rho_{|\Psi|^2} = \rho} \left\{ \hbar^2 \int_{\mathbb{R}^{dN}} \sum_{i=1}^N |\nabla_{x_i} \Psi|^2 + \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) |\Psi|^2(r_1, \dots, r_N) \right\}. \quad (8)$$

Nous avons ainsi exprimé l'énergie fondamentale  $E_{N,w}(V)$  non plus comme un problème variationnel sur l'ensemble des fonctions d'ondes  $\Psi \in L^2(\mathbb{R}^{dN})$ , mais bien comme un problème variationnel sur l'ensemble des densités électroniques  $\rho \in L^1(\mathbb{R}^d)$ , *a priori* bien plus petit, c'est-à-dire comme une *fonctionnelle de la densité*. Cette formulation est précisément le point de départ de la *Théorie de la fonctionnelle de la densité*, dont nous comprenons maintenant le nom.

*Remarque.* Si nous abusons volontairement du manque de rigueur mathématique, une petite remarque s'impose ici. Dans le cas classique, l'ensemble des densités électroniques  $\rho$  correspondait tout simplement à l'ensemble des mesures positives telles que  $\int_{\mathbb{R}^d} \rho = N$ . Dans le cas quantique, il faut être un peu plus prudent. En effet, nous travaillons avec des fonctions d'ondes  $\Psi$  qui vivent typiquement dans des espaces de Sobolev — pour s'assurer que le premier terme dans (4) soit fini. En particulier, l'ensemble des densités  $\rho$  dites *représentables*, c'est-à-dire telle qu'il existe une fonction d'onde  $\Psi$  pour laquelle  $\rho = \rho_{|\Psi|^2}$ , est l'ensemble des densités vérifiant certaines conditions de régularité et d'intégrabilité. Il est connu — voir [Lie83, Thm. 1.1] — que cet ensemble est donné par  $\{\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+) : \sqrt{\rho} \in H^1(\mathbb{R}^d)\}$ , où  $H^1(\mathbb{R}^d)$  est l'espace de Sobolev des fonctions  $u \in L^2(\mathbb{R}^d)$  dont la dérivée faible est aussi de carrée intégrable, c'est-à-dire  $u' \in L^2(\mathbb{R}^d)$ .

On voit que  $F_{\hbar}(\rho)$  correspond à l'énergie cinétique *plus* l'énergie d'interaction minimale qui peut être atteinte par un état quantique  $\Psi$  sous contrainte que  $\rho_{|\Psi|^2} = \rho$ . En particulier, à la *limite semi-classique*  $\hbar \rightarrow 0$ , on s'attend à ce que la fonctionnelle  $F_{\hbar}$  coïncide formellement avec la fonctionnelle  $F_{N,w}$  définie précédemment. Cela n'a du reste rien d'étonnant car  $F_{\hbar}$  correspond exactement à la contrepartie quantique de  $F_{N,w}$ . On trouvera une preuve rigoureuse de cette limite dans [Lew18; CFK13; BDP17; CFK18].

Nous voyons donc que la fonctionnelle  $F_{N,w}$ , que nous avons introduite dans un cadre purement classique, apparaît aussi dans le cadre d'une théorie pour le calcul numérique en chimie quantique. En pratique, la fonctionnelle  $F_{N,w}$  est utilisée comme une *fonctionnelle d'échange-corrélation* à l'intérieur du formalisme de *Kohn–Sham* (KS; [KS65]). Ce formalisme très populaire consiste à construire un système quantique sans interactions équivalent — en un certain sens — au système d'origine dont on cherche à calculer l'énergie fondamentale. En d'autres termes, ce formalisme correspond à considérer la limite  $\hbar \rightarrow \infty$  dans la fonctionnelle de Levy–Lieb  $F_{\hbar}(\rho)$ . Au contraire, nous avons mentionné que  $F_{N,w}$  apparaissait à la limite semi-classique  $\hbar \rightarrow 0$ . Ce formalisme porte le nom de *Strictly–Correlated Electrons* (SCE; [SPL99]) dans le cadre de la DFT, et correspond, à l'inverse de Kohn–Sham, à construire un système quantique sans énergie cinétique équivalent au système d'origine. Dans ce système, les électrons sont fortement corrélés et peuvent être considérés comme des particules classiques. Dans une série de travaux [MGG12; PLK13;



[Mal+14; MMGG14; GVGG18; Gro+20; Mar+22], la fonctionnelle  $F_{N,w}$  est utilisée comme un terme de correction dans *Kohn–Sham* efficace dans le cas de systèmes quantiques très corrélés, par exemple les *isolants de Mott* ou les systèmes cristallisés.

### §1.3. Le cas de la température positive

Après cette courte excursion dans le monde quantique, revenons à notre problème classique. Jusqu'à présent, nos systèmes étaient définis sans température. Bien évidemment, le cas de la température nulle est un idéal-type qui n'a d'existence que théorique. Il n'est pas atteignable thermodynamiquement, sinon asymptotiquement.

On peut réécrire le **Problème E** de la détermination de l'état d'équilibre d'un système à température positive  $T > 0$  en rajoutant, selon le formalisme mathématique de la mécanique statistique, l'entropie de Boltzmann

$$E_{N,w,T}(V) := \inf_{\mathbb{P}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) - T\mathcal{S}(\mathbb{P}) \right\}. \quad (E_T)$$

Ici, nous avons noté  $\mathcal{S}(\mathbb{P})$  l'entropie de la mesure  $\mathbb{P}$ , définie pour toute mesure absolument continue par rapport à la mesure de Lebesgue comme

$$\mathcal{S}(\mathbb{P}) = - \int_{\mathbb{R}^{dN}} \ln(\mathbb{P}) d\mathbb{P}. \quad (9)$$

Le paramètre  $T$  correspond à la température<sup>7</sup>. On notera aussi suivant l'usage habituel  $\beta$  la température inverse, c'est-à-dire  $\beta = T^{-1}$ . Nous pouvons maintenant réitérer dans les moindres détails ce que nous avons présenté au début de cette introduction dans le cadre du **Problème E**. Le lecteur pourra alors se convaincre qu'on obtient que l'énergie minimale  $E_{N,w,T}$  à température finie, appelée *énergie libre*, se réécrit comme

$$E_{N,w,T}(V) = \sup_{\rho \in \mathcal{Y}'} \left\{ F_{N,w,T}(\rho) - \int_{\mathbb{R}^d} V(x) \rho(x) \right\} \quad (D_T)$$

où, comme précédemment,  $F_{N,w,T}$  est la transformée de Legendre de l'énergie  $E_{N,w,T}(V)$  vue comme une fonctionnelle du potentiel extérieur  $V$ . On trouve ensuite que  $F_{N,w,T}$  se réécrit comme

$$F_{N,w,T}(\rho) = \inf_{\rho_{\mathbb{P}} = \rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) - T\mathcal{S}(\mathbb{P}) \right\}. \quad (F_T)$$

On remarque alors que le **Problème F<sub>T</sub>** est similaire au **Problème F**, si ce n'est pour l'ajout de l'entropie pondérée par la température  $T$ . En l'occurrence, dans le langage du transport optimal contemporain, il s'agit d'un problème

<sup>7</sup>On notera l'absence comme préfacteur de la constante de Boltzmann  $k_B$ . Cette dernière est plus une convention historique qu'une véritable nécessité physique.

de transport *entropique* [Lé12; Ben+15]. Si dans notre cas cette entropie joue un rôle physique, dans le cadre habituel du transport optimal, elle est plutôt utilisée pour régulariser le problème du transport. En particulier, le paramètre  $T$  joue alors le rôle d'un paramètre de régularisation.

Il suit d'un exercice classique de calcul des variations que le minimiseur du Problème  $F_T$ , qui est unique par stricte convexité de l'entropie, est une *mesure de Gibbs–Boltzmann*  $\mathbb{P}_\beta$  dont la densité est donnée par

$$\mathbb{P}_\beta(x_1, \dots, x_N) = \frac{1}{Z_\beta} \exp \left( -\beta \sum_{1 \leq i < j \leq N} w(x_i, x_j) - \beta \sum_{i=1}^N V_\beta(x_i) \right) \quad (10)$$

Ici, la constante  $Z_\beta$  assure que  $\mathbb{P}_\beta$  soit une mesure de probabilité sur  $(\mathbb{R}^d)^N$ , et  $V_\beta$  est un potentiel extérieur *a priori* inconnu qui assure que la contrainte de marge  $\rho_{\mathbb{P}_\beta} = \rho$  soit vérifiée. Dans le vocabulaire de la mécanique statistique, la mesure  $\mathbb{P}_\beta$  est appelée l'*ensemble canonique*. Il suit que la résolution du Problème  $F_T$  se résume à trouver le potentiel  $V_\beta$  telle que l'ensemble canonique à température  $\beta$  vérifie la contrainte de marge. Ce potentiel vérifie une équation non-linéaire, que l'on introduira plus tard en (75).

Un tel problème inverse, qui consiste à trouver le potentiel  $V$  qui donne naissance à la densité  $\rho$ , correspond au problème de la *théorie de la fonctionnelle de la densité* dans sa version *classique*. On pourra consulter la référence classique [Eva79], ou encore [Eva92; Sin91] ou [Wu09] pour un tour d'horizon plus récent du sujet. La DFT classique est une méthode populaire pour l'étude des fluides inhomogènes classiques, c'est-à-dire pour lesquels la densité  $\rho$  n'est pas constante en espace — comme par exemple à l'interface d'une séparation de phase [Gel+99]. Comme pour la DFT quantique, l'idée de la DFT classique est de construire des fonctionnelles en la densité  $\rho$  approchées [ESS76; EP79; SE77]. Pour ce qui est de sa justification théorique, la référence classique est un travail de Chayes, Chayes et Lieb dans [CCL84] — voir aussi [JLM23] pour un travail plus récent. Il est amusant de noter que l'article de Chayes *et al.* [CCL84], auquel nous référerons souvent, contient essentiellement ce qu'on appellerait volontiers aujourd'hui la *dualité de Kantorovich* pour le problème de transport entropique — du moins pour des coûts associés à des potentiels d'interaction répulsifs — même si la terminologie employée n'est pas la même.

Peut-être certains de nos lecteurs sont enfin rassurés de nous entendre mentionner le nom de Kantorovich et de la dualité qui porte son nom. Il est vrai que jusqu'à présent, nous n'avions pas accolé ce nom à chaque fois que nous parlions de dualité. Après tout, nous comprenons bien maintenant que lorsque nous avons défini la fonctionnelle  $F_{N,w}$  comme la transformée de Legendre de l'énergie fondamentale  $E_{N,w}$ , c'est-à-dire

$$F_{N,w}(\rho) = \sup_{V \in \mathcal{V}} \left\{ E_{N,w}(V) - \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}, \quad (11)$$

nous ne faisons du reste qu'écrire la dualité de Kantorovich pour le problème de transport optimal  $F_{N,w}(\rho)$  — *idem* pour l'énergie libre à température positive. Cependant, il aurait été étrange de faire ainsi, suivant la logique qui était la

nôtre. En effet, nous sommes partis en quelque sorte du dual — la fonctionnelle de l'énergie fondamentale  $E_{N,w}$  — pour arriver à ce qu'on nommerait volontiers, suivant la langage du transport optimal, le *primal* — c'est-à-dire la fonctionnelle  $F_{N,w}$ . En somme, dans le cadre de la physique, il s'agirait d'invertir la nomenclature — le dual étant le "primal" et *vice-versa*. D'autant plus que le transport optimal  $F_{N,w}$  n'arrive finalement que "par accident", étant acquis que nous étions principalement intéressé par l'énergie fondamentale  $E_{N,w}$ .

### Résumé

Avant de clore cette première partie, revenons sur nos pas, au prix de la répétition. Nous sommes partis du **Problème E** de la détermination de l'énergie fondamentale. Ce problème étant difficile, nous l'avions exprimé autrement en utilisant sa transformée de Legendre. Cette transformée, que nous avons notée  $F_{N,w}$ , est une fonctionnelle *a priori* tout aussi compliquée que  $E_{N,w}$ . Nous avons abouti au fait qu'elle correspondait exactement à un problème de transport optimal multimarge. Cependant, nous avons fait remarquer que  $F_{N,w}$  était une fonctionnelle qui ne dépendait pas du potentiel extérieur  $V$ , et qu'il n'était pas exclu d'en trouver des approximations dans certains régimes. Nous avons enfin expliqué, brièvement, qu'un tel travail était au coeur de la théorie de la fonctionnelle de la densité, une méthode très importante pour le calcul numérique en chimie quantique.

## §2. Le transport multimarge et sa résolution numérique

Nous avons précédemment expliqué comment le transport optimal, dans sa formulation multimarge, apparaissait naturellement dans le cadre de la physique statistique ou encore en chimie quantique comme dual du problème de la détermination de l'état fondamental d'un système particulaire. Dans cette partie, nous rappelons brièvement le formalisme multimarge en insistant sur l'interprétation physique de la dualité de Kantorovich.

### §2.1. Formulation générale du transport multimarge

Le transport multimarge [Pas15] correspond à l'extension naturelle à plusieurs marges du transport optimal classique à deux marges. On pourra se référer aux ouvrages classiques [Vil09; San15] pour une introduction à la théorie du transport. En toute généralité, le problème multimarge peut être défini de la façon suivante. Étant données  $N$  mesures de probabilité  $\mu_1, \dots, \mu_N$  sur  $\mathbb{R}^d$  et un *coût de transport*  $c : (\mathbb{R}^d)^N \rightarrow \mathbb{R}$ , on définit le *problème de Kantorovich* comme

$$\inf_{\pi \in \Pi(\mu_1, \dots, \mu_N)} \int_{\mathbb{R}^{dN}} c(x_1, \dots, x_N) d\pi(x_1, \dots, x_N) \quad (\text{MOT})$$

où  $\Pi(\mu_1, \dots, \mu_N)$  désigne l'ensemble des *plans de transport*, c'est-à-dire des mesures sur le produit cartésien  $\mathbb{R}^d \times \dots \times \mathbb{R}^d$  telles que la  $i$ -ième marge de  $\pi$  — selon à la  $i$ -ème copie de  $\mathbb{R}^d$  du produit — est donnée par  $\mu_i$ .

On note que l'existence d'un minimiseur pour le **Problème MOT** suit, comme pour le transport classique à deux marges, d'un argument classique de compacité sous des hypothèses assez faibles, par exemple lorsque le coût de transport  $c$  est une fonction semicontinue inférieurement et minorée uniformément [Vil09, Thm. 5.10]. En particulier, dans le cas des coûts de transport issus de la physique statistique, comme dans le cas du **Problème F** présenté à la partie précédente, c'est-à-dire

$$c(r_1, \dots, r_N) = \sum_{1 \leq i < j \leq N} w(x_i, x_j), \quad (12)$$

nous avons immédiatement l'existence d'un plan de transport sous l'hypothèse que  $w$  est semicontinue inférieurement et minorée uniformément. Ceci est par exemple le cas pour le potentiel de Coulomb en dimension trois d'espace, c'est-à-dire  $w(x, x') = |x - x'|^{-1}$ . Dans le cas où le coût n'est pas borné inférieurement, par exemple lorsque  $w$  est le potentiel de Coulomb en dimension deux d'espace, qui est donnée par l'interaction logarithmique  $w(x, x') = -\ln|x - x'|$ , l'existence d'un minimiseur s'obtient facilement sous l'hypothèse supplémentaire qu'il existe une fonction mesurable  $b$  telle que  $w(x, x') \leq b(x) + b(x')$  et telle que  $\int_{\mathbb{R}^d} |b| \rho < \infty$ , voir [Vil09, Thm. 5.10]. En particulier, pour le potentiel de Coulomb en dimension  $d = 2$ , il suffit que  $\int_{\mathbb{R}^2} |\ln(x)| \rho < \infty$ .

Comme pour le transport optimal à deux marges, nous pouvons aussi définir le *problème de Monge* — qu'on rappellera être la formulation historique du transport optimal. Dans le cas multimarge, le problème s'écrit

$$\inf_{T_i^\# \mu_1 = \mu_i} \int_{\mathbb{R}^{dN}} c(x, T_2(x), \dots, T_N(x)) d\mu_1(x) \quad (\text{Monge})$$

où l'*infimum* est pris sur l'ensemble des applications mesurables  $T_i$  telles que  $T_i^\# \mu_1 = \mu_i$  pour tout  $i = 2, \dots, N$  — où selon la notation habituelle  $f^\# \mu$  est la *mesure image* définie pour tout ensemble mesurable  $E$  et toute fonction mesurable  $f$  comme  $f^\# \mu(E) := \mu(f^{-1}(E))$ . Ce problème est très difficile à manier — *a priori* bien encore plus que son homologue à deux marges. Par ailleurs, beaucoup de résultats théoriques sur le problème de Monge dans le cas du transport optimal classique à deux marges sont obtenus grâce au lien très important qui l'unit à l'*équation de Monge–Ampère* [Fig18; Fig17; DPF14]. Cette analogie ne tient plus *a priori* dans le cas multimarge — ou du moins, si on peut formuler une équation de ce type, celle-ci semble tout simplement être bien trop compliquée pour fournir la moindre prise intéressante.

Un problème ouvert important consiste à déterminer s'il existe toujours un plan de transport  $\mathbb{P}$  pour le **Problème F** qui soit une solution du **Problème Monge**, c'est-à-dire telle qu'il existe des applications  $T_2, \dots, T_N$  qui préserve la densité  $\rho$  et telle que le plan de transport  $\mathbb{P}$  soit supportée sur le graphe des applications  $T_i$ , c'est-à-dire

$$\mathbb{P}(x_1, \dots, x_N) = \frac{\rho(x_1)}{N} \otimes \delta(x_2 - T_2(x_1)) \otimes \dots \otimes \delta(x_N - T_N(x_1)). \quad (13)$$

Comme toutes les marges sont égales pour le Problème F, on peut montrer que le problème est équivalent à trouver une unique application  $T$  préservant la densité  $\rho$  telle que  $T_i = T^{(i)}$ , où  $T^{(i)}$  dénote la  $i$ -ème composée de  $T$  avec elle-même, et telle que  $T^{(N)}$  soit l'identité [CDM15]. Dans le cas où  $N = 2$ , le réponse est positive pour le coût de Coulomb [BDPGG12; CFK13], voir aussi [DMGN15, Thm. 3.6]. Ce résultat suit d'un théorème tout à fait général pour le transport à deux marges, sous l'hypothèse que le coût de transport vérifie la *twist condition* [San15, Def. 1.16], ce qui est le cas pour le coût de Coulomb ou plus généralement si  $w(x, x') = \tilde{w}(|x - x'|)$  où  $\tilde{w}$  est une fonction strictement convexe. Un résultat similaire a été démontré dans le cas du transport multimarge par Kim et Pass [KP14]. Néanmoins, les hypothèses de ce théorème sont difficiles à vérifier en pratique, et le problème demeure ouvert pour un nombre arbitraires de particules  $N$  en dimension  $d \geq 2$ . En dimension une d'espace, le problème a été résolu par Colombo, De Pascale et Di Marino [CDPDM15] dans le cas des potentiels d'interaction convexes, qui ont donné de surcroît une construction explicite de l'application de transport  $T$ . Si l'existence d'un minimiseur demeure encore en grande partie inconnue, il est cependant connu que l'infimum du Problème Monge est égal au minimum du Problème MOT. Cela suit d'une extension au cadre multimarge du résultat obtenu par Pratelli [Pra07] dans le cas du transport optimal à deux marges, comme expliqué dans [CDM15, Rem. 1.3]. Le fait que les applications de transports puissent être prises *cycliques*, c'est-à-dire générées par la composition d'une unique application  $T$  avec elle-même comme expliqué ci-dessus, a été démontré par Colombo et Di Marino [CDM15].

Nous ne rentrerons pas plus en détails dans la théorie abstraite du transport optimal multimarge. Pour quelques références classiques, on pourra consulter [Pas15; DMGN15].

## §2.2. Interprétation de la dualité de Kantorovich

Comme tout problème de transport optimal, le transport multimarge admet une formulation duale suivant la fameuse *dualité de Kantorovich*. Pour un problème de transport multimarge générique telle que défini plus haut au Problème MOT, le dual de Kantorovich s'écrit

$$\sup_{V_1, \dots, V_N} \left\{ E(V_1, \dots, V_N) + \sum_{i=1}^N \int_{\mathbb{R}^d} V_i(x) \mu_i(dx) \right\} \quad (\text{MOT}_D)$$

où le *supremum* est pris sur l'ensemble des fonctions continues  $V_i : \mathbb{R}^d \rightarrow \mathbb{R}$  pour  $i = 1, \dots, N$ , et où

$$E(V_1, \dots, V_N) := \inf_{x_1, \dots, x_N} \left\{ c(x_1, \dots, x_N) - \sum_{i=1}^N V_i(x_i) \right\}. \quad (14)$$

Nous utilisons ici une formulation qui n'est pas *a priori* la plus usuelle [PC19, Rem. 2.23]. En effet, on demande généralement à ce que les fonctions  $V_i$  vérifient la contrainte d'admissibilité

$$V_1(x_1) + \dots + V_N(x_N) \leq c(x_1, \dots, x_N) \quad (15)$$

où l'inégalité est ou bien demandée pour toutes configurations, ou bien seulement  $\mu_1 \otimes \dots \otimes \mu_N$  presque partout — sous de bonnes hypothèses, ces deux contraintes sont en fait équivalentes. Notre formulation du **Problème MOT<sub>D</sub>** est équivalente à demander à ce que cette contrainte d'admissibilité habituelle ne soit vérifiée qu'à constante près. Cette constante est donnée par la quantité (14). En particulier, la contrainte (15) est équivalente à demander  $E(V_1, \dots, V_N) = 0$ . L'intérêt de cette formulation est de définir les potentiels de Kantorovich à constante près, ce qui est naturel du point de vue de la physique. Du point de vue de la résolution numérique du problème dual, elle permet de se débarrasser de la contrainte (15) qui n'est pas des plus aisées à manipuler.

Dans le cadre de la fonctionnelle  $F_{N,w}$ , toutes les marges sont égales  $\mu_1 = \dots = \mu_N$  et données par  $\rho/N$ . On rappelle que, par convention, on a  $\int_{\mathbb{R}^d} \rho = N$ . Quitte à considérer la moyenne arithmétique des  $V_i$  dans le **Problème MOT<sub>D</sub>**, on peut toujours réécrire le dual de Kantorovich avec pour variable d'optimisation une seule et unique fonction  $V$ . On (re)trouve alors

$$F_{N,w}(\rho) = \sup_{V \in \mathcal{V}} \left\{ E_N(V) - \int_{\mathbb{R}^d} V(x) \rho(x) \right\}, \quad (16)$$

c'est-à-dire la transformée de Legendre de l'énergie fondamentale  $E_{N,w}(V)$ . L'égalité des deux quantités — *dualité forte* — et l'existence d'une fonction  $V$  qui maximise le *supremum* — ce qu'on appellera un *potentiel de Kantorovich* selon la dénomination usuelle — suit des résultats de [CDMS19; BCDP18]. Nous notons qu'ici, nous avons changé le signe du potentiel par rapport au **Problème MOT<sub>D</sub>**. La raison à ce changement de signe, anodin du point de vue mathématique, suit d'une considération physique, comme nous allons maintenant le voir.

*Quel est l'interprétation du problème dual dans le cadre de la physique statistique ?* Pour cela, nous allons faire appel à la théorie de dualité dans le cadre général du transport optimal. En effet, il est connu qu'à l'optimalité [San15], si on considère un plan de transport optimal  $\mathbb{P}$  et un potentiel de Kantorovich  $V$ , on doit avoir

$$\sum_{1 \leq i < j \leq N} w(x_i, x_j) + \sum_{i=1}^N V(x_i) = E_N(V) \quad \text{sur le support de } \mathbb{P}, \quad (17)$$

et où l'égalité est une inégalité sur tout l'espace. Cette égalité dit exactement que les configurations  $(r_1, \dots, r_N) \in \text{supp}(\mathbb{P})$  minimisent l'énergie totale du système. D'un point de vue physique, il est intéressant de dériver cette équation pour faire apparaître la force. En effet, sous de bonnes hypothèses de régularité sur  $V$  et  $w$ , on obtient en dérivant par rapport à la première particule  $x_1$  l'égalité précédente que

$$-\nabla_{x_1} V(x_1) = \sum_{i=2}^N \nabla_{x_1} w(x_1, x_i). \quad (18)$$

Cette égalité est une équation d'équilibre de mécanique classique. À l'optimalité, la force exercée sur la première particule  $x_1$  due au potentiel extérieur  $V$  doit annuler la force d'interaction exercée par les autres particules  $x_2, \dots, x_N$  sur

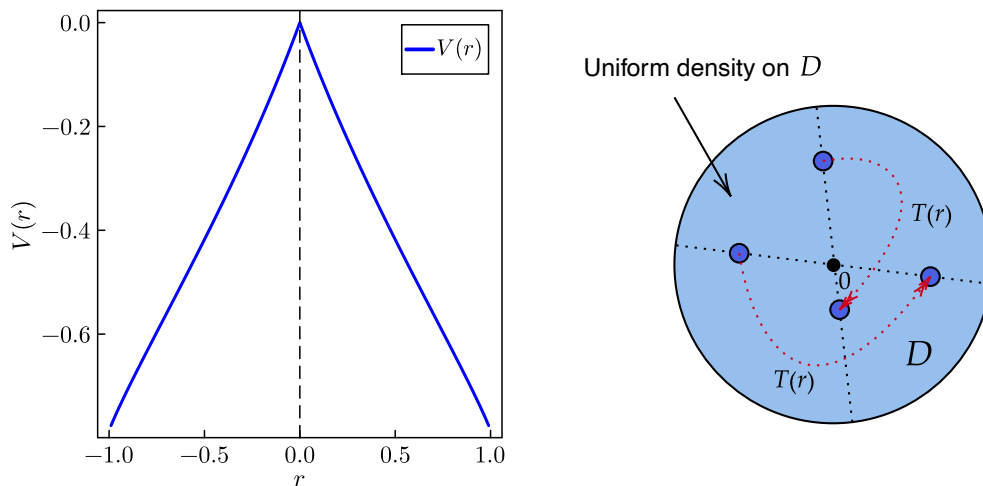


FIGURE 2 : On considère deux particules qui interagissent par le biais du potentiel logarithmique  $-\ln|x - x'|$  en dimension  $d = 2$ . On fixe pour densité cible la mesure uniforme sur le disque unité  $D$ . Le plan de transport est unique et supporté sur le graphe de l'application  $T$  telle que définie à (19). Le potentiel de Kantorovich  $V$  (73) est déterminé en intégrant l'équation d'équilibre (18).

la particule  $x_1$ . On obtient donc une caractérisation physique du potentiel de Kantorovich comme le potentiel extérieur qui force les particules à vivre sur le support d'un plan de transport optimal à l'équilibre.

*Exemple.* Considérons l'exemple d'un système à deux particules  $N = 2$  en dimension  $d = 2$  qui interagissent entre elles par le potentiel de Coulomb  $-\ln|r - r'|$ . On fixe pour densité cible  $\rho$  la mesure uniforme sur le disque unité  $D$  — normalisée telle que  $\int_D \rho = 2$ . Cette densité étant radiale, on peut utiliser les résultats obtenus par Pass dans [Pas13]. On sait alors que le plan transport optimal est unique et donné par

$$\mathbb{P}(x, x') = \frac{\rho(x)}{2} \otimes \delta(x' - T(x)) \quad \text{où } T(x) = -\frac{x}{|x|} \sqrt{1 - |x|^2}. \quad (19)$$

Le potentiel de Kantorovich  $V$  est alors déterminé implicitement par l'équation d'équilibre (18) — voir Figure 2. Si  $V$  peut-être approchée numériquement en intégrant cette équation d'équilibre, dans notre cas spécifique, on peut calculer explicitement  $V$ , qui est donné sur  $D$  par

$$V(x) = -\frac{1}{2} \sin^{-1}(x) - \frac{1}{2} \ln \left( 1 + \frac{|x|}{\sqrt{1 - |x|^2}} \right) - \frac{1}{4} \ln(1 - |x|^2). \quad (20)$$



### §2.3. Transport optimal entropique

Nous rappelons que le problème dual de la détermination de l'énergie libre  $E_{N,T}(V)$  à température positive  $T > 0$  s'exprime comme

$$F_{N,w,T}(\rho) = \inf_{\rho_{\mathbb{P}}=\rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) - T\mathcal{S}(\mathbb{P}) \right\}, \quad (\text{F}_T)$$

où  $\mathcal{S}(\mathbb{P})$  désignait l'entropie de la mesure  $\mathbb{P}$  par rapport à la mesure de Lebesgue. Nous avons indiqué qu'il s'agissait d'un problème de transport optimal entropique [Lé12; Ben+15]. Nous rappelons que le minimiseur de Problème  $\text{F}_T$ , qui est unique par stricte convexité de l'entropie, est donnée par la mesure de Gibbs  $\mathbb{P}_\beta$  — ou l'*ensemble canonique* selon la terminologie de la physique statistique — dont la densité est

$$\mathbb{P}_\beta(x_1, \dots, x_N) = \frac{1}{Z_\beta} \exp \left( -\beta \sum_{1 \leq i < j \leq N} w(x_i, x_j) - \beta \sum_{i=1}^N V_\beta(x_i) \right) \quad (21)$$

et où  $V_\beta$  est l'unique potentiel tel que  $\mathbb{P}_\beta$  vérifie la contrainte de marge  $\rho_{\mathbb{P}_\beta} = \rho$  du transport. Nous notons par ailleurs que cette contrainte force l'égalité  $V_\beta = +\infty$  en dehors du support de  $\rho$ . Ceci se comprend facilement, car en l'absence d'une telle barrière de potentiel, les particules pourraient sortir en dehors de  $\Omega$ . Par conséquent l'ensemble canonique  $\mathbb{P}_\beta$  chargerait *a priori* un ouvert  $U$  telle que  $U \cap \Omega^c \neq \emptyset$ . La densité  $\rho_{\mathbb{P}_\beta}$  serait alors supportée en dehors de  $\Omega$ , ce qui serait en contradiction avec la contrainte de marge.

En utilisant que  $V_\beta$  force la contrainte de marge pour  $\mathbb{P}_\beta$ , il suit de la définition de  $\rho_{\mathbb{P}_\beta}$  que le potentiel  $V_\beta$  vérifie une équation de point-fixe donnée par

$$\begin{aligned} & V_\beta(x) + \frac{\beta^{-1}}{N} \log \rho(x) \\ &= \beta^{-1} \log \int_{\mathbb{R}^{d(N-1)}} \exp \left( -\beta c(x, x_2, \dots, x_N) - \beta \sum_{i=2}^N V_\beta(x_i) \right) dx_2 \dots dx_N \end{aligned} \quad (22)$$

pour tout  $x$  dans le support  $\Omega$  de  $\rho$  — et *a priori* telle que  $\rho(x) > 0$ . Cette équation de point-fixe est parfois appelée *équation de Schrödinger* dans la communauté du transport optimal<sup>8</sup>. Nous n'utiliserons pas cette dénomination pour ne pas créer de confusion avec l'*autre* équation de Schrödinger, celle de la physique quantique. De même, le potentiel  $V_\beta$  qui est solution de l'équation de point-fixe est parfois appelé *potentiel de Schrödinger*. Encore une fois, nous n'utiliserons pas cette appellation, lui préférant celle de potentiel de Kantorovich, comme pour le cas de la température nulle.

<sup>8</sup>Cette dénomination un peu malheureuse provient de [Sch32], où E. Schrödinger introduit le problème suivant : *étant données  $N$  particules qui évoluent suivant un mouvement brownien, qu'on observe être distribuées selon une mesure  $\mu_0$  au temps  $t = 0$  et selon une autre mesure  $\mu_1$  au temps  $t = 1$ , que peut-on dire sur le comportement du système entre ces deux temps ?* Ce problème se trouve être équivalent au transport optimal entropique [Lé12; Lé13].



En notant  $W_\beta := V_\beta + \beta^{-1} \log \rho$ , l'équation précédente se réécrit

$$W_\beta(x) = \beta^{-1} \log \int_{\mathbb{R}^{d(N-1)}} \exp \left( -\beta c(x, x_2, \dots, x_N) - \beta \sum_{i=2}^N W_\beta(x_i) \right) d\mu^{\otimes N-1}$$

où  $\mu = \rho/N$ . Si le potentiel  $V_\beta$  est le véritable potentiel physique, la fonction  $W_\beta$  est une réécriture purement mathématique quoique plus commode. Par exemple, comme nous venons de le rappeler, le potentiel  $V_\beta$  doit être infini en dehors du support de  $\rho$ . Cette contrainte est automatiquement vérifiée pour  $W_\beta$  grâce au terme  $-\beta^{-1} \log \rho$ . De plus, comme noté dans [Lie83, Rem. p. 5], la décomposition  $V_\beta = -\beta^{-1} \log \rho + W_\beta$  est aussi utile car elle permet de retrancher le cas du gaz idéal — pour lequel  $W_\beta = 0$ . On notera aussi que dans le cadre du transport entropique, on considère généralement non pas l'entropie  $\mathcal{S}(\mathbb{P})$  par rapport à la mesure de Lebesgue sur  $\mathbb{R}^d$ , telle que nous l'avons fait, mais plutôt l'*entropie relative* par rapport à une mesure de référence  $\nu$ , qu'on considère généralement être le produit tensoriel des marges  $\nu = \mu_1 \otimes \dots \otimes \mu_N$ . Ce choix n'a *a priori* pas d'importance si les mesures  $\mu_i$  sont absolument continues, car à l'optimalité le plan de transport  $\mathbb{P}_\beta$  sera nécessairement absolument continue par rapport à la mesure produit  $\nu$ . La fonction  $W_\beta$  correspond en fait à la solution de l'équation de point-fixe qu'on aurait obtenue en considérant l'entropie relative par rapport à la mesure  $(\rho/N)^{\otimes N}$  plutôt que l'entropie par rapport à la mesure de Lebesgue.

### §3. Méthode de la charge duale (Chapitre 1)

Nous décrivons dans cette section les résultats obtenus au Chapitre 1. Le transport optimal multimarges est un problème dont la résolution numérique n'est pas aisée, particulièrement pour un nombre trop important de marges — c'est-à-dire, dans notre cadre physique, pour un nombre  $N$  trop important de particules. La complexité du problème numérique est typiquement exponentielle en le nombre de marges  $N$  et la taille de la discrétisation. Il est par ailleurs connu que le problème du transport optimal multimarges pour des coûts répulsifs est un problème NP-difficile [ABA21]. Pour un compte-rendu des méthodes numériques pour résoudre le Problème F, on peut se référer à [Vuc+23].

Du point de vue de la théorie de l'optimisation, le problème à température nulle est un *Programme Linéaire*. Il s'agit en effet de minimiser une fonction linéaire sous contrainte convexe. S'il existe toute une littérature d'algorithmes pour résoudre ce genre de problème, il demeure que la complexité numérique est très vite prohibitive, ne serait-ce que pour une dizaine de particules. Il s'agit donc d'être plus fin. On peut mentionner une série de travaux récents [FSV22; Vö21; FV18; FP23; EN23], où il est utilisé le fait que le plan de transport optimal devrait être génériquement parsimonieux — du moins dans un contexte discret. Une autre stratégie consiste à utiliser la régularisation entropique et à faire appel à l'algorithme du Sinkhorn [BCN16; Ben+15]. Cependant, cette méthode passe difficilement au cas  $N \gg 1$ , car le coût en mémoire nécessaire pour les tenseurs devient rapidement prohibitif. Nous pouvons aussi mentionner les travaux [Alf+20; ACE22; EN23] où il est question de relâcher la contrainte

de marges dans le transport optimal et de la remplacer par une contrainte sur les moments du plan de transport. Plus précisément, la contrainte de marge  $\rho_{\mathbb{P}} = \rho$  est remplacée par les contraintes de moments

$$\int_{\mathbb{R}^{dN}} \sum_{i=1}^N \phi_j(x_i) d\mathbb{P}(x_1, \dots, x_N) = \int_{\mathbb{R}^d} \phi_j(x) \rho(x) \quad (23)$$

pour un ensemble fini de fonctions  $\phi_j$  ( $j = 1, \dots, M$ ). Il se trouve que notre méthode, que nous allons maintenant présenter, est assez similaire à cette contrainte de moments. Plus précisément, elle peut être vue comme le dual de la méthode des moments.

Dans ce qui suit, nous considérons exclusivement le cas du transport optimal multimarge pour le *coût de Coulomb*, c'est-à-dire où le potentiel d'interaction  $w$  est donné par  $w(x, x') = g_d(|x - x'|)$  où  $g_d$  est le potentiel de Coulomb en dimension  $d \geq 1$ . On rappelle que  $g_d$  est défini comme la solution fondamentale de l'opérateur Laplacien  $-\Delta$ , c'est-à-dire  $-\Delta g_d = \delta_0$  où  $\delta_0$  est la mesure de Dirac à l'origine. Il suit que  $g_d$  est donnée à constante multiplicative près par

$$g_d(x) = \begin{cases} -|x| & \text{pour } d = 1, \\ -\ln|x| & \text{pour } d = 2, \\ \frac{1}{|x|^{d-2}} & \text{pour } d \geq 3. \end{cases} \quad (24)$$

Notre idée est la suivante. Étant donnée une densité électronique  $\rho$ , nous avons expliqué à la partie précédente que le potentiel de Kantorovich associé au problème du transport optimal  $F_{N,w}$  devait être interprété comme le potentiel physique qui force les particules à se positionner sur un plan de transport optimal à l'équilibre. Le problème étant de nature électrostatique, on peut se demander s'il est possible de réécrire le potentiel  $V$  comme un potentiel électrostatique généré par une distribution de charge  $\rho^{(e)}$  sur  $\mathbb{R}^d$ . Mathématiquement, cela revient à écrire

$$V(x) = \int_{\mathbb{R}^d} g_d(x - x') \rho^{(e)}(dx'), \quad (25)$$

ce qu'on peut encore écrire  $V = g_d * \rho^{(e)}$  où  $*$  dénote la convolution habituelle des fonctions et des mesures sur  $\mathbb{R}^d$ . Dans la suite, nous appelons  $\rho^{(e)}$  la *charge duale*. Si tel est le cas, il serait intéressant de s'essayer à résoudre le dual numériquement en discrétisant  $\rho^{(e)}$  plutôt que le potentiel  $V$  lui-même.

*Quelles propriétés peut-on attendre de la charge duale  $\rho^{(e)}$  ?* La charge duale doit attirer les particules et les forcer à se positionner sur un plan de transport optimal. Les électrons étant chargés négativement, on s'attend à ce que la charge duale soit une mesure positive. De plus, étant donnée la densité cible  $\rho$  de support  $\Sigma$ , on peut s'attendre à ce que la charge  $\rho^{(e)}$  soit supportée dans  $\Sigma$ . Si cela n'était pas le cas, une particule pourrait être attirée en dehors du support de  $\rho$ , ce qui contredirait l'interprétation que nous avons donnée du potentiel de Kantorovich. Finalement, d'après l'équation d'équilibre des forces à l'optimalité, qu'on rappelle s'écrire

$$-\nabla_{x_1} V(x_1) = \sum_{i=2}^N \nabla_{x_1} w(x_1, x_i), \quad (26)$$

on voit que le champ de force  $-\nabla V$  créé par le potentiel de Kantorovich sur une particule située à un point  $x_1$  doit contrebalancer la force répulsive exercée sur cette même particule par les  $N - 1$  autres. Par conséquent, nous sommes amenés à intuiter que le masse totale de cette mesure positive doit vérifier  $\int_{\mathbb{R}^d} \rho^{(e)} \geq N - 1$ . En effet, si la masse était strictement inférieure à  $N - 1$ , le champ  $-\nabla V$  ne devrait pas pouvoir écranter la force répulsive générée par les particules, et par conséquent le système ne pourrait pas être stable.

L'idée d'employer comme variable la charge duale a déjà été employée numériquement, dans le cadre du transport optimal pour le coût de Coulomb, par Mendl et Lin [ML13]. On peut aussi mentionner, dans le cadre général de la DFT, les travaux [VLGG17], ou encore [Gö99] qui semble être la référence historique (voir les références de la partie suivante). Dans [ML13], les auteurs écrivent le potentiel de Kantorovich  $V$  comme un potentiel électrostatique générée par une charge extérieure — qu'ils appellent *pseudo-charge*, et qui est équivalente à notre charge duale  $\rho^{(e)}$ . Ils discrétisent ensuite la charge duale à température nulle dans le cas de petites molécules de taille  $N = 2, 3$ , en la décomposant sur des gaussiennes. Le dual de Kantorovich est alors optimisé en utilisant un algorithme *derivative-free* de type *quasi-Newton*. Cependant, cette stratégie ne passe pas à l'échelle pour un nombre de particules trop important, comme noté par les auteurs : « *the derivative-free methods are not suitable for optimizing with respect to a large number of degrees of freedom. More efficient numerical methods need to be developed in order to obtain the Kantorovich dual solution for more general systems* ». Notre première contribution consiste donc à considérer le problème à température positive, qui est plus arrangeant du point de vue de l'optimisation.

En effet, dans le cas de la température positive, on peut attaquer numériquement le problème par les méthodes habituelles de gradient. Rappelons que le dual à température positive s'écrit

$$\sup_V \left\{ -\beta^{-1} \log Z_{N,\beta}(V) - \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}. \quad (27)$$

où  $Z_{N,\beta}(V)$  est la fonction de partition associée à l'ensemble canonique  $\mathbb{P}_\beta(V)$  telle que définie en (21). Dans le cas de la température nulle, l'énergie libre  $-\beta^{-1} \log Z_{N,\beta}(V)$  doit être remplacée par l'énergie fondamentale  $E_N(V)$ . Comme indiqué ci-dessus, il est difficile d'accéder aussi bien analytiquement que numériquement à la dérivée fonctionnelle de  $E_N(V)$  par rapport au potentiel extérieur  $V$  — il n'est d'ailleurs pas évident de savoir dans quel sens il faudrait considérer cette dérivée. Au contraire l'énergie libre peut être facilement dérivée. En effet, un calcul donne

$$\frac{\delta}{\delta V(x)} \left( -\beta^{-1} \log Z_{N,\beta}(V) + \int_{\mathbb{R}^d} V(x) \rho(x) \right) = \rho_{\mathbb{P}_\beta(V)}(x) - \rho(x). \quad (28)$$

où on rappelle que la quantité  $\rho_{\mathbb{P}_\beta(V)}$  correspond à la densité électronique de l'ensemble canonique  $\mathbb{P}_\beta(V)$ . Le problème de maximisation étant concave, la condition d'optimalité au premier ordre est donc équivalente à l'égalité  $\rho_{\mathbb{P}_\beta(V)} = \rho$ . Ceci n'a rien de surprenant, et est bien connu dans le cadre du transport optimal entropique.

Nous allons maintenant discrétiser la charge duale  $\rho^{(e)}$ . L'idée la plus simple consiste à se donner une base finie de fonctions  $B = \{\mu_i \in L^1(\mathbb{R}^d, \mathbb{R}_+) : i = 1, \dots, M\}$  et d'approcher  $\rho^{(e)}$  par une combinaison linéaire des éléments de la base  $B$ , c'est-à-dire d'approcher  $\rho^{(e)}$  par  $\rho^{(e)}[\nu]$  où

$$\rho^{(e)}[\nu] := \sum_{i=1}^M \nu_i \mu_i, \quad \nu = (\nu_i)_{i=1}^M \in \mathbb{R}_+^M. \quad (29)$$

Ceci est équivalent à approcher le potentiel de Kantorovich  $V$  par le potentiel  $V[\nu]$  où

$$V_\nu = \sum_{i=1}^M \nu_i V_i \quad \text{où } V_i = g_d * \rho_i \quad \text{pour } i = 1, \dots, M. \quad (30)$$

On optimise alors sur  $\nu \in \mathbb{R}_+^M$  le problème

$$\sup_{\nu \in \mathbb{R}_+^M} \left\{ -\beta^{-1} \log Z_{N,\beta}(V_\nu) - \int_{\mathbb{R}^d} V_\nu(r) \rho(r) \right\}. \quad (31)$$

Si on note  $G(\nu)$  la fonction à maximiser, la dérivée par rapport à  $\nu_i$  de  $G$  est alors donnée par

$$\partial_{\nu_i} G(\nu) = \int_{\mathbb{R}^d} V_i \rho_{\mathbb{P}_\beta(V_\nu)} - \int_{\mathbb{R}^d} V_i \rho. \quad (32)$$

Le deuxième terme dans le membre de droite, qui reste fixé tout au long de l'optimisation, peut être facilement calculé en amont. Son coût numérique est donc virtuellement nul. Le premier terme, qui fait intervenir la densité  $\rho_{\mathbb{P}_\beta(V_\nu)}$  de l'ensemble canonique  $\mathbb{P}_\beta(V_\nu)$ , est plus compliqué à calculer, et tout le coût numérique de l'optimisation lui est associé. Si une première stratégie consisterait à calculer la densité  $\rho_{\mathbb{P}_\beta(V_\nu)}$  en discrétisant  $\mathbb{R}^d$ , cela ne serait pas de tout efficace. En utilisant plutôt que cette densité est par définition ( $N$  fois) la marge de la distribution de probabilité  $\mathbb{P}_\beta(V_\nu)$ , il suit qu'on peut écrire

$$\int_{\mathbb{R}^d} V_i \rho_{\mathbb{P}_\beta(V_\nu)} = \int_{\mathbb{R}^{dN}} \sum_{j=1}^N V_i(x_j) d\mathbb{P}_\beta(V_\nu)(x_1, \dots, x_N). \quad (33)$$

On se retrouve donc avec un problème classique d'estimation numérique de l'intégrale d'une fonction sous une mesure. Cette quantité peut donc être estimée à partir des *méthodes de Monte-Carlo*. Bien évidemment, cela requiert de pouvoir échantillonner la mesure  $\mathbb{P}_\beta(V_\nu)$ , ce qui n'est pas chose facile.

Nous rappelons que nous avons défini l'ensemble canonique par rapport à la mesure produit  $(\rho/N)^{\otimes N}$ . Par conséquent, lorsque  $\rho$  est à support compact, il est possible de tirer des échantillons selon la mesure uniforme sur le support. Cependant, il serait beaucoup plus efficace d'échantillonner directement l'ensemble canonique. Ceci est d'autant plus crucial lorsque la température est petite — ce qui est précisément le régime qui nous intéresse. En effet, lorsque la température est petite, l'ensemble canonique va génériquement se concentrer sur les minimiseurs de l'énergie à température nulle. Ces minimiseurs vivent typiquement sur une variété singulière de petite dimension. Par conséquent,

plus la température diminue, plus un échantillonnage uniforme sera mauvais. Il s'agit donc d'échantillonner directement l'ensemble canonique.

Il n'existe pas de méthode directe pour échantillonner *exactement* la mesure  $\mathbb{P}_\beta(V)$  pour un potentiel extérieur  $V$  quelconque. Dans quelques cas particuliers, on peut relier l'ensemble canonique à des modèles de matrices aléatoires et ainsi obtenir une procédure d'échantillonnage exacte. Tel est le cas, par exemple, de l'ensemble canonique en dimension  $d = 1, 2$  pour le potentiel d'interaction logarithmique  $-\ln|x - x'|$  et le potentiel harmonique  $V(x) = |x|^2$  — ce qu'on nomme l'ensemble de Ginibre [Gin04] quand  $\beta = 2$  plus généralement  $\beta$ -ensemble [DE02; GBV21] pour un  $\beta > 0$  quelconque. Par conséquent, il faut faire appel à des méthodes d'échantillonnages approchées, comme par exemple les méthodes de *Markov Chain Monte-Carlo* [RC10; RC04].

Au Chapitre 1, nous présentons quelques résultats numériques. Nous appliquons notre discrétisation en dimension  $d = 1$  pour le potentiel de Coulomb  $-|x - x'|$ , où l'ensemble des plans de transport optimaux est connu — par les résultats de [CDPDM15] et un petit résultat supplémentaire que nous présentons au Chapitre 1 — ainsi qu'en dimension  $d = 3$  jusqu'à un nombre de particules  $N = 30$  pour le cas d'une densité cible  $\rho$  uniforme sur une boule. Les résultats sont ensuite comparés à ceux de Räsänen *et al.* dans [RSGG11]. Nous notons que les résultats de ces auteurs ont été obtenus en utilisant une construction due à Seidl pour les densités cibles  $\rho$  radiales [SGGS07]. S'il n'est pas connu rigoureusement que ces états tests fournissent un minimiseur dans le cas de la densité  $\rho$  uniforme sur une boule, nos résultats montrent que ces derniers ont néanmoins une énergie très proche de la valeur minimale.

Il nous reste à justifier théoriquement l'utilisation de cette charge duale.

### Justification théorique

*Existe-t-il toujours une charge duale ?* C'est-à-dire, étant donnée une densité cible  $\rho$ , existe-t-il toujours une mesure  $\rho^{(e)}$  telle que le potentiel électrostatique  $V = g_d * \rho^{(e)}$  généré par  $\rho^{(e)}$  soit un potentiel de Kantorovich pour le problème de transport associé ? Nous apportons tout d'abord une réponse positive dans le cas de la température nulle. Tous les théorèmes qui suivent sont énoncés et démontrés pour  $d \geq 3$ . Cela nous permet de nous affranchir de quelques détails techniques pour les dimensions  $d = 1, 2$ , pour lesquelles le potentiel de Coulomb n'est pas borné inférieurement. Cependant, on s'attend à ce que des résultats similaires soient vrais pour  $d = 1, 2$ . Dans le cas de la température nulle, on obtient le théorème qui suit.

**Théorème** (Théorème 1 du Chapitre 1, page 83). *Étant donnée une densité cible  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N \geq 2$ , il existe un potentiel de Kantorovich de la forme  $V(x) = \rho^{(e)} * g_d(x)$  pour une certaine mesure  $\rho^{(e)}$  positive de masse  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N - 1$ . De plus, si le support  $\Sigma$  de la densité  $\rho$  est borné, on peut toujours supposer que la mesure  $\rho^{(e)}$  est supportée sur  $\Sigma$ .*

L'existence d'une charge duale  $\rho^{(e)}$  suit de considérations en théorie du potentiel [Lan72], moyennant quelques résultats de la théorie du transport

optimal. En effet, il est connu [BCDP18] qu'on peut toujours trouver un potentiel de Kantorovich  $V$  qui vérifie l'équation

$$V(x) = \inf_{x_2, \dots, x_N} \left\{ c(x, x_2, \dots, x_N) - \sum_{i=2}^N V(x_i) \right\} \quad (34)$$

où on rappelle que  $c$  est le coût de transport associé au potentiel de Coulomb  $g_d$  en dimension  $d \geq 3$ , c'est-à-dire

$$c(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^{d-2}}. \quad (35)$$

Lorsque les particules  $x_2, \dots, x_N$  sont fixées, la fonction  $x \rightarrow c(x, x_2, \dots, x_N)$  étant superharmonique, il suit que la fonction  $V$  est superharmonique — sous l'hypothèse de sa continuité, qui n'est pas tout à fait triviale à obtenir. On peut alors conclure, suivant un résultat important de théorie du potentiel, que  $V$  peut s'écrire (à constante près) comme un potentiel de Coulomb généré par une mesure positive — précisément notre charge duale  $\rho^{(e)}$ . Qu'on puisse supposer que cette mesure soit supportée sur le support  $\Sigma$  de la densité cible  $\rho$  dans le cas où  $\Sigma$  est bornée suit aussi un théorème connu en théorie du potentiel. Nous étendons ensuite ce théorème au cas de la température positive :

**Théorème** (Théorème 3 du Chapitre 1, page 85). *Étant donnée une densité cible  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N \geq 2$  et qui vérifie que le potentiel  $\rho * g_d$  est uniformément borné, i.e.  $\rho * g_d \in L^\infty(\mathbb{R}^d)$ , alors il existe une mesure positive  $\rho_\beta^{(e)}$  telle que  $V_\beta(x) = \rho_\beta^{(e)} * g_d(x)$  sur le support de  $\rho$  (et à constante près), où  $V_\beta$  est l'unique potentiel de Kantorovich du problème de transport à température finie associé à la densité  $\rho$ . De plus, si le support  $\Sigma$  de  $\rho$  est borné, on peut toujours supposer que la mesure  $\rho_\beta^{(e)}$  est supportée sur  $\Sigma$ .*

La démonstration suit essentiellement les mêmes lignes que pour le cas de la température nulle. Une petite subtilité consiste à étendre correctement  $V_\beta$  à l'entièreté de l'espace. On rappelle en effet que  $V_\beta = \infty$  en dehors du support de  $\rho$ . On démontre ensuite que  $V_\beta$  est superharmonique en utilisant l'équation de point-fixe vérifiée par  $V_\beta$ , voir (75).

La charge duale n'est pas unique, du moins dans le cas où le support de  $\rho$  est compact, voir Remarque 2 du Chapitre 1, à la page 84. Cependant, on peut obtenir l'unicité de cette dernière sur le support de  $\rho$ .

**Théorème** (Théorème 2 du Chapitre 1, page 84). *Soit une densité cible  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N \geq 2$ , et  $\rho^{(e)}, \rho^{(e)'} \in \mathcal{M}(\mathbb{R}^d)$  deux charges duales pour le transport optimal associé à la densité  $\rho$ . Alors, les deux mesures coïncident sur l'intérieur topologique du support de  $\rho$ . En particulier, si le support est donnée par  $\mathbb{R}^d$ , alors les deux mesures coïncident partout. Ce résultat d'unicité est valide à température nulle et positive.*

Nous avons l'idée de rajouter de la température à notre système et de faire tendre cette température vers zéro pour retrouver numériquement le problème à température nulle. Du point de vue du coût total du transport  $F_{N,w}(\rho)$  il est

bien connu que  $F_{N,w,T}(\rho) \rightarrow F_{N,w}(\rho)$  [Car+17], ce qui montre la consistance de cette approche. *Qu'en est-il de la convergence de la charge duale ?* Nous démontrons que cette charge  $\rho_\beta^{(e)}$  à température positive converge effectivement vers une charge à température nulle  $\rho^{(e)}$  lorsque la température tend vers 0. Ce résultat suit en fait d'un théorème plus général sur la convergence des potentiels de Kantorovich :

**Théorème** (Théorème 5 du Chapitre 1, page 86). *Soit une densité cible  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N \geq 2$  et telle que  $\rho * g_d$  soit uniformément borné. On demande de surcroît que le support  $\Sigma$  de  $\rho$  soit borné. Soit  $V_\beta$  le potentiel de Kantorovich du problème de transport à température positive associé à la densité  $\rho$ . Alors, le potentiel  $V_\beta$  est, à constante additive près, borné dans  $W^{1,\infty}(\mathbb{R}^d)$  uniformément en  $\beta$  lorsque  $\beta \rightarrow \infty$ . En d'autres termes,  $V_\beta$  est uniformément Lipschitz. En particulier, à extraction d'une sous-suite près, il existe  $V \in W^{1,\infty}(\mathbb{R}^d)$  telle que*

$$V_\beta \xrightarrow[\beta \rightarrow \infty]{} V \quad \text{uniformément sur tout compact.} \quad (36)$$

*De plus, la fonction  $V$  est un potentiel de Kantorovich pour le problème de transport à température nulle.*

La convergence des potentiels de Kantorovich pour le problème régularisé vers un potentiel optimal pour le problème sans entropie a été démontré récemment dans [NW22]. Si les résultats dans [NW22] sont plus génériques, notre théorème donne une convergence plus forte, quoiqu'*a priori* seulement dans le cas du coût de Coulomb. Il suit de la convergence au niveau des potentiels de Kantorovich que le charge duale à température positive converge vers une charge duale à température nulle, comme consigné dans le théorème qui suit.

**Théorème** (Théorème 4 du Chapitre 1, page 85). *Soit  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N \geq 2$  et telle que  $\rho * g_d$  soit uniformément borné. Soit  $\rho_\beta^{(e)}$  une charge duale telle que  $\text{supp}(\rho_\beta^{(e)}) \subset \Sigma$ , c'est-à-dire telle que  $V_\beta(x) = \rho_\beta^{(e)} * g_d(x)$  soit l'unique potentiel de Kantorovich à constante près du problème de transport à température positive associé à la densité  $\rho$ . Alors, à extraction d'une sous-suite près, il existe une mesure  $\mu$  dans  $\mathcal{M}_+(\Sigma)$  telle que*

$$\rho_\beta^{(e)} \xrightarrow[\beta \rightarrow \infty]{} \mu \quad \text{pour la topologie faible sur } \mathcal{M}(\Sigma). \quad (37)$$

*De plus, la mesure  $\mu$  est une charge duale pour le problème de transport à température nulle.*

Nous avons vu que la charge duale était toujours de masse  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N - 1$ . Cependant, comme nous l'avons expliqué, nous avons l'intuition que la charge doit être nécessairement de masse *au moins*  $N - 1$ , sans quoi le système ne devrait pas pouvoir être stable par un simple raisonnement sur la neutralité. Il est en fait possible de démontrer que la charge est *exactement* de masse  $N - 1$  — du moins dans le cas où le support de la densité cible  $\rho$  n'est pas borné, auquel cas on peut toujours modifier la charge à l'extérieur de  $\Omega$  à notre bon



vouloir — voir Remarque 2 au Chapitre 1, page 84. Cette égalité suit d’une conjecture sur le comportement asymptotique du potentiel de Kantorovich, que nous présentons maintenant, et qui sera l’objet du Chapitre 2.

## §4. Asymptote du potentiel de Kantorovich (Chapitre 2)

Nous présentons maintenant les résultats consignés dans le Chapitre 2 de cette thèse, qui est une reproduction de notre article [Lel22b]. Il a été conjecturé [FGGG22] que le potentiel de Kantorovich  $V$  associé au transport optimal multimarges à  $N$  particules pour le coût Coulomb doit satisfaire l’asymptote

$$V(x) \sim -\frac{N-1}{|x|} \quad \text{lorsque } |x| \rightarrow \infty. \quad (38)$$

À notre connaissance, si cette conjecture fût formulée pour la première fois par Friesecke *et al.* dans [FGGG22], la question est plus ancienne et on la retrouve à plusieurs reprises dans les travaux de chimistes, voir par exemple [SGGS07; VGG17]. Jusqu’à présent, l’asymptote était rigoureusement connue dans le cas de la dimension  $d = 1$  pour tout nombre de particules  $N$  en utilisant les résultats de Colombo, De Pascale et Di Marino [CDPDM15] déjà mentionnés, et pour  $N = 2$  en toute dimension dans le cas des densités  $\rho$  radiales, auquel cas la solution du transport est explicite comme l’a montré Pass dans [Pas15]. Au Chapitre 2, nous démontrons cette conjecture en toute généralité pour les coûts de Riesz.

**Théorème** (Théorème 19 du Chapitre 2, page 123). *Soit  $\Omega \subset \mathbb{R}^d$  un ouvert connexe et non-borné. Soit  $\rho \in L^1(\Omega, \mathbb{R}_+)$  une densité telle que  $\int_{\mathbb{R}^d} \rho = N$  et telle que  $\rho(r) > 0$  presque partout sur  $\Omega$ . Soit  $V$  un potentiel de Kantorovich pour le transport de transport multimarge avec le coût de Riesz  $w_s(x) = |x|^{-s}$  pour  $s > 0$ . Alors, il existe une constante  $C$  telle que*

$$V(x) + C \sim -\frac{N-1}{|x|^s} \quad \text{lorsque } |x| \rightarrow \infty \text{ et } x \in \Omega. \quad (39)$$

Pour démontrer ce théorème, nous suivons un résultat de [FGGG22], où il est indiqué que l’asymptote suit plus ou moins immédiatement d’une autre conjecture. Notre contribution principale au Chapitre 2 est la preuve de cette conjecture :

**Théorème** (Théorème 21 du Chapitre 2, page 125). *Soit  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N$ . Si  $\mathbb{P}$  est un plan de transport (symétrique) optimal pour le problème de transport multimarge pour le coût de Riesz  $w_s(x) = |x|^{-s}$  avec  $s > 0$ , alors il existe un  $R > 0$  assez grand tel que*

$$\mathbb{P} \left( (\mathbb{R}^d \setminus B_R) \times (\mathbb{R}^d \setminus B_R) \times \mathbb{R}^{d(N-2)} \right) = 0, \quad (40)$$

où  $B_R \subset \mathbb{R}^d$  désigne la boule ouverte de rayon  $R$  centrée à l’origine



Ce théorème dit que si  $\mathbb{P}$  est un plan de transport optimal, alors si une particule — disons  $x_1$  — est envoyée à l'infini  $x_1 \rightarrow \infty$ , toutes les autres particules doivent rester dans un compact de l'espace. Notre démonstration utilise la notion de *c-monotonie cyclique* [San15 ; Vil09].

*Comment ce théorème entraîne-t-il l'asymptote ?* Comme noté par Friesecke *et al.* [FGGG22], étant donnée une densité cible  $\rho$ , nous rappelons que le potentiel de Kantorovich  $V$  est le potentiel qui force les particules à vivre sur un plan de transport optimal  $\mathbb{P}$  pour le problème multimarge à  $\rho$  fixé. En particulier, nous avons vu que les conditions d'optimalité pour le transport optimal devait s'interpréter comme une équation d'équilibre des forces, qu'on rappelle s'écrire

$$\nabla V(x_i) = \sum_{j \neq i}^N \nabla_{x_i} w_s(x_i - x_j) \quad (41)$$

pour tout  $i = 1, \dots, N$ . Cette équation d'équilibre étant vérifiée sur tout le support de  $\mathbb{P}$  et comme le support de la densité  $\rho$  n'est pas compact, on peut envoyer une particule, par exemple  $x_1$ , à l'infini. D'après le théorème susmentionné, toutes les autres particules  $x_2, \dots, x_N$  doivent restées dans un compact. Par conséquent, on obtient lorsque  $x_1 \dots \infty$  que

$$\nabla V(x) \sim (N - 1)w'_s(x) \quad \text{lorsque } x \rightarrow \infty \quad (42)$$

ce qui permet essentiellement de conclure.

Le fait que l'asymptote soit vérifiée est important dans le cadre de la DFT. Sans rentrer dans les détails, l'asymptote assure que l'approximation réalisée par le formalisme SCE permet de rendre compte de certaines propriétés importantes du véritable système — que d'autres approximations, comme par exemple Kohn-Sham, manquent irrémédiablement. On pourra se référer à [VLB94 ; TH03] pour plus de détails, ainsi qu'aux références données à la fin de cette partie. On note aussi que cette asymptote, suivant un argument de Seidl *et al.* dans [SVGG16] — voir aussi [LO81, Annexe A] — permet d'obtenir des informations sur les minimiseurs de l'*inégalité de Lieb–Oxford* [LO81 ; Lie79 ; Sei+22 ; LS09 ; LLS19b]. Cette inégalité est utilisée en DFT pour la calibration des fonctionnelles d'échange-corrélation [LP93 ; PBE96 ; PS22 ; Sun+16]. Nous reviendrons brièvement sur cette question à la fin du Chapitre 2, qui est le sujet d'un article en préparation avec S. Di Marino [DML].

### Lien avec la charge duale

Nous avons énoncé que la charge duale vérifie  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N - 1$ . Grâce à l'asymptote, on peut maintenant affirmer, comme notre intuition nous l'avait dicté au tout début, que cette inégalité est en fait une égalité. Au Chapitre 2, nous démontrons le théorème suivant.

**Théorème** (Corollaire 20 du Chapitre 2, page 124). *Soit  $\Omega \subset \mathbb{R}^d$  un ouvert connexe et non-borné. On suppose que  $\Omega$  ne se "rétrécit pas trop à l'infini" dans certaines directions, c'est-à-dire que  $|A_\Omega| > 0$ , où  $|\cdot|$  est la mesure de Lebesgue sur la sphère  $\mathbb{S}^{d-1}$  et où*

$$A_\Omega := \left\{ \xi \in \mathbb{S}^{d-1} : \text{pour tout } r_0 \text{ il existe } r \geq r_0 \text{ t.q. } r\xi \in \Omega \right\}.$$

Soit  $\rho \in L^1(\Omega, \mathbb{R}_+)$  telle que  $\int_{\mathbb{R}^d} \rho = N$  et telle que  $\rho(x) > 0$  presque partout sur  $\Omega$ . Soit  $\rho^{(e)}$  une mesure positive telle que  $-|\cdot|^{2-d} * \rho^{(e)}$  soit un potentiel de Kantorovich pour le problème de transport multimarge avec le coût de Coulomb. Alors, on a

$$\int_{\mathbb{R}^d} \rho^{(e)} = N - 1. \quad (43)$$

Nous finirons cette partie de notre introduction, consacrée aux résultats du Chapitre 1 et du Chapitre 2, par un commentaire rapide sur la philosophie de la charge duale. Nous avons rapidement mentionné que cette charge, si elle apparaissait dans le cadre du transport optimal pour le coût de Coulomb en [ML13], était du reste un objet que l'on retrouvait aussi dans la littérature de la DFT [Gö99; LAP99]. Dans le cadre de la DFT, il est parfois plus intéressant de considérer la charge qui génère le potentiel d'échange-corrélation plutôt que le dit potentiel en question. La raison étant qu'il est alors beaucoup plus aisée d'un point de vue numérique de forcer l'asymptote. Comme noté dans [VLGG17] "*The main advantage of approximating [the exchange-correlation potential  $V$ ] by modeling [the dual charge  $\rho^{(e)}$ ] is the fact that if a model for  $[\rho^{(e)}]$  integrates to  $[N - 1]$  and vanishes at infinity, then the corresponding  $[V]$  will have the correct asymptotic behaviour [...] which is usually missed by the standard density functional approximations.*". On pourra consulter [Tok05; VLGG17; Mar02; LAP99; KS13; Gö99; GL12; AL01; AAG11; AU85] pour plus de détails sur cette question.

## §5. Transition de phases en dimension une pour les gaz de Riesz (Chapitre 3)

Nous présentons dans cette partie les résultats du Chapitre 3. On y étudie le *gaz de Riesz* [Lew22a] en dimension  $d = 1$ , c'est-à-dire des particules qui vivent sur la droite réelle et qui interagissent par le biais du potentiel de Riesz comme défini ci-dessous (97). On s'intéresse au cas des interactions non-singulières, c'est-à-dire lorsque l'exposant  $s$  dans le potentiel de Riesz est négatif. Dans ce cas, le potentiel de Riesz s'écrit  $-|r|^{-s}$ . On étudie plus particulièrement le cas où  $s$  est compris entre le potentiel de Coulomb  $s = -1$  et l'interaction logarithmique  $s = 0$ . Pour  $s = -1$ , on obtient le *gaz de Coulomb*. Il est connu que ce système remarquable est cristallisé à toute température [Kun74; AM80]. D'autre part, dans le cas de l'interaction logarithmique  $s = 0$ , le modèle correspondant est appelée *log-gaz* de Dyson et il est admis que ce système est un fluide à toute température. Au Chapitre 3, suivant une question soulevée récemment dans [Lew22a], nous montrons numériquement qu'il existe une transition de phase en la température et en l'exposant  $-1 < s < 0$  qui interpolent entre les diagrammes de phases du gaz de Coulomb et du log-gaz.

### §5.1. Définition du gaz de Riesz en dimension arbitraire

Nous définissons le *gaz de Riesz* [Lew22a]. Nous considérons des particules ponctuelles et classiques situées en  $x_1, \dots, x_N$  qui interagissent entre eux par le biais du *potentiel de Riesz*  $w_s$ . Ce potentiel est une généralisation naturelle du potentiel de Coulomb. Pour tout  $s \in \mathbb{R}$ , il est défini par

$$w_s(x) = \begin{cases} |x|^{-s} & \text{pour } s > 0, \\ -\ln|x| & \text{pour } s = 0, \\ -|x|^{-s} & \text{pour } s < 0. \end{cases} \quad (44)$$

Dans le cas où l'exposant  $s$  est négatif, le signe change de telle façon à ce que le potentiel demeure de nature répulsive. On voit que le potentiel de Coulomb en dimension trois correspond au choix  $s = 1$ . Plus généralement le choix  $s = d - 2$  donne le potentiel de Coulomb en dimension arbitraire  $d \geq 1$ , dont nous avons rappelé la définition à la partie précédente, voir (77). En dimension deux d'espace, on retrouve bien le potentiel d'interaction logarithmique  $-\ln|x - x'|$  correspondant au cas  $s = 0$  dans (97) — convention naturelle car elle correspond au premier ordre de  $w_s$  lorsque  $s \rightarrow 0^+$ .

On fixe un domaine  $\Omega \subset \mathbb{R}^d$ , et on contraint les particules à vivre dans  $\Omega$ . Dans ce qui suit, nous nous intéressons au cas de la *longue-portée*, c'est-à-dire lorsque  $s < d$ . Le cas longue-portée est particulièrement intéressant du point de vue de la physique, car il permet l'apparition de transition de phases. Il est aussi généralement plus compliqué pour l'analyse mathématique que le cas *courte-portée*  $s > d$ . Il suit par exemple d'un raisonnement de théorie du potentiel que pour  $s < d$  les électrons, qui cherchent la meilleure configuration possible pour compenser la forte répulsion qu'ils exercent mutuellement les uns sur les autres, vont nécessairement se concentrer sur la frontière de  $\Omega$ . Pour empêcher cela, nous allons rajouter un *fond uniforme* de charge positive sur  $\Omega$ . Ce fond va alors générer un potentiel extérieur qui va permettre de ramener les particules à l'intérieur du domaine  $\Omega$ . On peut alors définir l'énergie du système

$$H_{s,\Omega}(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} w_s(x_i - x_j) - \sum_{i=1}^N \int_{\Omega} w_s(x_i - x') dx' + \frac{1}{2} \iint_{\Omega \times \Omega} w_s(x - x') dx dx'. \quad (45)$$

Le premier terme correspond à l'interaction des particules entre elles, et le deuxième terme à l'interaction des particules avec le fond uniforme. Le troisième et dernier terme correspond à l'énergie d'interaction du fond avec lui-même. Celui-ci est une constante, qui peut donc être négligée lorsqu'on travaille à  $N$  fixé. Cependant, lorsqu'on considère la *limite thermodynamique*, c'est-à-dire la limite  $N \rightarrow \infty$ , ce terme est très important et ne peut pas être négligé lorsque  $s < d$ .

L'idée de rajouter un fond uniforme est bien connu en physique. Dans le cas du potentiel de Coulomb  $s = d - 2$ , on retrouve alors le modèle du *Jellium* [Tho04; Wig34; Lew22a], qui est un modèle très important en physique

de la matière condensée. Dans ce contexte, le fond uniforme doit être vu comme une modélisation simplifiée du réseau atomique dans lequel les électrons sont délocalisés. En pratique, on utilise plutôt en laboratoire des potentiels extérieurs qui peuvent varier en espace et dont la charge associée n'est pas uniforme [CI94; Tho+94; Tho+94; HT94]. Cependant, dans une région où le potentiel varie lentement, on s'attend à ce que le Jellium soit *localement* une bonne approximation.

À température inverse  $\beta \in (0, \infty]$ , nos particules sont modélisées par l'*ensemble canonique*, c'est-à-dire la mesure de Gibbs dont la densité est donnée par

$$\mathbb{P}_{\beta,s,\Omega}(x_1, \dots, x_N) = \frac{1}{Z(\beta, s, \Omega)} \exp(-\beta H_{s,\Omega}(x_1, \dots, x_N)). \quad (46)$$

Ici  $Z(\beta, s, \Omega)$  est la *fonction de partition*, c'est-à-dire la constante de normalisation qui assure que  $\mathbb{P}_{\beta,s,\Omega}$  soit bien une mesure de probabilité, telle que nous l'avons déjà rencontré lors de l'étude de transport optimal entropique. L'*énergie libre* de l'ensemble canonique est donnée par

$$F_s(\beta, \Omega) := -\beta^{-1} \log Z(\beta, s, \Omega). \quad (47)$$

Le gaz de Riesz est l'objet obtenu lorsqu'on considère la limite thermodynamique  $N \rightarrow \infty$  dans les quantités précédentes. En ce qui concerne l'énergie libre, il faut renormaliser par le volume  $|\Omega|$ . On peut alors définir l'énergie libre du gaz de Riesz par unité de volume comme la limite

$$f(s, \beta) := \lim_{\substack{N \rightarrow \infty \\ \Omega \nearrow \mathbb{R}^d \text{ et } |\Omega|=N}} \frac{F_s(\beta, \Omega)}{|\Omega|}. \quad (48)$$

Pour le gaz de Riesz à interaction de courte-portée  $s > d$ , l'existence de cette limite est bien connue. On pourra consulter [Rue99; Geo76; Geo11; DM67]. Dans le cas longue-portée  $s < d$ , généralement plus compliqué à étudier, on pourra se référer à [Lew22a, Sec. III] et aux références mentionnées. Dans le cas spécifique du potentiel de Coulomb  $s = d - 2$ , l'existence a été démontré Lieb et Nahnofer [LN75] en dimension  $d = 3$ , par un argument qui a fait date. L'existence a ensuite été généralisée aux dimensions  $d \geq 2$  par Sari et Merlini [SM76].

En ce qui concerne maintenant la limite de l'ensemble canonique  $\mathbb{P}_{\beta,s,\Omega}$ , on s'attend à ce que l'objet obtenu à la limite  $N \rightarrow \infty$  soit un *processus de points* de type *gibbsien* [Der19; Geo11; Lew22a; Jan]. Il existe toute une famille de caractérisations/définitions rigoureuses cet objet, qui sont généralement équivalentes. Par exemple, on peut mentionner les équations de *Dobrushin–Landford–Ruelle* (DLR) ou encore la hiérarchie de *Bogolioubov–Born–Green–Kirkwood–Yvon* (BBGKY) [GLM80]. Le processus limite est notamment caractérisé par ses *fonctions de corrélations à  $k$ -points*, définies en volume fini  $N < \infty$  par

$$\rho^{(k)}(x_1, \dots, x_k) := \frac{N!}{(N-k)!} \int_{\mathbb{R}^{d(N-k)}} \mathbb{P}_{\Omega,s,\beta}(x_1, \dots, x_k, dx'_{k+1}, \dots, dx'_N). \quad (49)$$

Une première question fondamentale dans l'étude des gaz de Riesz est l'existence de ce processus limite, qui n'est pas facile. Si le cas des interactions courte-portées  $s > d$  est bien connu [Rue99; Geo76; Geo11; DM67], le cas de la longue-portée  $s < d$  l'est moins [Lew22a, Sec. III]. L'existence étant acquise, une autre question fondamentale est celle de l'unicité de ce processus limite. En effet, la non-unicité du processus est intrinsèquement liée aux brisures de symétrie et aux transitions de phases. Pour finir, il se pose aussi le problème de déterminer la nature de la phase (*e.g* solide, liquide, gaz etc.), qui est reliée aux propriétés des fonctions de corrélations (102), et tout particulièrement à la fonction de corrélation à deux points  $\rho^{(2)}(x, x')$ , comme nous le verrons plus tard.

Il est aussi intéressant de définir le gaz de Riesz avec des conditions périodiques aux bords [Lew22a, Sec. IV], où les particules interagissent entre elles ainsi qu'avec leurs images dans les périodisés du domaine  $\Omega$ . Dans le cas courte-porté  $s > d$ , le potentiel de Riesz périodisé  $\widetilde{v}_s$  s'écrit alors

$$\widetilde{v}_s(x) = \sum_{z \in \mathcal{L}} \frac{1}{|x - z|^s}, \quad (50)$$

où  $\mathcal{L}$  désigne le réseau périodique. Si cette série est convergente lorsque  $s > d$ , cela n'est plus le cas pour les potentiels de Riesz de longue-portée  $s < d$ . On peut cependant montrer que la fonction  $\widetilde{v}_s(x)$ , vue comme une fonction de la variable complexe  $s \in \mathbb{C}$ , admet une extension méromorphe sur  $\mathbb{C} \setminus \{d\}$  avec un pôle à  $s = d$  [BBS89; Bor+88; BBT85; BBS14], voir aussi [Lew22a, Sec. IV], à l'image de la fonction zeta de Riemann à laquelle elle est reliée. On peut en fait définir ce potentiel périodisé grâce à sa transformée de Fourier, qui est donnée à une constante multiplicative près par

$$\widehat{\widetilde{v}_s}(k) = \sum_{k \in \mathcal{L}^* \setminus \{0\}} \frac{\delta_k}{|k|^{d-s}}. \quad (51)$$

Ici,  $\mathcal{L}^*$  désigne le réseau dual de  $\mathcal{L}$ , c'est-à-dire le réseau engendré par la base duale de celle de  $\mathcal{L}$ . Il se trouve en fait que l'extension méromorphe à  $d - 2 < \Re(s) < d$  correspond exactement à l'ajout dans (103) du fond uniforme telle que nous l'avons introduit précédemment. On se référera à [Lew22a, Sec. IV] pour plus de détails sur ces questions.

En pratique, les conditions périodiques aux bords sont particulièrement utiles du point de vue du numérique lorsque  $s < d$ . Ces dernières permettent notamment de supprimer les effets de bords, qui sont peuvent être considérables dans le cas des interactions à longue-portée. Au Chapitre 3, nos expériences numériques sont réalisées avec ces conditions périodiques aux bords.

## §5.2. Le cas de la dimension $d = 1$

Nous nous intéressons à partir de maintenant et jusqu'à la fin de cette introduction au cas de la dimension  $d = 1$ . Nous présentons les résultats théoriques connus pour le gaz de Riesz en dimension une, et nous discutons de deux

modèles intégrables très importants, que sont le *gaz de Coulomb* et le *log-gaz*, qui correspondent respectivement à  $s = -1$  et  $s = 0$

En ce qui concerne l'existence de l'énergie libre en limite thermodynamique (101), celle-ci a été démontrée par Kunz [Kun74] pour le cas du potentiel de Coulomb  $s = -1$ . Dans le cas  $s \geq 0$ , l'existence est contenue dans les travaux de Serfaty *et al.* On pourra se référer [Ser15] et les références de [Lew22a]. Si dans ces travaux le cas des exposants négatifs  $s < 0$  n'est pas traité *sic*, ils seraient tout de même couverts par la théorie [Ser]. On peut aussi mentionner les travaux de physiciens [GLM80; GLM81; BGM84] — voir les références de [Lew22a]. La limite thermodynamique de l'énergie libre existe donc pour tout  $s \geq -1$ . Il est d'ailleurs probable que l'existence tienne jusqu'à  $s > -2$  [Lew22a], même si le résultat n'est pas connu. Dans le cas où on impose des conditions aux bords périodiques sur l'intervalle, la limite a été démontrée dans [Lew22a] suivant une extension d'un résultat de [Har+15].

En ce qui concerne l'existence et l'unicité du processus de points limite en dimension  $d = 1$ , il est connu que le processus limite est invariant par translation pour  $s > 1$  [FP81] et qu'il est unique pour  $s > 2$  [Pap87]. Par conséquent, le gaz de Riesz pour  $s > 2$  n'a pas de transition de phases et il est un fluide à toute température. L'invariance par translation n'étant pas brisée pour  $s > 1$ , on s'attend en droit à ce que le processus soit unique pour  $s > 1$ . En fait, on pense que cela est certainement le cas pour tout  $s \geq 0$ . Le cas  $0 < s < 1$  a été étudié récemment par Dereudre et Vasseur [DV23], qui ont démontré l'existence des équations DLR pour le processus limite, et par Boursier [Bou22] qui a obtenu une série de résultats dans le cas périodique. Un autre cas, tout à fait remarquable, où l'existence et la (non-)unicité du processus limite sont connues est pour le potentiel de Coulomb  $s = -1$ , comme nous en discutons maintenant.

Un cas très spécifique est le *gaz de Coulomb*  $s = -1$ . Ce modèle intégrable est très bien étudié dans la littérature [AM80; AJJ10; BLC63; CGZJ20; Cho75; Kun74]. On doit à Kunz [Kun74] plusieurs résultats remarquables sur le gaz de Coulomb, dont l'existence de l'énergie libre déjà mentionnée ainsi que l'existence des *fonctions de corrélations* du processus limite, dont il donne de surcroît une formule explicite. Ces fonctions ne sont pas uniques, ce qui permet d'établir la non-unicité du processus et par conséquent la brisure de symétrie à toute température. Il montre de plus que ces fonctions sont périodiques, ce qui prouve que le système est un cristal à toute température. Cependant, il faut encore que ces fonctions ne soient pas constantes. Par un argument d'analyticit , Kunz d montre que c'est effectivement le cas   toute temp rature assez petite. Ce r sultat sera g n ralis  peu apr s   toute temp rature par Aizenman et Martin [AM80], qui ont aussi d montr  que les processus limites v rifiaient les  quations de BBGKY et DLR. Le cas du gaz de Coulomb quantique a  t  r solu par Brascamp et Lieb [BL02] ainsi que Jansen et Jung [JJ14]. On insistera qu'il est tout   fait fortuit que le gaz de Coulomb soit un mod le int grable. Ceci est notamment d    la forme tr s sp cifique du potentiel de Coulomb en dimension une, c'est- -dire  $-|r|$ . Par exemple, on peut voir que la force qu'exerce une particule sur une autre ne d pend pas de leur distance mutuelle, ce qui permet d'obtenir des propri t s tr s int ressantes qui simplifient grandement l'analyse math matique.



Un autre cas très important est celui de l'interaction logarithmique  $s = 0$ , pour laquelle on trouve le modèle du *log-gaz* en dimension  $d = 1$ . Ce modèle a été introduit dans les travaux de Dyson [Dys62a; Dys62b; Dys62c; DM63; MD63] qui sont avec les travaux de Wigner [Wig55; Wig67] à l'origine de la *théorie des matrices aléatoires* en mathématiques [For10; Meh04]. Dans ce contexte, le log-gaz est aussi appelé  $\beta$ -ensemble ou encore *sine- $\beta$* . Pour  $\beta = 1, 2$  et  $4$ , le log-gaz correspond aux trois *ensembles gaussiens* bien connus dans la théorie des matrices aléatoires, que sont respectivement le GOE (pour *Gaussian Orthogonal Ensemble*), le GUE (pour *Gaussian Unitary Ensemble*) et le GSE (pour *Gaussian Symplectic Ensemble*) [For10]. Le log-gaz est aussi un modèle intégrable qui est abondamment étudié dans la littérature. On référera le lecteur aux très nombreuses références de [Lew22a], ou encore au monographe [For10]. Si l'unicité du log-gaz n'est pas *a priori* connu rigoureusement en toute généralité, il est prédit qu'il ne peut pas y avoir de brisure de symétrie [RW90]. En l'occurrence, l'unicité a été démontrée dans le cas particulier où l'on se restreint aux processus de points stationnaires [EHL21]. Par conséquent, on s'accorde sur le fait que le log-gaz en dimension une n'a pas de transition de phases, et qu'il est un fluide à toute température, à l'exception de la température nulle où il est cristallisé [SS15; Leb15].

Si le log-gaz ne présente pas de brisure de symétrie à température positive, il existe cependant une transition de type *Berezinskii–Kosterlitz–Thouless* (BKT) [Mud14; Her07; KT73] à  $\beta = 2$ , dont il nous faut dire quelques mots. En dimension une, cette transition s'exprime par un changement qualitatif du comportement de la fonction de corrélation à deux points  $\rho^{(2)}(r, r')$  (102). En utilisant une théorie des fluides quantiques en une dimension de Haldane [Hal81], Forrester a conjecturé dans [For84] que

$$g(r) \underset{r \rightarrow \infty}{\sim} \begin{cases} 1 - \frac{1}{\pi^2 \beta r^2} & \text{for } \beta < 2, \\ 1 + \frac{\cos(2\pi r)}{2\pi^2 r^2} - \frac{1}{2\pi^2 r^2} & \text{for } \beta = 2, \\ 1 + c \frac{\cos 2\pi r}{r^{4/\beta}} - \frac{1}{2\pi^2 r^2} & \text{for } \beta > 2 \end{cases} \quad (52)$$

pour une constante universelle  $c$ . Ici, la fonction de corrélation à deux points  $\rho^{(2)}(r, r')$  ne dépendant que de la distance  $r - r'$ , on pose  $g(r) := \rho^{(2)}(0, r)$ . Nous voyons qu'à la température inverse  $\beta = 2$ , on passe d'une décroissance *universelle* de la fonction corrélation à une décroissance dont l'exposant dépend de la température. De plus, on observe l'apparition d'un terme oscillant. Cependant, à l'inverse de l'ordre cristallin, ces oscillations sont peu à peu atténuées pour finalement disparaître lorsque  $r \rightarrow \infty$ . La conjecture de Forrester est vérifiée dans les cas particuliers  $\beta = 1, 2$  et  $4$ , et plus généralement pour tout  $\beta$  pair ou rationnel, voir [For93] et [For10, Chap. 13].

Pour finir, on peut mentionner le cas  $s = 2$ , qui correspond au modèle classique de *Calogero–Sutherland–Moser* (CSM) [Sut03a; Sut04; Sut03b; Cal71], qui est entièrement intégrable [Cho00; GM73]. Un fait remarquable est que la version quantique de ce modèle est équivalente au log-gaz — voir les références

susmentionnées. C'est d'ailleurs par cette analogie et par les travaux de Haldane que Forrester a obtenu le comportement asymptotique mentionné au-dessus.

Dans cette thèse, nous nous intéressons au cas des exposants  $-1 < s < 0$  qui interpolent entre le gaz de Coulomb et le log-gaz. Suivant les résultats qui ont été énoncés, Lewin dans [Lew22a] a posé la question suivante : *Existe-il une transition de phases pour les gaz de Riesz d'exposants  $-1 < s < 0$  qui interpolent entre le diagramme de phases du gaz de Coulomb, pour lequel nous sommes toujours un solide, et le log-gaz pour lequel nous sommes toujours un fluide ?* Le reste de cette introduction est dédiée à la présentation de nos résultats numériques du Chapitre 3, qui répondent par l'affirmative à cette question.

### §5.3. Transition de phases dans les gaz de Riesz : résultats numériques

Nous cherchons donc à déterminer s'il existe une transition de phases en la température  $T$  pour les gaz de Riesz en une dimension d'espace pour les exposants  $-1 < s < 0$ . Nous notons que la majorité des résultats concernant les transitions de phases dans les gaz de Riesz en dimension  $d \geq 1$  sont des résultats numériques. Pour répondre à notre question par un raisonnement théorique, il conviendrait d'abord de pouvoir définir rigoureusement le gaz de Riesz pour ces exposants, par exemple en exhibant une solution aux équations DLR ou à la hiérarchie BBGKY associés, comme cela a été fait pour le cas  $0 < s < 1$  par Dereudre et Vasseur [DV23]. Plus encore, il faudrait pouvoir répondre à la question de l'unicité ou de la non-unicité de ce gaz. Ces problèmes sont difficiles et très largement ouverts actuellement. Nous rappelons que le seul résultat connu rigoureusement pour les transitions de phases en dimension une est l'unicité du gaz de Riesz pour  $s > 2$  [Pap87]. Nos résultats du Chapitre 3 sont donc entièrement numériques.

Avant d'en venir à ces résultats, il nous semble intéressant de mentionner rapidement ce qui est connu dans le cas des dimensions  $d = 2, 3$ .

#### Le cas de la dimension $d = 2$ et $d = 3$

La dimension  $d = 3$  est la dimension pour laquelle les résultats numériques sont les plus abondants dans la littérature. À haute température, le gaz de Riesz est dans un état gazeux — ce qui peut être rigoureusement démontré dans le cas courte-portée  $s > d$  [Lew22a, Thm. 46]. Il est ensuite admis qu'il existe une transition de phase *solide-fluide* à température assez petite pour tout  $s > 0$ . Dans le régime solide, il est de surcroît connu qu'il existe une transition *solide-solide* où le système transite entre un cristal cubique centré (BCC) et un cristal cubique à faces centrées (FCC) pour certaines valeurs de  $s$ , mais pas pour le cas Coulombien  $s = 1$ , où seul le cristal BCC semble exister. On pourra consulter [Lew22a, Sec. VI. B.] pour des références classiques sur ce sujet.

En dimension  $d = 2$ , les choses sont plus subtiles. L'existence de transition de phases en dimension deux d'espace fut le sujet de plusieurs controverses dans l'histoire de la physique contemporaine, notamment car des travaux numériques



semblaient mettre en évidence l'existence de transitions là où un théorème mathématique bien connu semblait l'interdire. Il est en effet connu qu'il ne peut pas y avoir de brisure de symétrie spontanée en petite dimension  $d \leq 2$  pour des potentiels à courte-portée. Une telle interdiction est énoncée par le fameux théorème de *Hohenberg–Mermin–Wagner* [Hoh67; Mer67; Mer68; MW66]. Il est cependant à noter que ce théorème ne s'applique *a priori* que pour des potentiels d'interaction de courte-portée. En particulier, le potentiel de Coulomb  $s = 0$  en dimension deux ou plus généralement les potentiels de Riesz de longue-portée  $s < d$  pour  $d = 1, 2$  ne rentrent pas dans les clous de ce théorème. En l'occurrence, comme nous l'avons indiqué, le gaz de Coulomb en dimension une est cristallisé à toute température [Kun74; AM80]. Alder et Wainwright<sup>9</sup>[AW62] furent les premiers à mettre en lumière une transition de phase dans un système bidimensionnel là où la théorie semblait l'interdire. Une décennie plus tard, Berezinskii, Kosterlitz et Thouless [Kos; Ber71; Ber72] mirent en lumière l'existence de transitions de phase d'un type très particulier, qui sont dites *topologiques*, et que nous avons déjà rencontré dans le cas du log-gaz en dimension une (voir au-dessus). Kosterlitz et Thouless reçurent le Prix Nobel de 2016 pour leurs travaux. Kosterlitz, dans sa réception du prix, cita notamment les travaux de Alder et Wainwright —“*It also helped that there existed some experimental and numerical evidence for transitions to more ordered low temperature phases in 2D crystals*” [Kos]. En ce qui concerne nos gaz de Riesz en dimension  $d = 2$ , si plusieurs travaux numériques mentionnaient pendant longtemps l'existence d'une transition du premier ordre dans le cas longue-portée  $s < 2$  [Str88], des travaux plus récents mettent plutôt en évidence une transition d'ordre topologique à la BKT — on réfère le lecteur à [Lew22a, Sec. VI. B.] pour une liste de références.

<sup>9</sup>Pionniers de l'utilisation des ordinateurs en physique statistique. On pourra consulter [BC18] qui retrace le parcours du physicien Berni Alder (1925 – 2020) et par la même occasion l'histoire de l'introduction des ordinateurs en physique. Histoire qui ne s'est pas déroulée non sans quelques réticences d'une partie de la communauté des physiciens, et qui s'est transfigurée peu à peu par l'arrivée d'ordinateurs toujours plus puissants sur le marché. L'article [BC18] relate une anecdote amusante, sur un vote à main levée : “*In the round table on statistical mechanics that took place just before Alder's presentation, Kirkwood summed up the situation as follows : “the most recent evidence for the existence of the liquid-solid transition was provided by Dr. Alder [...] Following his work, Dr. William Wood of Los Alamos again took up the Monte Carlo calculations... it seems that the Rosenbluth calculations were in error at high densities ; there is a very definite evidence for a first-order transition, which we would identify as a transition from a solid or crystalline state to a liquid state”. The exchange of viewpoints that followed showed clearly that, in spite of the evidence presented by both methods, the question was still far from being settled. [...] George Uhlenbeck half jokingly put the question [...] on a vote, remembering the audience that a vote taken in a similar occasion the previous fall, as to whether a gas of hard spheres has a transition point or not, had ended up even ; “Now of course, with Mr. Kirkwood here, we have weighted the evidence a little bit. May I ask first of the panel to put up their hands if they believe there is a transition in the classical case... Now those who believe there is not a transition... Even, again !”*”

### Le cas de la dimension $d = 1$ et $-1 < s < 0$

Nous présentons ici les résultats obtenus au Chapitre 3, sur le diagramme de phases du gaz de Riesz en une dimension d'espace pour  $-1 < s < 0$ . En dimension  $d = 1$ , la littérature sur les transitions de phases est bien moins abondante qu'en dimensions  $d = 2, 3$ . Suivant une reproduction de [Lew22a, Fig. 5], nous résumons à la Figure 9 les résultats connus rigoureusement ou du moins admis, que nous avons déjà mentionnés précédemment. On rappelle que le *gaz de Coulomb*  $s = -1$  cristallisé à toute température [Kun74; AM80], et qu'il est prédit que le *log-gaz*  $s = 0$  est toujours un fluide à température positive, avec une transition BKT à  $\beta = 2$ .

Au Chapitre 3, nous montrons numériquement que l'intuition formulée dans [Lew22a] semble correcte, c'est-à-dire qu'il existe bien une température critique  $T_s$  dépendant de l'exposant  $-1 < s < 0$  telle qu'en dessous de cette température le gaz de Riesz soit un solide, tandis qu'au dessus le système est un fluide. De plus, cette température interpole bien les résultats connus — ou du moins attendus — pour le gaz de Coulomb et le log-gaz, à savoir que  $T_s \rightarrow \infty$  lorsque  $s \rightarrow -1$ , ce qui est consistant à le fait que le gaz de Coulomb est cristallisé à toute température, et que  $T_s \rightarrow 0$  lorsque  $s \rightarrow 0$ , ce qui consistant à le fait que le log-gaz est un fluide à toute température. De plus, nos expériences numériques semblent indiquer qu'il existe une deuxième transition de phase  $\tilde{T}_s$  correspondant à une transition de type BKT d'une phase liquide à une phase quasi-solide. On observe que  $\tilde{T}_s$  converge vers  $1/2$  lorsque  $s \rightarrow 0$ , ce qui est consistant avec le point de transition BKT du log-gaz. Le diagramme de phase schématique pour  $-1 < s < 0$  est alors donné par la Figure 10.

Pour obtenir ce diagramme, nous avons simulé le gaz de Riesz par des méthodes de Monte Carlo, en imposant des conditions périodiques aux bords. On calcule ensuite la fonction de corrélation  $g(r)$ . Cette fonction, qui exprime comment la densité de particules d'un système varie par rapport à une particule de référence fixée, est une quantité très importante en mécanique statistique pour l'étude des transitions de phases. Par exemple, dans le cas d'un cristal parfait, la fonction de corrélation est périodique — à température nulle, c'est même un peigne de Dirac. En effet, étant donnée la position d'une particule, les positions des autres particules sont déterminées par la géométrie du réseau cristallin. Au contraire, dans le cas d'un fluide parfait, les particules sont indépendantes les unes des autres, si bien que  $g(r)$  est une fonction constante égale à la densité moyenne du système. Plus généralement, en présence d'un (quasi-)ordre cristallin, on s'attend à ce que  $g(r)$  présente des oscillations et/ou une décroissance non intégrable à longue-portée. Au contraire, dans le cas d'un système désordonné, on s'attend à ce que  $g(r)$  décroisse rapidement à l'infini vers la densité moyenne du système, ce qui est équivalent à dire que deux particules assez éloignées l'une de l'autre ne sont pas corrélées, comme dans un gaz ou un liquide. Pour le gaz de Coulomb avec des conditions périodiques aux bords, Kunz a démontré que  $g(r)$  était une fonction proprement périodique, et par conséquent que le système était cristallisé.

Suivant cette phénoménologie, nous cherchons donc à déterminer si la fonction de corrélation pour le gaz de Riesz aux exposants  $-1 < s < 0$  devient

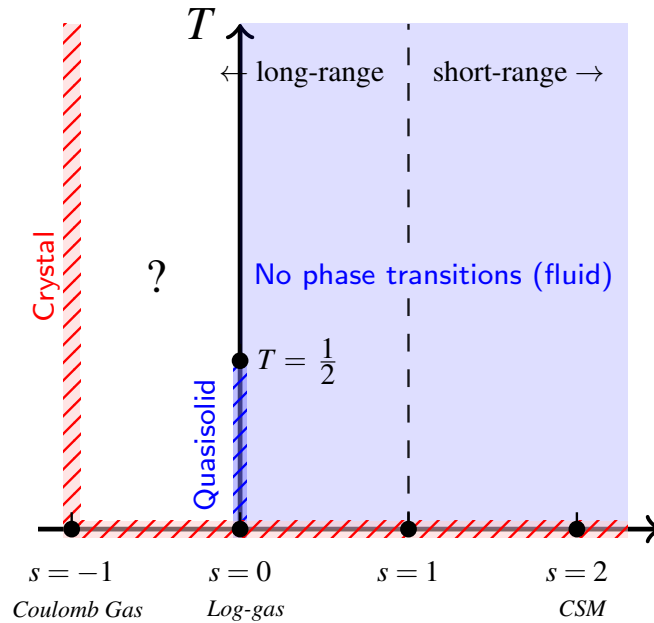


FIGURE 3 : Reproduction de [Lew22a, Fig. 5] du diagramme de phases du gaz de Riesz en une dimension d'espace en fonction de la température et de l'exposant  $s$  du potentiel de Riesz. Notre but au Chapitre 3 est de compléter le diagramme pour  $-1 < s < 0$ . Suivant [Lew22a], nous nous demandons s'il existe une transition de phase qui interpole entre le diagramme de phases du gaz de Coulomb et celui du log-gaz.

périodique lorsque la distance  $r$  est grande, ce qui serait une manifestation très claire de l'ordre cristallin. Au contraire, si la fonction de corrélation  $g(r)$  converge rapidement et de façon monotone vers la densité moyenne, on s'attend à ce que le système soit un fluide. Dans le cas de la transition BKT et de la phase quasi-solide associée,  $g(r)$  doit avoir des oscillations qui doivent progressivement s'atténuer pour finalement disparaître. Ceci est cependant difficile à vérifier en pratique dans le cas d'un calcul numérique où nécessairement  $N < \infty$ . À la Figure 11 ci-dessous, l'image de gauche montre la fonction  $g(r)$  calculée à trois températures différentes pour le gaz de Riesz d'exposant  $s = -0.5$  par notre algorithme. On observe que pour la température la plus haute, la fonction  $g(r)$  décroît rapidement vers la densité moyenne, qui ici est unitaire, comme dans un fluide. Pour les deux autres températures, plus basses, on voit au contraire que  $g(r)$  est une fonction oscillante. Cependant, il est difficile de déterminer si ces oscillations finissent par disparaître lorsque  $r \rightarrow \infty$ , et donc de décider si nous sommes en présence d'une phase solide ou quasi-solide, comme cela est le cas pour le log-gaz à température  $\beta > 2$ . C'est le même problème que celui

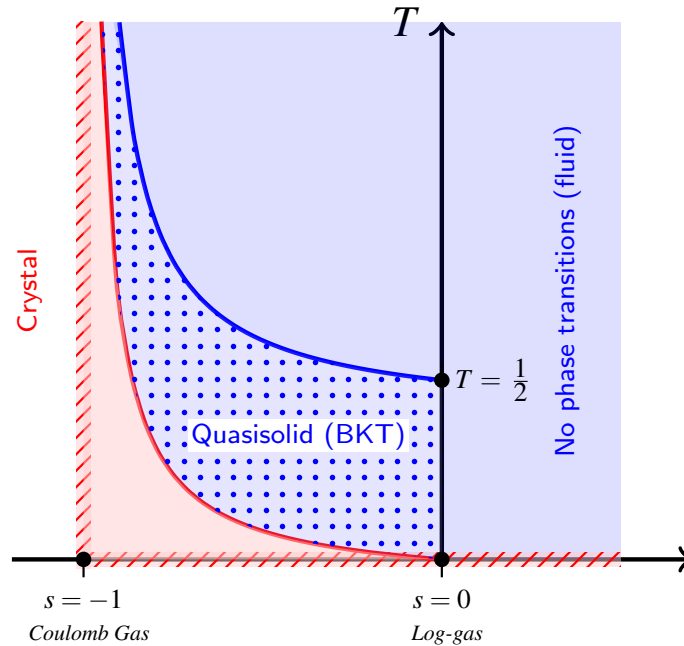


FIGURE 4 : Diagramme de phases du gaz de Riesz en une dimension d'espace complétée dans la région  $-1 < s < 0$  selon les résultats numériques présentés au Chapitre 3. L'allure est consistante avec l'intuition de [Lew22a], à savoir qu'il existe une température critique qui sépare une phase solide et une phase désordonnée, consistante avec les diagrammes de phases du gaz de Coulomb  $s = -1$  et du log-gaz  $s = 0$ . Nous trouvons de plus l'existence d'une transition de type BKT, correspondant à la formation d'un quasi-solide.

ayant engendré les nombreuses controverses en dimension  $d = 2$ , comme nous l'avons mentionné plus haut.

Pour mieux comprendre cette transition, nous étudions donc le *facteur de structure*  $S(k)$  [HM13, Chap. 4]. Cette quantité est définie comme la transformée de Fourier de la fonction de corrélation tronquée  $g(r) - 1$ , c'est-à-dire

$$S(k) := 1 + \frac{1}{2\pi} \int_{\mathbb{R}} e^{ikr} (g(r) - 1) dr. \quad (53)$$

La fonction  $S(k)$  est utilisée en physique de la matière condensée et en cristallographie, car elle est plus facilement accessible expérimentalement que la fonction de corrélation  $g(r)$ , que l'on détermine donc en pratique comme transformée de Fourier inverse de  $S(k)$ . Elle est liée aux propriétés de diffusion — *i.e. scattering* — du matériau [DB08 ; GH04 ; Sir+89 ; Yar+73]. En effet, pour un (quasi)-solide de taille caractéristique  $a > 0$ , on peut accéder à la géométrie du réseau cristallin par diffusion d'ondes de longueur  $\lambda \gg a$ , ou de façon équivalente de petit nombre d'onde  $k \ll a^{-1}$ .

Si la fonction de corrélation possède une composante périodique, le facteur de structure doit avoir un pic de Dirac associé à la période. Dans le cas où la densité moyenne du système est unitaire, ce pic doit être présent à  $k = 1$ . Dans le cas d'un quasi-solide où les oscillations disparaissent, comme pour le log-gaz, il ne s'agira pas d'une masse de Dirac mais plutôt d'une singularité intégrable. En fait, dans le cas du log-gaz, il vient à partir du comportement de  $g(r)$  conjecturé par Forrester telle que rappelée plus haut au (105), qu'il existe trois régimes différents. Au dessus de la transition BKT  $\beta < 2$ , la fonction  $S(k)$  est monotone en  $k = 1$ . En dessous de la transition BKT et  $2 < \beta < 4$ , la fonction  $S(k)$  n'est plus monotone en  $k = 1$ , mais elle reste cependant bornée. Finalement, pour  $\beta > 4$ , la fonction  $S(k)$  diverge en  $k = 1$  — logarithmiquement pour  $\beta = 4$  et comme  $|1 - k|^{4/\beta - 1}$  pour  $\beta > 4$ . Ce qui est ici important à noter, c'est que cette singularité est dans tous les cas intégrable. Ceci est donc différent d'un phase solide, où on s'attend à ce que  $S(k)$  ait une masse de Dirac en  $k = 1$ .

Il existe un lien non-trivial entre la brisure de symétrie et le comportement de  $S(k)$  proche de l'origine. Ce lien ne tient *a priori* que pour les systèmes unidimensionnels invariants par translation. En effet, dans ce contexte, Aizemann, Goldstein et Lebowitz [AGL01] ont donné une condition suffisante pour qu'il y ait brisure de symétrie. Ce résultat, qui est lié à la notion d'hyperuniformité [TS03; Tor18], énonce que si  $S(k)$  se comporte comme  $C|k|^\eta$  lorsque  $k \rightarrow 0$  pour une certaine constante  $C$  et pour un exposant  $\eta > 1$ , alors nécessairement l'invariance par translation doit être brisée. Ceci est consistant avec le gaz de Coulomb, pour lequel  $\eta = 2$ . Pour le log-gaz, il suit de la conjecture de Forrester (105) que  $\eta = 1$ . En toute généralité, il semble que l'exposant naturel soit  $\eta = 1 - s$ . Ceci est connu dans le cas  $0 < s < 1$  d'après les résultats obtenus récemment par Boursier [Bou22]. Forrester a d'ailleurs donné dans [For10, Chap. 11] — voir aussi [AM85] — une heuristique dans le cas  $s = 0$  qui s'étend facilement au cas  $s$  général, et que nous reproduisons au Chapitre 3. Cependant, cet argument se base sur une prémisse essentiellement équivalente à la cristallisation. Aussi, nous conjecturons plutôt que *si le gaz de Riesz est cristallisé, alors nécessairement  $\eta = 1 - s$ .*

À partir de ces différents critères — (non-)oscillations de  $g(r)$ , (non-)monotonie et/ou singularité de  $S(k)$  en  $k = 1$ , et comportement de  $S(k)$  lorsque  $k \rightarrow 0$  — il est possible de déterminer numériquement le diagramme de phase que nous avons présentée à la Figure 10. Si nous revenons à la Figure 11, l'image de droite montre le facteur de structure  $S(k)$  pour nos trois températures. On voit pour la température  $T = 1$ , la fonction  $S(k)$  se comporte linéairement lorsque  $k \rightarrow 0$  et présente un pic à  $k = 1$ , ce qui semble donc indiquer qu'à cette température le gaz de Riesz est un quasi-solide. Au contraire, pour  $T = 0.2$ , la fonction  $S(k)$  se comporte comme  $C|k|^{1-s}$  lorsque  $k \rightarrow 0$  pour une certaine constante  $C > 0$ , ce qui semble indiquer que le système est un solide à cette température d'après [AGL01].

Nous sommes donc amenés à conjecturer l'existence de deux températures critiques. Une première, que nous notons  $\tilde{T}_s$ , correspondant à une transition BKT et à la phase quasi-solide associée, et une deuxième température critique, notée  $T_s$ , qui correspond à la transition solide. En utilisant les différents critères

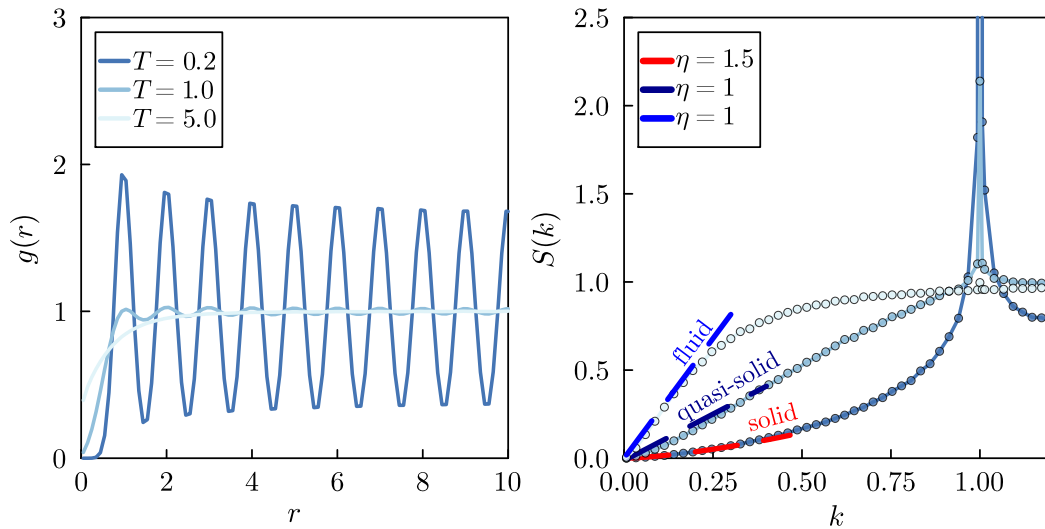


FIGURE 5 : On affiche ici la fonction de corrélation  $g(r)$  (à gauche) pour le gaz de Riesz d'exposant  $s = -0.5$ , obtenu par notre algorithme, ainsi que le facteur de structure  $S(k)$  déterminé à partir de la transformée de Fourier de  $g(r)$  (à droite). On voit que pour la température  $T = 5$ , la fonction  $g(r)$  décroît rapidement et de façon monotone vers la densité moyenne  $\rho$  de notre système (ici  $\rho = 1$ ) comme dans un fluide. Le facteur de structure ne présente alors pas de pic à  $k = 1$ . Au contraire, pour des températures plus basses, la fonction  $g(r)$  est oscillante et on voit l'apparition du pic à  $k = 1$  pour  $S(k)$ . Pour  $T = 1$ , la fonction  $S(k)$  se comporte linéairement lorsque  $k \rightarrow 0$ , ce qui laisse à penser que le système est un quasi-solide. Pour  $T = 0.5$ , elle se comporte plutôt comme  $C|k|^{1-s}$  et le pic en  $k = 1$  se comporte plus comme une masse de Dirac, ce qui d'après Aizenman *et al.* [AGL01], laisse à penser que le système est solide.

mentionnés, nous pouvons tracer un diagramme de phases approché en fonction de la température  $T$  et de l'exposant  $s$ , représentée à la Figure 12. Déterminer précisément les deux températures  $T_s$  et  $\tilde{T}_s$  est compliqué de par la nature approchée du numérique. Cependant, indépendamment de cette certaine part d'arbitraire, il nous semble que les résultats montrent bien qualitativement l'existence d'une transition de phase pour les gaz de Riesz d'exposant  $-1 < s < 0$ , dont le comportement est consistant avec le diagramme de phases du gaz de Coulomb et du log-gaz, répondant ainsi par l'affirmative à la question soulevée par Lewin [Lew22a].

Pour vérifier que notre algorithme est fonctionnel, nous l'avons aussi testé sur le log-gaz. Cela nous permet de confirmer la conjecture de Forrester (105), notamment pour ce qu'elle implique sur le comportement du facteur de structure  $S(k)$  proche de  $k = 1$  et de l'origine  $k = 0$ .

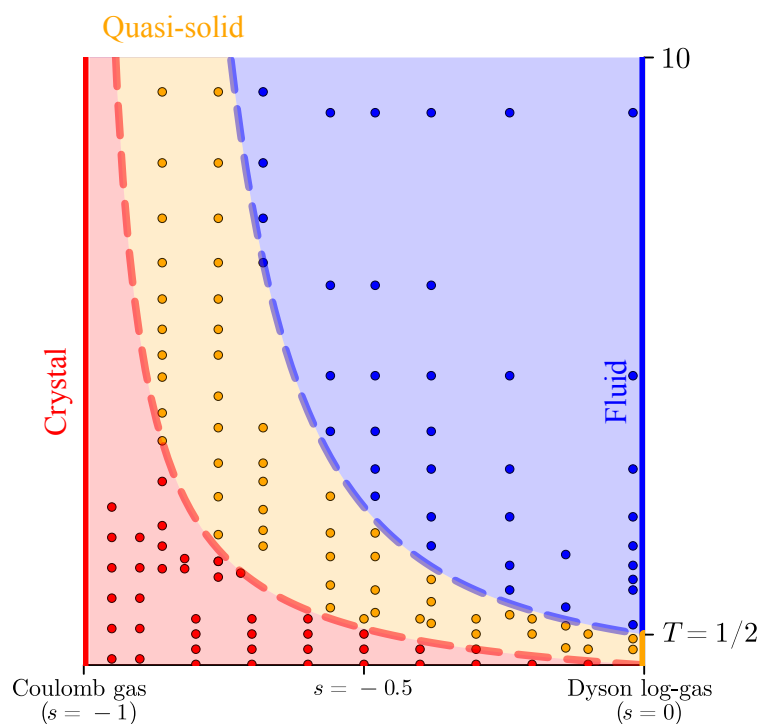


FIGURE 6 : Diagramme de phases du gaz de Riesz en une dimension d'espace en fonction de la température  $T$  et de l'exposant  $-1 < s < 0$ . Les points bleus correspondent à la phase liquide. Les points oranges correspondent à la phase quasi-solide, pour laquelle le facteur de structure a un pic à  $k = 1$  mais se comporte linéairement lorsque  $k \rightarrow 0$ . Finalement, les points rouges correspondent à la phase solide, pour laquelle  $S(k)$  se comporte sous-linéairement lorsque  $k \rightarrow 0$ . On trace alors  $T_s$  et  $\tilde{T}_s$  schématiquement.

# INTRODUCTION (IN ENGLISH)

*In this introductory chapter<sup>10</sup> we begin by introducing a generic problem in statistical physics, that of determining the ground-state energy of a system of interacting classical particles, and we show how this problem can be expressed using the theory of multimarginal optimal transport. We then recall some important results regarding the multimarginal optimal transport with repulsive costs as arising in physics. We then introduce a discretization of this problem in the case of the Coulomb cost, based on the physical interpretation of the Kantorovich duality, and which consists in writing the Kantorovich potential as an electrostatic potential generated by an external charge distribution. We state several properties of this charge, which will be proved in Chapter 1. We then state the results of Chapter 2 concerning the asymptotic behaviour of the Kantorovich potential for repulsive costs. Finally, in the last part of this introduction, we present the numerical results obtained in Chapter 3 regarding the existence of phase transitions for Riesz gases in one space-dimension with very long-range interaction.*

## Summary of the present chapter

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<sup>10</sup>This chapter is a translation in english of the previous introductory chapter in french.



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## §1. A generic problem in statistical physics

We start with a very generic problem in physics, that of determining the ground-state energy of a system of interacting particles. We show how this problem, which is difficult to solve numerically, can be rewritten in such a way as to make the *theory of optimal transport* appear.

### §1.1. The ground-state energy problem

We consider a system of  $N$  classical pointlike particles located at  $x_1, \dots, x_N \in \mathbb{R}^d$ . These particles interact with each other *via* a two-body interaction potential, which we will denote by  $w(x, x')$ , and which we will always assume hereafter to be symmetric, that is such that  $w(x, x') = w(x', x)$ . In addition, the particles are subjected to an external potential  $V(x)$ . According to the laws of physics, we know that, at equilibrium, these particles will arrange themselves in such a way as to minimise the total energy of the system. Therefore, we seek to solve the following minimisation problem

$$E_{N,w}(V) := \inf_{x_1, \dots, x_N \in \mathbb{R}^d} \left\{ \sum_{1 \leq i < j \leq N} w(x_i, x_j) + \sum_{i=1}^N V(x_i) \right\}. \quad (\text{E})$$

We have put the interaction potential  $w$  in subscript in the notation of the *ground-state energy*  $E_{N,w}$ . The reason for this, other than a simple stylistic convention, is as follows. Remember that the potential  $w$  is typically fixed by the nature of the particles under consideration. Although it can be artificially modified within reasonable limits in the laboratory, it should be remembered that it is usually given as a premise that is beyond the control of the experimenter. As for the external potential  $V$ , it describes the action that the experimenter applies to the system and it can therefore be more easily modified — always within reasonable limits. However, if we imply by our notation that  $V$  can vary, the reason for this is different, as it will soon become apparent.

The **Problem E** is a very general problem, which can be written for molecular or atomic systems as well as for gravitational systems. Although in this section we will remain at a certain level of generality by not specifying the form of the interaction potential  $w$ , in this thesis we will focus on systems of interacting charged particles. For example, we can think of ions interacting with each other through the *Coulomb potential*, as is the case in the study of the core of *white dwarfs* in astrophysics [VH19].

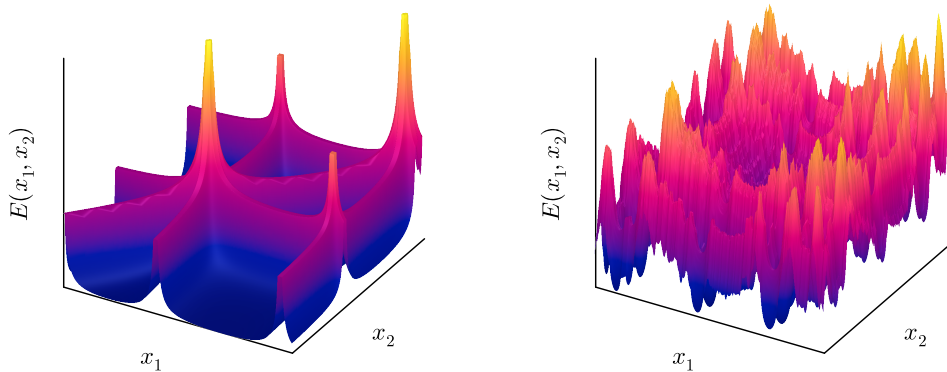


Figure 7: We consider particles  $x_1, \dots, x_N \in \mathbb{R}$  interacting through the potential  $|x - x'|^{-1}$ . On the left, we consider that  $N = 4$  and that the particles  $x_3$  and  $x_4$  are arbitrarily fixed. The surface then depicts the energy  $E(x_1, x_2)$  of the two remaining particles. On the right, we do the same with  $N = 50$ . We notice that the energy  $E(x_1, x_2)$  features many local *minima*.

It would be tempting to try to solve the **Problem E** analytically. At the exception of a few specific and marginal cases, this attempt is certainly doomed to fail. Indeed, the analytical resolution of such a problem is in general impossible without additional information on the nature of the minimisers. If the mathematician may find this frustrating, the physicist will be satisfied with an approximate solution obtained from numerical calculations. However, things are not necessarily simpler numerically. First of all, it should be noted that, in statistical physics at least, the number of particles  $N$  is typically expected to be of the order of the *Avogadro number*. This expectation is immediately dashed by the limitations of our computers. We will therefore have to reduce our expectations by considering a few hundred or thousands of particles. The difficulty of such a problem lies in the well-known fact that in large dimension the total energy that we seek to minimise will generally feature a large number of local *minima*, which are the pet peeve of optimisation algorithms (see Figure 7). Although such a task is by no means impossible, it is not entirely trivial.

We will take a different approach to the **Problem E**. First, we note that  $E_{N,w}(V)$  is a concave function of the external potential, as a minimisation of a linear function. We can then consider the *Legendre transform*<sup>11</sup> of the ground-state energy  $E_{N,w}(V)$  seen as a functional of the external potential  $V$ .

<sup>11</sup>Recall that, given a Banach space  $X$ , the Legendre transform of a function  $F : X \rightarrow \mathbb{R}$  is defined as the function  $F^* : X^* \rightarrow \mathbb{R}$  on the topological dual  $X^*$  of  $X$  endowed with its duality pairing  $\langle \cdot, \cdot \rangle$  by the equality (according to the convention that this transform is convex)

$$F^*(v) = \sup_{u \in X} \{F(u) - \langle u, v \rangle\}.$$

While it may seem at first sight that such an idea is somewhat arbitrary, it should be remembered that this is a fairly common technique in physics — *e.g.* in classical mechanics, where it allows to go from the *Lagrangian formalism* to the *Hamiltonian formalism* and *vice-versa*, or in statistical physics, where the entropy is the Legendre transform of free energy. For a discussion on the Legendre transform in physics, see [ZRM09]. We therefore consider  $F_{N,w}$  the Legendre transform of  $E_{N,w}$ , given by

$$F_{N,w}(\rho) := \sup_{V \in \mathcal{V}} \left\{ E_{N,w}(V) - \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}. \quad (\text{F})$$

Here the *supremum* runs over a functional space  $\mathcal{V}$  which characterises the set of admissible external potentials  $V$ , and  $\rho$  is an element of its topological dual  $\mathcal{V}'$ , typically a measure. For the sake of clarity, we can assume that  $\mathcal{V}$  is the set of continuous and bounded functions, and therefore that its dual  $\mathcal{V}'$  is given by the set of Radon measures. Once the Legendre transform  $F_{N,w}$  of the ground-state energy  $E_{N,w}$  has been defined, we are going to consider the Legendre transform of  $F_{N,w}$  itself. It follows from a classical theorem of convex analysis<sup>12</sup> [Sim11] that under appropriate assumptions the Legendre transform is an involution. Consequently, we obtain

$$E_{N,w}(V) = \sup_{\rho \in \mathcal{V}'} \left\{ F_{N,w}(\rho) - \int_{\mathbb{R}^d} V(r) \rho(r) dr \right\}. \quad (\text{D})$$

We have therefore obtained a new expression of **Problem E**. *Is this problem simpler? A priori*, it seems that this new problem is just as complicated as our original one — if not more. For instance, we went from a minimisation problem over the set of configurations  $(\mathbb{R}^d)^N$  to a variational problem over the much bigger set of measures  $\rho \in \mathcal{V}'$  — which also poses some challenges when it comes to its numerical treatment. However, the **Problem D** has a few advantages. First, it is a convex problem. More interestingly, since the functional  $F_{N,w}$  is independent of the external potential  $V$ , we can imagine that a perfect knowledge of  $F_{N,w}$  could eventually provide us with an algorithm to solve the **Problem D** which would be in some way *universal* with respect to the external potential  $V$ , as the latter only appears as a linear term in **Problem D**. Let us therefore further study the functional  $F_{N,w}$  to gain some insights. In practice, we will then try to approximate  $F_{N,w}$  by a simpler functional.

*Remark.* In the language of optimisation theory and convex analysis, we note that the **Problem D** exactly corresponds to a *duality principle* for the ground-state energy  $E_{N,w}$ . Recall that such a principle states that to each convex minimisation problem, we can associate an equivalent maximisation problem, known as a *dual problem*. By abuse of language, we may say that  $F_{N,w}$  is the *dual* functional of the ground-state energy  $E_{N,w}$ . Incidentally, this is not at all surprising, given that the Legendre transform is exactly the analytical counterpart of the duality principle as defined in optimisation [Sim11].

<sup>12</sup>The said theorem is the *Fenchel-Moreau theorem*, which states that under the assumption that  $F$  is a proper, convex and lower semicontinuous function, then  $(F^*)^* = F$ .

### Determining the functional $F_{N,w}$

Can we better understand the functional  $F_{N,w}$ ? It turns out that we can give a much more explicit formulation of  $F_{N,w}$ , which will lead us very quickly to the heart of our subject, namely the theory of optimal transport. We first point out that, in the definition of the ground-state energy  $E_{N,w}$ , it is always possible to substitute for the minimisation over the space of configurations  $(x_1, \dots, x_N) \in \mathbb{R}^{dN}$  the minimisation over the set of probability measures  $\mathbb{P}(x_1, \dots, x_N)$  on  $\mathbb{R}^{dN}$ . In other words, we are moving to a probabilistic view of our particles — which not only is not surprising from the standpoint of statistical physics, but which will be mandatory in the case where one adds temperature to the system, as we shall see later. Since we have assumed that the interaction potential  $w$  is symmetric, we can always restrict ourselves to symmetric  $\mathbb{P}$ , that is such that  $\mathbb{P}(x_1, \dots, x_N) = \mathbb{P}(x_{\sigma(1)}, \dots, x_{\sigma(N)})$  for any permutation of indices  $\sigma \in \mathfrak{S}_N$ . We then find that

$$E_{N,w}(V) = \inf_{\mathbb{P}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) + \int_{\mathbb{R}^d} V(x) \rho_{\mathbb{P}}(x) \right\}.$$

Here, we have used that the external potential  $V$  is a one-body potential. In particular, the total potential energy of a distribution of particles  $\mathbb{P}$  can be rewritten as

$$\int_{\mathbb{R}^{dN}} \sum_{i=1}^N V(x_i) d\mathbb{P}(x_1, \dots, x_N) = \int_{\mathbb{R}^d} V(x) \rho_{\mathbb{P}}(x) \quad (54)$$

where  $\rho_{\mathbb{P}} \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  is the positive measure defined by

$$\rho_{\mathbb{P}}(x) = N \int_{\mathbb{R}^{d(N-1)}} \mathbb{P}(x, x_2, \dots, x_N)$$

*Remark.* The function  $\rho_{\mathbb{P}}$  is often called the *electronic density* in the case of charged particles such as electrons or ions. We will simply refer to it as the *density*. Note that this function is normalised such that  $\int_{\mathbb{R}^d} \rho_{\mathbb{P}} = N$ . If the particles are distributed according to the probability measure  $\mathbb{P}$ , the function  $\rho_{\mathbb{P}}$  corresponds to the average density of charge in space. We can think of it as a *mean-field* representation, such that a particle located far away from the system will not "see" all the peculiar geometric configurations in which the system may be found, but rather an averaged version corresponding exactly to  $\rho_{\mathbb{P}}$ .

The ground-state energy  $E_{N,w}$  has been rewritten as an *infimum* over the set of measures  $\mathbb{P}$  on the space of  $(\mathbb{R}^d)^N$ . We will now separate this minimisation into two-step minimisation. First, we fix a density  $\rho$ , and we consider the minimisation of the energy over the set of measures  $\mathbb{P}$  that satisfy the constraint  $\rho_{\mathbb{P}} = \rho$ . We then optimise on the density  $\rho$ . We obtain

$$E_{N,w}(V) = \inf_{\rho} \left\{ \int_{\mathbb{R}^d} V(x) \rho(x) + \inf_{\rho_{\mathbb{P}} = \rho} \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) \right\}, \quad (55)$$

where the first *infimum* is taken over the set of positive measures  $\rho$  such that  $\int_{\mathbb{R}^d} \rho = N$ . Such a rewriting echoes with *Lieb's variational problem* and the *Levy-Lieb functional* [Lev79; Lie83; HT22], as we shall see below when we will discuss the *Density functional theory* (DFT). We now insert this rewriting into the definition of  $F_{N,w}$  as the Legendre transform of the energy  $E_{N,w}$  — which is equivalent to the *Hohenberg-Kohn variational problem* in DFT [HT22; HK64]. We can then invert the *supremum* of the Legendre transform and the *infima*, so as to obtain the equality

$$F_{N,w}(\rho) = \inf_{\mathbb{P}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) + \sup_{V \in \mathcal{V}} \int_{\mathbb{R}^d} V(x) (\rho_{\mathbb{P}}(x) - \rho(x)) \right\}.$$

We then notice that the *supremum* over the set of potentials  $V \in \mathcal{V}$  is zero if and only if  $\rho_{\mathbb{P}} = \rho$ , and infinite otherwise. We are therefore implicitly forced to only consider the set of measures  $\mathbb{P}$  whose density is  $\rho$ , that is such that  $\rho_{\mathbb{P}} = \rho$ . We finally get

$$\boxed{F_{N,w}(\rho) = \inf_{\rho_{\mathbb{P}} = \rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) \right\}.} \quad (\text{F})$$

When  $\rho$  is not a positive measure such that  $\int_{\mathbb{R}^d} \rho = N$ , we may use the convention that  $F_{N,w}(\rho) = \infty$ . We have now arrived at a satisfactory expression for the Legendre transform  $F_{N,w}$  of the ground-state energy functional  $E_{N,w}$ . We see that given a density  $\rho$ , *the quantity  $F_{N,w}(\rho)$  corresponds to the minimum interaction energy that can be achieved by a system of  $N$  particles under the constraint that its density is equal to  $\rho$* . We will now be able to make the theory of optimal transport appear — if the informed reader has not already unravelled it.

### The functional $F_{N,w}$ and its link with the optimal transport

We can now state that the functional  $F_{N,w}$ , which we originally defined as the Legendre transform of the ground-state energy  $E_{N,w}(V)$  seen as a functional of the external potential  $V$ , can be expressed — in the case of symmetrical  $w$  interaction potentials at least — as a *multimarginal optimal transport* problem [BDPGG12; CFK13; DMGN15; FGGG22] where the number of marginals is given by the number of particles  $N$ , where all the marginals are constrained to the same density  $\rho$ , and where finally the cost of transport  $c(x_1, \dots, x_N)$  is given by the interaction energy

$$c(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} w(x_i, x_j). \quad (56)$$

Recall that we introduced the **Problem F** and consequently the functional  $F_{N,w}$  to obtain a rewriting of the **Problem E** of the determination of the ground-state energy  $E_{N,w}(V)$ , where the external potential  $V$  is *a priori* fixed. We hoped that this new point of view would provide us with tools for solving this

problem, which is complicated. The Problem F did not seem any simpler than the original problem — but it did have some advantages. Now that we have obtained a more explicit expression for the functional  $F_{N,w}(\rho)$  as a multimarginal optimal transport problem, we can confirm our intuition. Indeed, it is well known that the optimal transport problem is complicated, both theoretically and numerically. However, from a physical standpoint, we may try to come up with good approximations of the functional  $F_{N,w}$ . Incidentally, this is at the heart of the *density functional theory* (DFT). This important theory can be written both for classical and quantum systems. In the following section, we briefly introduce DFT in the quantum case, and return to the classical case a little later on.

### §1.2. Density functional theory in quantum chemistry

*Density Functional Theory* (DFT) is a very popular computational modelling method in quantum chemistry. We explain hereafter the main idea of this theory, without going into the technical details, as it requires the mathematical formalism of quantum mechanics [Lew22b]. In this section, we consider the *quantum* version of DFT, and we will explain its classical counterpart later on once we introduce temperature to our initial Problem E.

We consider a system of  $N$  quantum particles interacting *via* a two-body interaction potential  $w$  and subjected to an external potential  $V$ . This time, the energy of the system is given by the quadratic form

$$\begin{aligned} Q_{N,w,V}(\Psi) &= \hbar^2 \int_{\mathbb{R}^{dN}} \sum_{i=1}^N |\nabla_{x_i} \Psi(x_1, \dots, x_N)|^2 \\ &+ \int_{\mathbb{R}^{dN}} \sum_{i=1}^N V(x_i) |\Psi|^2(x_1, \dots, x_N) + \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) |\Psi|^2(x_1, \dots, x_N), \end{aligned} \quad (57)$$

for any *wavefunction*  $\Psi$ . Recall that  $\Psi$  is by definition a square-integrable function  $\Psi \in L^2(\mathbb{R}^{dN})$  normalised such that  $\int_{\mathbb{R}^{dN}} |\Psi|^2 = 1$ , and which also satisfies a symmetry constraint such that the function  $|\Psi|^2$  is invariant with respect to the permutation of the indices of the particles. The normalisation ensures that  $|\Psi|^2$  is a probability density on the space  $(\mathbb{R}^d)^N$ , and it is equivalent to our previous  $\mathbb{P}$ . The first term in (57) corresponds to the kinetic energy. Note that this term cannot be expressed directly as a function of  $\mathbb{P}$ , at the exception of the special case where the sign of  $\Psi$  is constant. The second (resp. third) term corresponds to the potential (resp. interaction) energy. The constant  $\hbar$  is the *Planck constant*. The problem of determining the ground-state energy, that is Problem E, that we introduced at the very beginning of this introduction now reads in its quantum version [Lew22b, Chap. 1] as

$$E_{N,w}(V) := \inf_{\substack{\Psi \in L^2(\mathbb{R}^{dN}) \\ \int_{\mathbb{R}^{dN}} |\Psi|^2 = 1}} Q_{N,w,V}(\Psi). \quad (58)$$

This variational problem is equivalent to determining the smallest eigenvalue  $E = E_{N,w}(V)$  (and its corresponding eigenvector  $\Psi$ ) of the associated Schrödinger operator,

$$\left( -\hbar^2 \sum_{i=1}^N \Delta_{x_i} + \sum_{i=1}^N V(x_i) + \sum_{1 \leq i < j \leq N} w(x_i, x_j) \right) \Psi = E\Psi. \quad (59)$$

As in the classical case, finding an analytical solution to this problem is impossible, with the exception of a few textbook examples such as the hydrogen atom [Lew22b, Sec. 1.3]. As for solving it numerically, the story repeats itself for the worse as we find ourselves with an  $L^2$  space which needs to be discretized. The computational burden is then typically exponential in the number of particles and the size of the discretisation, which is dramatic. The main idea behind DFT, which we have already encountered, is to separate the minimisation over the set of wavefunctions by a two-stage minimisation. The first one is carried over the set of densities  $\rho$ , and the second over the set of wavefunctions such that the associated particle distribution  $\mathbb{P} = |\Psi|^2$  verifies  $\rho_{\mathbb{P}} = \rho$ . We then find

$$E_{N,w}(V) = \inf_{\rho} \left\{ F_h(\rho) - \int_{\mathbb{R}^d} V(r)\rho(r)dr \right\} \quad (60)$$

where the *Levy-Lieb functional*  $F_h$  [Lev79; Lie83; HT22] is defined by

$$F_h(\rho) := \inf_{\rho_{|\Psi|^2} = \rho} \left\{ \hbar^2 \int_{\mathbb{R}^{dN}} \sum_{i=1}^N |\nabla_{x_i} \Psi|^2 + \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) |\Psi|^2(r_1, \dots, r_N) \right\}. \quad (61)$$

We have thus expressed the ground-state energy  $E_{N,w}(V)$  as the solution to a variational problem on the set of densities  $\rho \in L^1(\mathbb{R}^d)$ , which is much smaller than that of the wavefunctions  $\Psi$ . This formulation is precisely the starting point of the density functional theory.

*Remark.* If we are deliberately abusing of the lack of mathematical rigour, a small remark is in order here. In the classical case, the set of densities  $\rho$  was simply the set of positive measures such that  $\int_{\mathbb{R}^d} \rho = N$ . In the quantum case, we have to be a little more careful. Indeed, we are working with wavefunctions  $\Psi$  that typically belong to Sobolev spaces to ensure that the first term in (57), namely the kinetic energy, is well-defined and finite. In particular, the set of densities  $\rho$  which are said to be *representable*, that is such that there exists a wavefunction  $\Psi$  for which  $\rho = \rho_{|\Psi|^2}$ , is non-trivial. It is known from [Lie83, Thm. 1.1] that this set is given by  $\{\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+) : \sqrt{\rho} \in L^1(\mathbb{R}^d)\}$  where  $H^1(\mathbb{R}^d)$  is the Sobolev space of functions  $u \in L^2(\mathbb{R}^d)$  such that their weak derivatives are also square-integrable, that is  $u' \in L^2(\mathbb{R}^d)$ .

We see that  $F_h(\rho)$  corresponds to the minimum kinetic energy *plus* interaction energy that can be reached by a quantum state  $\Psi$  under the constraint that  $\rho_{|\Psi|^2} = \rho$ . In particular, in the semiclassical limit  $\hbar \rightarrow 0$ , we expect that the functional  $F_h$  will coincide with the functional  $F_{N,w}$  defined previously in the classical setting. This is not surprising, since  $F_h$  corresponds exactly to the



quantum counterpart of  $F_{N,w}$ . A rigorous proof of this limit can be found in [CFK13; Lew18; BDP17; CFK18].

In addition, as with  $F_{N,w}$ , the functional  $F_h$  generically has no analytical expression, and it is just as difficult to approximate numerically. It will therefore be necessary to craft good approximations to it, and this is where DFT comes into play.

We see that the functional  $F_{N,w}$ , which we introduced in a purely classical framework, also appears in the context of a concrete set of numerical methods in quantum chemistry. In practice, the functional  $F_{N,w}$  is used as an *exchange-correlation functional* within the *Kohn–Sham* (KS; [KS65]) formalism. This very popular formalism consists in constructing a fictitious non-interacting quantum system that is equivalent — in a certain sense — to the original system whose ground-state energy we are trying to compute. In other words, this formalism corresponds to considering the limit  $\hbar \rightarrow \infty$  in the Levy–Lieb functional  $F_h(\rho)$ . On the contrary, we mentioned that  $F_{N,w}$  appeared in the semiclassical limit  $\hbar \rightarrow 0$ . This formalism is called *Strictly–Correlated Electrons* (SCE; [SPL99]) in DFT, and corresponds, in contrast to Kohn–Sham, to constructing a fictitious quantum system with no kinetic energy which is equivalent to the original system. In this system, the electrons are strongly correlated and can be considered to be classical particles. In a series of works [MGG12; PLK13; Mal+14; MMGG14; GVGG18; Gro+20; Mar+22], the functional  $F_{N,w}$  is used as an efficient correction term in *Kohn–Sham* for strongly correlated quantum systems, such as *Mott insulators* or crystallized systems.

### §1.3. The free energy at positive temperature

After this short excursion into the quantum world, let us return to our classical problem. So far, our systems have been defined without any temperature. Of course, the zero-temperature case is only an ideal problem, as it is not reachable except asymptotically.

We can rewrite the Problem E of the determination of the equilibrium of a system at positive temperature  $T > 0$  by adding, according to the mathematical formalism of statistical mechanics, the Boltzmann entropy into the problem. This leads to

$$E_{N,w,T}(V) := \inf_{\mathbb{P}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) - T\mathcal{S}(\mathbb{P}) \right\}. \quad (E_T)$$

Here, we have denoted  $\mathcal{S}(\mathbb{P})$  the entropy of the measure  $\mathbb{P}$ , defined for any measure which is absolutely continuous with respect to the Lebesgue measure as

$$\mathcal{S}(\mathbb{P}) = - \int_{\mathbb{R}^{dN}} \ln(\mathbb{P}) d\mathbb{P}. \quad (62)$$

The parameter  $T$  corresponds to the temperature<sup>13</sup>. Following the usual notation, we will write  $\beta$  the inverse temperature, *i.e.*  $\beta = T^{-1}$ . We can now

<sup>13</sup>Note the absence of *Boltzmann’s constant*  $k_B$  as a prefactor. The latter is more a historical convention than a real physical necessity



reiterate what has been done previously for Problem E. The *free energy*  $E_{N,w,T}$  at positive temperature then reads

$$E_{N,w,T}(V) = \sup_{\rho \in \mathcal{Y}'} \left\{ F_{N,w,T}(\rho) - \int_{\mathbb{R}^d} V(x) \rho(x) \right\} \quad (D_T)$$

where, as before,  $F_{N,w,T}$  is the Legendre transform of the energy  $E_{N,w,T}(V)$  seen as a functional of the external potential  $V$ . We then find that  $F_{N,w,T}$  can be rewritten as

$$F_{N,w,T}(\rho) = \inf_{\mathbb{P}=\rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) - T\mathcal{S}(\mathbb{P}) \right\}. \quad (F_T)$$

We see that the Problem  $F_T$  is similar to the Problem F, except for the addition of the entropy. In the language of optimal transport, this problem is nothing but an *entropic optimal transport* problem [Lé12; Ben+15]. While in our case this entropy plays a physical role, by representing the thermal fluctuations in the system, in the usual framework of entropic optimal transport, it is rather used to regularise the original transport problem and  $T$  is seen as a penalty.

It follows from a classical exercise in calculus of variations that the minimiser of the Problem  $F_T$ , which is unique by strict convexity of the entropy, is given by the *Gibbs–Boltzmann measure*  $\mathbb{P}_\beta$  whose density is defined by

$$\mathbb{P}_\beta(x_1, \dots, x_N) = \frac{1}{Z_\beta} \exp \left( -\beta \sum_{1 \leq i < j \leq N} w(x_i, x_j) - \beta \sum_{i=1}^N V_\beta(x_i) \right). \quad (63)$$

Here,  $Z_\beta$  is a normalising constant that ensures that  $\mathbb{P}_\beta$  is a probability measure on  $(\mathbb{R}^d)^N$ , and  $V_\beta$  is the external potential which ensures that the marginal constraint  $\rho_{\mathbb{P}_\beta} = \rho$  is satisfied. In the context of statistical mechanics, the measure  $\mathbb{P}_\beta$  is called the *canonical ensemble* [Rue99]. It follows that to solve Problem  $F_T$  it suffices to find the potential  $V_\beta$  such that the Gibbs measure  $\mathbb{P}_\beta$  satisfies the marginal constraint. In fact, this potential verifies a nonlinear equation, which we will introduce later in (75).

Such an inverse problem, which consists in finding the potential  $V$  that gives rise to the density  $\rho$ , corresponds to *Density Functional Theory*, as introduced before, in its *classical* version. We refer to the references [Eva79; Eva92; Sin91] or [Wu09] for a more recent overview of the subject. Classical DFT is a popular method for studying classical inhomogeneous fluids, such as liquids for which the density  $\rho$  is not constant in space. This is for instance very useful when studying the interface of a phase separation [Gel+99]. As with quantum DFT, the idea of classical DFT is to construct approximate functionals for  $F_{N,w,T}$  [ESS76; EP79; SE77]. As for its theoretical justification, the classic reference is a work by Chayes, Chayes and Lieb in [CCL84] — see also [JLM23] for a more recent work. It is interesting to note that the paper of Chayes *et al.* [CCL84], to which we will often refer, essentially contains what we would now call the *Kantorovich duality* for the (entropic) optimal transport — at least for costs arising in statistical physics — even though the terminology used is not the same.

Perhaps some of our readers are finally reassured to hear us mention the name of Leonid Kantorovich and the duality that bears his name. It is true that up until now, we did not mention his name. After all, we now understand that when we defined the functional  $F_{N,w}$  as the Legendre transform of the ground-state energy  $E_{N,w}$ , that is

$$F_{N,w}(\rho) = \sup_{V \in \mathcal{V}} \left\{ E_{N,w}(V) - \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}, \quad (64)$$

we were simply writing the *Kantorovich duality* for the optimal transport problem  $F_{N,w}(\rho)$ . However, it would have been strange to do so, following our didactic. Indeed, we somehow went backward by starting from the dual, that is the ground-state energy functional  $E_{N,w}(V)$ , which is the most relevant physically, to arrive at what we would call the *primal* of the optimal transport problem, that is the functional  $F_{N,w}$ . In the context of physics, this terminology should be inverted — the dual being the “primal” and *vice-versa*.

### Summary

Let us retrace what has been said so far, at the cost of repetition. We started from the problem of determining the ground-state energy of system  $E_{N,w}(V)$ , that is **Problem E**. We emphasized that this problem was complicated, both theoretically and numerically. By resorting to the Legendre transform  $F_{N,w}$  of  $E_{N,w}$ , we were able to obtain another expression of this problem, namely **Problem F**. We showed that this problem was a multimarginal optimal transport problem. Although it is *a priori* as complicated as the original one, we pointed out that the functional  $F_{N,w}$  was independent of the external potential  $V$ , and that it was not excluded to find good approximations to it. We explained that such work was at the heart of density functional theory, a very important modelling method in quantum chemistry.

## §2. Multimarginal optimal transport and Kantorovich duality

We have previously explained how optimal transport, in its multimarginal setting, naturally occurs in statistical physics or in quantum chemistry as the dual of the problem of the determination of the ground-state energy of a system. In this section, we briefly review the multimarginal formalism, focusing on the physical interpretation of the Kantorovich duality.

### §2.1. Abstract formulation of multimarginal optimal transport

Multimarginal optimal transport corresponds to the natural extension to several marginal of the classical two-marginal optimal transport. For an introduction to the theory of optimal transport, we refer to the references [Vil09; San15].

In general, the multimarginal problem can be defined as follows. Given  $N$  probability measures  $\mu_1, \dots, \mu_N$  on  $\mathbb{R}^d$  and a *cost of transportation*  $c : (\mathbb{R}^d)^N \rightarrow \mathbb{R}$ , we define the *Kantorovich formulation* of the multimarginal problem as

$$\inf_{\pi \in \Pi(\mu_1, \dots, \mu_N)} \int_{\mathbb{R}^{dN}} c(x_1, \dots, x_N) d\pi(x_1, \dots, x_N) \quad (\text{MOT})$$

where  $\Pi(\mu_1, \dots, \mu_N)$  denotes the set of *transport plans*, that is the set of measures on the Cartesian product  $\mathbb{R}^d \times \dots \times \mathbb{R}^d$  such that the  $i$ -th marginal of  $\pi$  — according to the  $i$ -th copy of  $\mathbb{R}^d$  of the Cartesian product — is given by  $\mu_i$ .

We note that the existence of a minimiser to the **Problem MOT** follows, as for the classical two-marginal case, from a straightforward argument of compactness under rather weak assumptions on  $c$ , typically that  $c$  is a lower semicontinuous and bounded from below function [Vil09, Thm. 5.10]. In particular, for the costs of transportation arising in physics, as in the case of the **Problem F** derived in the previous section, that is

$$c(r_1, \dots, r_N) = \sum_{1 \leq i < j \leq N} w(x_i, x_j), \quad (65)$$

we immediately have obtain the existence of an optimal transport plan under the assumption that  $w$  is lower semicontinuous and bounded from below. This is for example the case of the *Coulomb potential* in three space-dimension, that is  $w(x, x') = |x - x'|^{-1}$ . When the cost is not bounded from below, for example when  $w$  is the Coulomb potential in two space-dimension, that is  $w(x, x') = -\ln|x - x'|$ , the existence of a minimiser is easily obtained under rather mild additional assumption, for instance that there exists a measurable function  $b$  such that  $w(x, x') \leq b(x) + b(x')$  and such that  $\int_{\mathbb{R}^d} |b| \rho < \infty$ , see [Vil09, Thm. 5.10]. In particular, for the Coulomb potential in dimension  $d = 2$ , it suffices that  $\int_{\mathbb{R}^2} |\ln(x)| \rho < \infty$ .

As for the optimal transport with two marginals, we can also define the *Monge formulation* to the multimarginal problem — which we will recall to be the historical formulation of the optimal transport. In the multimarginal case, this formulation reads

$$\inf_{T_i^\# \mu_1 = \mu_i} \int_{\mathbb{R}^{dN}} c(x, T_2(x), \dots, T_N(x)) d\mu_1(x) \quad (\text{Monge})$$

where the *infimum* runs over the set of measurable maps  $T_i$  such that  $T_i^\# \mu_1 = \mu_i$  for all  $i = 2, \dots, N$  — where according to the usual notation  $f^\# \mu$  is the *pushforward measure*, which is defined for any measurable set  $E$  and any measurable function  $f$  as  $f^\# \mu(E) := \mu(f^{-1}(E))$ . This problem is very difficult to handle, even more so than its two-marginal counterpart. We note that many theoretical results on **Problem Monge** for two marginals are obtained using to tenacious link between it and the *Monge–Ampère equation* [Fig18; Fig17; DPF14]. This analogy no longer holds in the multimarginal setting — or at least, if such an equation can be formulated, it simply seems to be far too complicated to provide any interesting insights on the problem.

An important open problem is to determine whether or not there always exists an optimal transport plan  $\mathbb{P}$  for **Problem F** which is a solution of **Problem Monge**, that is such that there exist maps  $T_2, \dots, T_N$  which preserve the density  $\rho$  and such that the transport plan  $\mathbb{P}$  is supported on the graphs of the  $T_i$ 's, that is

$$\mathbb{P}(x_1, \dots, x_N) = \frac{\rho(x_1)}{N} \otimes \delta(x_2 - T_2(x_1)) \otimes \dots \otimes \delta(x_N - T_N(x_1)). \quad (66)$$

In the case of **Problem F**, since all the marginal are equal, we can show that the problem is equivalent to finding a unique application  $T$  preserving the density  $\rho$  such that  $T_i = T^{(i)}$ , where  $T^{(i)}$  denotes the  $i$ -th fold composition of  $T$  with itself, and such that  $T^{(N)}$  is the identity mapping [CDM15]. In the case  $N = 2$ , the answer is positive for the Coulomb cost [BDPGG12; CFK13], see also [DMGN15, Thm. 3.6]. This result follows from a very general theorem of optimal transport, which states that, under the assumption that  $c$  verifies the *twist condition* [San15, Def. 1. 16], the minimiser of **Problem MOT** is unique and of the form (66). This condition is verified for the Coulomb cost or more generally if  $w(x, x') = \tilde{w}(|x - x'|)$  where  $\tilde{w}$  is a strictly convex function. A similar result was proved in the multimarginal setting by Kim and Pass [KP14]. Nevertheless, the assumptions of this theorem are difficult to verify in practice, and the problem remains largely open for an arbitrary number of particles  $N$  in dimension  $d \geq 2$ . In dimension  $d = 1$ , the problem was entirely solved by Colombo, De Pascale and Di Marino [CDPDM15] in the case of convex interaction potentials. They also gave an explicit construction of the transport map  $T$ . We finally note that, while the existence of a minimiser to **Problem Monge** is still largely unknown, it is known that the *infimum* is equal to the minimum of the **Problem MOT**. This follows from the seminal result obtained by Pratelli [Pra07] which, although originally proved in the two-marginal case, extends easily to the multimarginal case, see [CDM15, Rem. 1.3]. The stronger statement that the Monge maps can be taken *cyclical*, that is stemming from a unique application  $T$  and its compositions with itself as explained above, was proved by Colombo and Di Marino [CDM15].

We will not go into further details on the abstract theory of the multimarginal optimal transport problem. We refer to [DMGN15; Pas15].

## §2.2. Kantorovich duality and its physical interpretation

As for any optimal transport problem, multimarginal optimal transport admits a dual formulation according to the famous *Kantorovich duality*. For an abstract multimarginal optimal transport problem as defined above in **Problem MOT**, the Kantorovich dual reads

$$\sup_{V_1, \dots, V_N} \left\{ E(V_1, \dots, V_N) + \sum_{i=1}^N \int_{\mathbb{R}^d} V_i(x) \mu_i(dx) \right\} \quad (\text{MOT}_D)$$

where the *supremum* runs over the set of continuous functions  $V_i : \mathbb{R}^d \rightarrow \mathbb{R}$  for  $i = 1, \dots, N$ , and where

$$E(V_1, \dots, V_N) := \inf_{x_1, \dots, x_N} \left\{ c(x_1, \dots, x_N) - \sum_{i=1}^N V_i(x_i) \right\}. \quad (67)$$

We used here a formulation which is not *a priori* the most usual, see [PC19, Rem. 2.23]. Indeed, it is usually required that the  $V_i$ 's satisfy the following constraint

$$V_1(x_1) + \dots + V_N(x_N) \leq c(x_1, \dots, x_N) \quad (68)$$

where the inequality is either required for all configurations, or only  $\mu_1 \otimes \dots \otimes \mu_N$ -almost-everywhere. Under appropriate assumptions, these two constraints are in fact equivalent. Our formulation of the **Problem MOT<sub>D</sub>** is equivalent to asking that this constraint be verified up to an additive constant. This constant is exactly given by the quantity (67). In particular, the constraint (68) is equivalent to asking  $E(V_1, \dots, V_N) = 0$ . The advantage of this formulation is that the Kantorovich potentials are only defined up to additive constants, which is very natural from the standpoint of physics. From the point of view of the numerics, it allows to get rid of the constraint (68) which is not the easiest to handle.

In **Problem F**, all the marginal are equal  $\mu_1 = \dots = \mu_N$  and given by  $\rho/N$ . Remember that, by convention, we have  $\int_{\mathbb{R}^d} \rho = N$ . By considering the arithmetic mean of the  $V_i$ 's, we can rewrite the Kantorovich dual as a variational problem of a single function  $V$ . We then find

$$F_{N,w}(\rho) = \sup_{V \in \mathcal{V}} \left\{ E_N(V) - \int_{\mathbb{R}^d} V(x) \rho(x) \right\}, \quad (69)$$

that is the Legendre transform of the ground-state energy  $E_{N,w}(V)$ . The equality of the two quantities, the so-called *strong duality*, and the existence of a function  $V$  that maximises the *supremum* — which we call a *Kantorovich potential* according to the usual terminology — follows from the results of [CDMS19; BCDP18]. We note that here we have switched the sign of the potential compared to **Problem MOT<sub>D</sub>**. The reason for this change of sign, which is harmless from a mathematical point of view, follows from a physical consideration, as we shall now see.

*What is the physical interpretation of the dual problem?* Let us appeal to the theory of duality in the context of optimal transport. It is known that at optimality [San15], if we consider an optimal transport plan  $\mathbb{P}$  and a Kantorovich potential  $V$ , we must have

$$\sum_{1 \leq i < j \leq N} w(x_i, x_j) + \sum_{i=1}^N V(x_i) = E_N(V) \quad \text{on the support of } \mathbb{P}, \quad (70)$$

and where the equality becomes an inequality when considered over the whole space. This equality says exactly that the configurations  $(r_1, \dots, r_N) \in \text{supp}(\mathbb{P})$  minimise the total energy of the system. From a physical point of view, it is

interesting to differentiate this equation. Under appropriate assumptions on the regularity on  $V$  and  $w$ , we then obtain by differentiating the previous equation with respect to the first particle  $x_1$  that

$$-\nabla_{x_1} V(x_1) = \sum_{i=2}^N \nabla_{x_1} w(x_1, x_i). \quad (71)$$

This equality is nothing but an equilibrium equation from classical mechanics. It says that at optimality the force exerted on the first particle  $x_1$  due to the external potential  $V$  must cancel out the interaction force exerted by the other particles  $x_2, \dots, x_N$ . We therefore obtain a physical characterisation of the Kantorovich potential as *the external potential which forces the particles to live on the support of an optimal transport plan at equilibrium*.

*Example.* Let us consider the example of a two-particle system  $N = 2$  in dimension  $d = 2$ . We suppose that the particles interact *via* the Coulomb potential  $-\ln|r - r'|$ . The target density  $\rho$  is taken to be the uniform measure on the unit disc  $D$ , normalised such that  $\int_D \rho = 2$ . Since this density is radial, we can appeal to the results of Pass in [Pas13]. We then know that the optimal transport plan is unique and given by

$$\mathbb{P}(x, x') = \frac{\rho(x)}{2} \otimes \delta(x' - T(x)) \quad \text{where } T(x) = -\frac{x}{|x|} \sqrt{1 - |x|^2}. \quad (72)$$

The Kantorovich potential  $V$  is then implicitly determined by the equilibrium equation (18) — see Figure 8. If  $V$  can be approximated numerically by integrating this equilibrium equation, in our specific case we can calculate  $V$  explicitly. We find that it is given on  $D$  by

$$V(x) = -\frac{1}{2} \sin^{-1}(x) - \frac{1}{2} \ln \left( 1 + \frac{|x|}{\sqrt{1 - |x|^2}} \right) - \frac{1}{4} \ln(1 - |x|^2). \quad (73)$$

### §2.3. Entropic optimal transport

We recall that the dual problem of the determination of the free energy  $E_{N,T}(V)$  at positive temperature  $T > 0$  reads

$$F_{N,w,T}(\rho) = \inf_{\rho_{\mathbb{P}} = \rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w(x_i, x_j) d\mathbb{P}(x_1, \dots, x_N) - T \mathcal{S}(\mathbb{P}) \right\}, \quad (\text{F}_T)$$

where  $\mathcal{S}(\mathbb{P})$  is the entropy of the  $\mathbb{P}$  measure with respect to the Lebesgue measure. We noticed earlier that this problem was an instance of entropic optimal transport [L  12; Ben+15]. We recall that the minimiser of Problem  $\text{F}_T$ , which is unique by strict convexity of entropy, is given by the Gibbs measure  $\mathbb{P}_\beta$  whose density is

$$\mathbb{P}_\beta(x_1, \dots, x_N) = \frac{1}{Z_\beta} \exp \left( -\beta \sum_{1 \leq i < j \leq N} w(x_i, x_j) - \beta \sum_{i=1}^N V_\beta(x_i) \right) \quad (74)$$

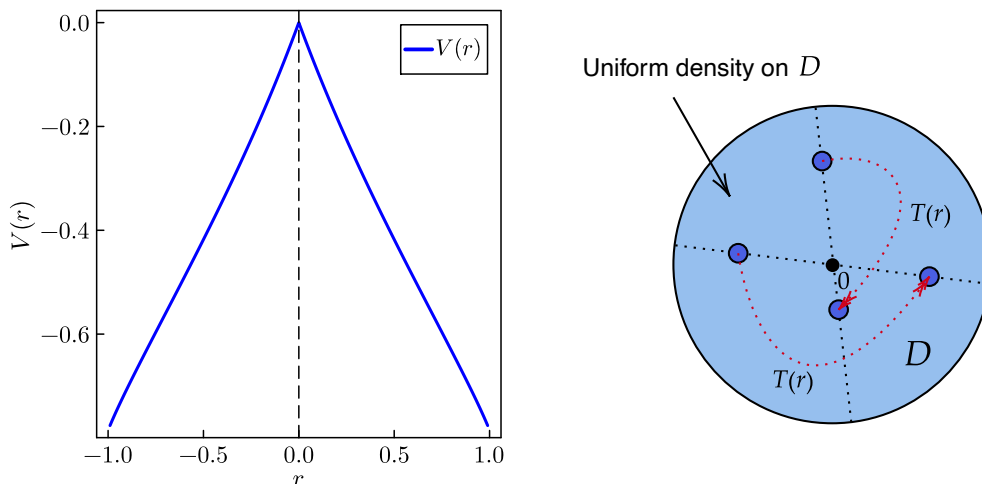


Figure 8: We consider two particles interacting through the logarithmic potential  $-\ln|x - x'|$  in dimension  $d = 2$ . We fix  $\rho$  to be the uniform density of the unit disk  $D$ . The optimal transport plan is then unique and supported on the graph of the map  $T$  as defined in (72). The Kantorovich potential  $V$  (73) is defined by integrating (18).

and where  $V_\beta$  is the unique potential such that  $\mathbb{P}_\beta$  satisfies the marginal constraint  $\rho_{\mathbb{P}_\beta} = \rho$ . We also note that this constraint forces the equality  $V_\beta = +\infty$  outside the support of  $\rho$ . Indeed, in the absence of such a potential barrier, the particles may exit outside  $\Omega$ . Consequently the measure  $\mathbb{P}_\beta$  would charge an open  $U$  such that  $U \cap \Omega^c \neq \emptyset$ . The density  $\rho_{\mathbb{P}_\beta}$  would then be supported outside  $\Omega$ , which would contradict the marginal constraint.

Using that  $V_\beta$  forces the marginal constraint for the Gibbs measure  $\mathbb{P}_\beta$ , it follows from the definition of  $\rho_{\mathbb{P}_\beta}$  that the potential  $V_\beta$  verifies a fixed-point equation given by

$$\begin{aligned} V_\beta(x) + \frac{\beta^{-1}}{N} \log \rho(x) \\ = \beta^{-1} \log \int_{\mathbb{R}^{d(N-1)}} \exp \left( -\beta c(x, x_2, \dots, x_N) - \beta \sum_{i=2}^N V_\beta(x_i) \right) dx_2 \dots dx_N \end{aligned} \quad (75)$$

for all  $x$  in the support  $\Omega$  of  $\rho$  — and *a priori* such that  $\rho(x) > 0$ . This fixed-point equation is sometimes called *Schrödinger equation* in the optimal transport community<sup>14</sup>. We will not use this name so as not to create confusion with the *other* Schrödinger equation, that of quantum physics. Similarly, the potential  $V_\beta$  which is the solution to the fixed-point equation is sometimes called the

<sup>14</sup>This rather unfortunate name comes from [Sch32], where E. Schrödinger introduced the following problem: *Given  $N$  particles evolving according to the brownian motion, which are observed to be distributed according to the measure  $\mu_0$  at time  $t = 0$  and according to another measure  $\mu_1$  at time  $t = 1$ , what can be said about the behaviour of the system between these two times?* This problem turns out to be equivalent to entropy-optimal transport [Lé12; Lé13].



*Schrödinger potential.* Once again, we will not use this terminology, preferring that of the Kantorovich potential, as in the case of the zero temperature.

By noting  $W_\beta := V_\beta + \beta^{-1} \log \rho$ , the previous equation can be rewritten as follows

$$W_\beta(x) = \beta^{-1} \log \int_{\mathbb{R}^{d(N-1)}} \exp \left( -\beta c(x, x_2, \dots, x_N) - \beta \sum_{i=2}^N W_\beta(x_i) \right) d\mu^{\otimes N-1}$$

where  $\mu = \rho/N$ . While the potential  $V_\beta$  pertains to physics, the function  $W_\beta$  is a just a convenient mathematical rewriting. For example, as we have just recalled, the potential  $V_\beta$  must be infinite outside the support of  $\rho$ . This constraint is automatically verified for  $W_\beta$  thanks to the term  $-\beta^{-1} \log \rho$ . Moreover, as noted in [Lie83, Rem. p. 5], the decomposition  $V_\beta = -\beta^{-1} \log \rho + W_\beta$  is also useful because it allows to deduct the case of the *ideal gas*, for which  $W_\beta = 0$ . We also note that in the context of entropic optimal transport, the *relative entropy* with respect to a reference measure  $\nu$  rather than the entropy  $\mathcal{S}(\mathbb{P})$  with respect to the Lebesgue measure on  $\mathbb{R}^d$ , as we have done. The reference measure  $\nu$  is usually considered to be the tensor product of the marginals  $\nu = \mu_1 \otimes \dots \otimes \mu_N$ . This choice does not matter if the  $\mu_i$  are absolutely continuous with respect to the Lebesgue measure, for at optimality the measure  $\mathbb{P}_\beta$  will necessarily be absolutely continuous with respect to the  $\nu$  product measure. The function  $W_\beta$  corresponds in fact to the solution of the fixed-point equation that we would have obtained by considering the relative entropy with respect to the measure  $(\rho/N)^{\otimes N}$  rather than the entropy with respect to the Lebesgue measure.

### §3. Dual charge method (Chapter 1)

In this section we describe the results obtained in Chapter 1. The multimarginal optimal transport problem is not an easy problem to solve numerically, especially for too many marginals  $N$ . Its computational complexity is typically exponential in the number of margins  $N$  and the size of the discretisation. It is in fact known that this problem is NP-hard from repulsive costs [ABA21]. We refer to [Vuc+23] for a review on the numerical methods to solve **Problem F**.

From the point of view of optimisation theory, the zero temperature problem is a *Linear Program*. It involves minimising a linear function under convex constraints. While there is a whole literature of algorithms for solving this type of problem, it remains that the numerical complexity becomes very quickly prohibitive, even for just a dozen of particles. Therefore, we need to be more clever. We can mention a series of recent works [FSV22; Vö21; FV18; FP23; EN23], which use the fact that the optimal transport plan is believed to be sparse — at least in a discrete setting. Another strategy is to use entropic regularisation and make use of the *Sinkhorn algorithm* [BCN16; Ben+15; Cut13]. However, this method does not scale easily when  $N \gg 1$ , as the memory cost required for the tensors quickly becomes prohibitive. We can also mention the works [Alf+20; ACE22; EN23] where the authors relax the marginal constraint and replace it with a set of moments constraints. More precisely, the marginal



constraint  $\rho_{\mathbb{P}} = \rho$  is replaced by

$$\int_{\mathbb{R}^{dN}} \sum_{i=1}^N \phi_j(x_i) d\mathbb{P}(x_1, \dots, x_N) = \int_{\mathbb{R}^d} \phi_j(x) \rho(x) \quad (76)$$

for a finite set of functions  $\phi_j$  ( $j = 1, \dots, M$ ). Our method, which we will now present, is quite similar to these moments constraints. More precisely, it can be seen as the dual of this method.

In what follows, we consider exclusively the case of the multimarginal optimal transport with the *Coulomb cost*, that is where the interaction potential  $w$  is given by  $w(x, x') = g_d(|x - x'|)$  where  $g_d$  is the Coulomb potential in dimension  $d \geq 1$ . Recall that  $g_d$  is defined as the fundamental solution of the Laplacian operator  $-\Delta$ , that is  $-\Delta g_d = \delta_0$  where  $\delta_0$  is the Dirac measure at the origin. The function  $g_d$  is given up to a multiplicative constant by

$$g_d(x) = \begin{cases} -|x| & \text{for } d = 1, \\ -\ln|x| & \text{for } d = 2, \\ \frac{1}{|x|^{d-2}} & \text{for } d = 3. \end{cases} \quad (77)$$

Our idea is as follows. Given a density  $\rho$ , we explained in the previous section that the Kantorovich potential associated with the optimal transport problem should be interpreted as the physical potential that forces the particles to position themselves on an optimal transport plan at equilibrium. Since the problem is electrostatic in nature, we ask ourselves whether it is possible to rewrite the potential  $V$  as an *electrostatic potential* generated by a charge distribution  $\rho^{(e)}$  on  $\mathbb{R}^d$ . Mathematically, this amounts to writing

$$V(x) = \int_{\mathbb{R}^d} g_d(x - x') \rho^{(e)}(x'), \quad (78)$$

which can also be written as  $V = g_d * \rho^{(e)}$  where  $*$  denotes the usual convolution of functions and measures on  $\mathbb{R}^d$ . In the following, we call  $\rho^{(e)}$  the *dual charge*. If this is the case, it would be interesting to try and solve the dual numerically by discretising  $\rho^{(e)}$  rather than the potential  $V$  itself.

*What properties can we expect from the dual charge?* The dual charge should attract the particles and force them into an optimal transport plan. If we consider that our particles are negatively charged, then we expect the dual charge to be a positive measure. Furthermore, if we denote  $\Sigma$  the support of the target density  $\rho$ , we expect the charge  $\rho^{(e)}$  to be supported in  $\Sigma$ . If this were not the case, a particle could be attracted away from the support of  $\rho$ , which would contradict our interpretation of the Kantorovich potential. Finally, according to the equilibrium equation at optimality, which we recall to read

$$-\nabla_{x_1} V(x_1) = \sum_{i=2}^N \nabla_{x_1} w(x_1, x_i), \quad (79)$$

we see that the electric field  $-\nabla V$  induced by the Kantorovich potential on a particle located at a point  $x_1$  should counterbalance the repulsive force

exerted on this same particle by the  $N - 1$  others particles. Consequently, we are led to believe that the total mass of this positive measure must verify  $\int_{\mathbb{R}^d} \rho^{(e)} \geq N - 1$ . Indeed, if the mass were to be strictly less than  $N - 1$ , the electric field  $-\nabla V$  should not be able to properly screen the repulsive force generated by the particles, and consequently the system could not be stable. Turning this intuition into a rigorous proof is however not so straightforward since the equality (79) only holds on the support of an (unknown) optimal transport plan.

The idea of using the dual charge as a variable has already been used numerically in the context of optimal transport by Mendl and Lin [ML13]. We can also mention, in the general framework of DFT, the works [VLGG17], or [Gö99] which seems to be the historical reference. We also refer to the references at the end of the next section. In [ML13], the authors write the Kantorovich potential  $V$  as an electrostatic potential generated by an external charge, which they call *pseudo-charge*, and which is equivalent to our dual charge  $\rho^{(e)}$ . Then, they discretise the dual charge at zero temperature in the case of small molecules of size  $N = 2, 3$  by decomposing it onto a set of Gaussians. The Kantorovich dual is then optimised using a *derivative-free* algorithm of a *quasi-Newton* type. However, this strategy does not scale for too many particles, as noted by the authors: « *the derivative-free methods are not suitable for optimizing with respect to a large number of degrees of freedom. More efficient numerical methods need to be developed in order to obtain the Kantorovich dual solution for more general systems* ». Our first contribution is therefore to consider the positive temperature problem, which is more amenable from the point of view of optimisation.

Indeed, the problem at positive temperature can be more easily tackled numerically using gradient methods. Recall that the dual at positive temperature reads

$$\sup_V \left\{ -\beta^{-1} \log Z_{N,\beta}(V) - \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}, \quad (80)$$

where  $Z_{N,\beta}(V)$  is the partition function associated with the Gibbs measure  $\mathbb{P}_\beta(V)$  as defined in (74). At zero temperature, the *free energy* term, namely  $-\beta^{-1} \log Z_{N,\beta}(V)$ , must be replaced by the ground-state energy  $E_N(V)$ . As mentioned above, it is difficult to access either analytically or numerically the functional derivative of  $E_N(V)$  with respect to the external potential  $V$ . In fact, it is not self-evident in which sense this derivative should be understood. On the other hand, the free energy can easily be differentiated. A calculation gives

$$\frac{\delta}{\delta V(x)} \left( -\beta^{-1} \log Z_{N,\beta}(V) + \int_{\mathbb{R}^d} V(x) \rho(x) \right) = \rho_{\mathbb{P}_\beta(V)}(x) - \rho(x). \quad (81)$$

where we recall that the quantity  $\rho_{\mathbb{P}_\beta(V)}$  corresponds to the density of the Gibbs measure  $\mathbb{P}_\beta(V)$ . Since the dual is a concave maximisation problem, we can write and solve for the first-order optimality condition, which then reads  $\rho_{\mathbb{P}_\beta(V)} = \rho$ . This optimality condition is not surprising, and is well known in the context of entropic optimal transport.

We will now discretise the dual charge  $\rho^{(e)}$ . The simplest idea is to give ourselves a finite basis of functions  $B = \{\mu_i \in L^1(\mathbb{R}^d, \mathbb{R}_+) : i = 1, \dots, M\}$  and

to approximate  $\rho^{(e)}$  by a linear combination of the elements of  $B$ , that is to approximate  $\rho^{(e)}$  by  $\rho^{(e)}[\nu]$  where

$$\rho^{(e)}[\nu] := \sum_{i=1}^M \nu_i \mu_i, \quad \nu = (\nu_i)_{i=1}^M \in \mathbb{R}_+^M. \quad (82)$$

This is equivalent to approximating the Kantorovich potential  $V$  by the potential  $V[\nu]$  where

$$V_\nu = \sum_{i=1}^M \nu_i V_i \quad \text{where } V_i = g_d * \rho_i \quad \text{for } i = 1, \dots, M. \quad (83)$$

We then optimise on  $\nu \in \mathbb{R}_+^M$  the problem

$$\sup_{\nu \in \mathbb{R}_+^M} \left\{ -\beta^{-1} \log Z_{N,\beta}(V_\nu) - \int_{\mathbb{R}^d} V_\nu(r) \rho(r) \right\}. \quad (84)$$

If we denote  $G(\nu)$  the function to be maximised, the derivative with respect to  $\nu_i$  of  $G$  then reads

$$\partial_{\nu_i} G(\nu) = \int_{\mathbb{R}^d} V_i \rho_{\mathbb{P}_\beta(V_\nu)} - \int_{\mathbb{R}^d} V_i \rho. \quad (85)$$

The second term in the right-hand side, which remains fixed throughout the optimisation, can easily be calculated beforehand. The first term, which involves the density  $\rho_{\mathbb{P}_\beta(V_\nu)}$  of the Gibbs measure  $\mathbb{P}_\beta(V_\nu)$ , is more complicated to calculate, and all the numerical cost of the optimisation is hidden in it. A first strategy would be to calculate the density  $\rho_{\mathbb{P}_\beta(V_\nu)}$  by discretising  $\mathbb{R}^d$ . Nevertheless, this would not be at all efficient. Instead, by using that this density is by definition ( $N$  times) the first marginal of  $\mathbb{P}_\beta(V_\nu)$ , it follows that

$$\int_{\mathbb{R}^d} V_i \rho_{\mathbb{P}_\beta(V_\nu)} = \int_{\mathbb{R}^{dN}} \sum_{j=1}^N V_i(x_j) d\mathbb{P}_\beta(V_\nu)(x_1, \dots, x_N). \quad (86)$$

We therefore find ourselves with a classical problem of numerical integration with respect to a probability measure. This quantity can be estimated using *Monte-Carlo methods* [RC04; RC10]. Of course, this requires being able to sample from the Gibbs measure  $\mathbb{P}_\beta(V_\nu)$ , which is not a trivial task at all.

Recall that we defined the Gibbs measure  $\mathbb{P}_\beta(V)$  with respect to the product measure  $(\rho/N)^{\otimes N}$ . Therefore, when  $\rho$  has compact support, it is possible to appeal to vanilla Monte-Carlo sampling by drawing samples according to the uniform measure on the support. However, it would be much more efficient to sample directly from the  $\mathbb{P}_\beta(V)$ , especially in the small temperature regime. Indeed, when the temperature is small, the Gibbs measure  $\mathbb{P}_\beta(V)$  will typically concentrate on the minimisers of the zero temperature energy. These minimisers typically live on low-dimensional and singular manifolds.

Sadly, there is no direct method for sampling  $\mathbb{P}_\beta(V)$  for any external potential  $V$ . In a few special cases, it is possible to relate the Gibbs measure  $\mathbb{P}_\beta(V)$  to random matrices models and thus obtain an exact sampling procedure. This

is the case, for example, in dimension  $d = 1, 2$  for the logarithmic interaction potential  $-\ln|x - x'|$  and the harmonic potential  $V(x) = |x|^2$ , in which case we recover the *Ginibre ensemble* [Gin04] when  $\beta = 2$  more generally the  $\beta$ -ensembles [DE02; GBV21] for any  $\beta > 0$ . Consequently, we need to use approximate sampling methods, such as the *Markov Chain Monte-Carlo* methods [RC10; RC04].

In Chapter 1, we present some numerical results. We apply our discretisation in dimension  $d = 1$  for the Coulomb potential  $-|x - x'|$ , where the set of optimal transport plans is known exactly — by the results of [CDPDM15] and by a small contribution of ours presented in the Appendix of Chapter 1 — as well as in dimension  $d = 3$  up to  $N = 30$  particles when the target density  $\rho$  is the uniform measure on the unit ball, so called *droplets* in physics. The results are then compared with those of Räsänen *et al.* in [RSGG11]. We note that the results of these authors were obtained using a construction due to Seidl for radial densities  $\rho$  [SGGS07]. Although it is not rigorously known that these trial states provide a minimiser in the case of droplets, our results show that they have nevertheless an energy which is very close to the minimum value.

It remains to justify all our previous claims mathematically.

### Theoretical justification

*Does there exist a dual charge?* That is, given a target density  $\rho$ , is there always a measure  $\rho^{(e)}$  such that the electrostatic potential  $V = g_d * \rho^{(e)}$  generated by  $\rho^{(e)}$  is a Kantorovich potential for the associated optimal transport problem? All the following theorems are stated and proved for  $d \geq 3$ , to serve away for few technical details in dimensions  $d = 1, 2$ , for which the Coulomb potential is not bounded from below. However, we expect similar results to be true for  $d = 1, 2$ . At zero temperature, we obtain the following theorem.

**Theorem** (Theorem 1 of Chapter 1, page 83). *Given a target density  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  such that  $\int_{\mathbb{R}^d} \rho = N \geq 2$ , there exists a Kantorovich potential of the form  $V(x) = \rho^{(e)} * g_d(x)$  for some positive measure  $\rho^{(e)}$  of mass  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N - 1$ . Moreover, if the support  $\Sigma$  of  $\rho$  is bounded, we can always assume that the measure  $\rho^{(e)}$  is supported on  $\Sigma$ .*

The existence of a dual charge  $\rho^{(e)}$  follows from considerations in *potential theory* [Lan72]. It is known that we can always find a Kantorovich potential  $V$  which satisfies the equation

$$V(x) = \inf_{x_2, \dots, x_N} \left\{ c(x, x_2, \dots, x_N) - \sum_{i=2}^N V(x_i) \right\} \quad (87)$$

where  $c$  is the cost of transportation associated with the Coulomb potential  $g_d$  in dimension  $d \geq 3$ , that is

$$c(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^{d-2}}. \quad (88)$$

When the particles  $x_2, \dots, x_N$  are fixed, the function  $x \rightarrow c(x, x_2, \dots, x_N)$  being superharmonic, it follows that the potential  $V$  which verified the above equation is superharmonic — under the hypothesis that it is continuous, which is not entirely trivial. We can then conclude, appealing to a well-known theorem from potential theory, that  $V$  can be written up to an additive constant as the Coulomb potential generated by a positive measure. This measure is precisely our dual charge  $\rho^{(e)}$ . That this measure can be assumed to be supported on the support  $\Sigma$  of the density  $\rho$  when  $\Sigma$  is bounded also follows from another well-known theorem in potential theory. We then extend the previous theorem to the positive temperatures.

**Theorem** (Theorem 3 from Chapter 1, page 85). *Given a target density  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  such that  $\int_{\mathbb{R}^d} \rho = N \geq 2$  and which verifies that the potential  $\rho * g_d$  is uniformly bounded, that is  $\rho * g_d \in L^\infty(\mathbb{R}^d)$ , then there exists a positive measure  $\rho_\beta^{(e)}$  such that  $V_\beta(x) = \rho_\beta^{(e)} * g_d(x)$  up to an additive constant on the support of  $\rho$ , where  $V_\beta$  is the unique Kantorovich potential for the optimal transport problem at positive temperature with target density  $\rho$ . Moreover, if the support  $\Sigma$  of  $\rho$  is bounded, we can always assume that the measure  $\rho_\beta^{(e)}$  is supported on  $\Sigma$ .*

The proof essentially follows the same lines as for the zero temperature case. A small subtlety consists in correctly extending  $V_\beta$  to the whole of space. Remember that  $V_\beta = \infty$  outside the support of  $\rho$ . We then show that  $V_\beta$  is superharmonic using the fixed-point equation verified by  $V_\beta$ , see (75).

The dual charge is not unique, at least in the case where the support of  $\rho$  is compact, see Remark 2 of Chapter 1, on page 84. However, the uniqueness of the latter can be obtained on the support of  $\rho$ , as stated in the following theorem.

**Theorem** (Theorem 2 of Chapter 1, page 84). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  be such that  $\int_{\mathbb{R}^d} \rho = N \geq 2$ , and  $\rho^{(e)}, \rho^{(e)'} \in \mathcal{M}(\mathbb{R}^d)$  be two dual charges for the optimal transport associated with the density  $\rho$ . Then the two measures coincide on the topological interior of the support of  $\rho$ . In particular, if the support is given by  $\mathbb{R}^d$ , then the two measures coincide everywhere. This uniqueness result is valid both at zero and positive temperature.*

We had the idea of adding temperature to our system and making this temperature tend towards zero in order to recover the problem at zero temperature. From the point of view of the value of  $F_{N,w}(\rho)$ , it is well known that  $F_{N,w,T}(\rho) \rightarrow F_{N,w}(\rho)$  [Car+17], which shows the consistency of this approach. *What about the convergence of the dual charge?* We show that the dual charge  $\rho_\beta^{(e)}$  at positive temperature does indeed converge to a dual charge at zero temperature as the temperature tends towards 0. This result follows from a more general theorem on the convergence of Kantorovich potentials:

**Theorem** (Theorem 5 of Chapter 1, page 86). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  be such that  $\int_{\mathbb{R}^d} \rho = N \geq 2$  and such that  $\rho * g_d$  is uniformly bounded. We also require that the support  $\Sigma$  of  $\rho$  be bounded. Let  $V_\beta$  be the Kantorovich potential of*

the optimal transport problem at positive temperature with the target density  $\rho$ . Then the potential  $V_\beta$  is bounded up to an additive constant in  $W^{1,\infty}(\mathbb{R}^d)$  uniformly in  $\beta$  when  $\beta \rightarrow \infty$ . In other words,  $V_\beta$  is uniformly Lipschitz. In particular, up to a subsequence, there exists  $V \in W^{1,\infty}(\mathbb{R}^d)$  such that

$$V_\beta \xrightarrow[\beta \rightarrow \infty]{} V \quad \text{uniformly over compact sets.} \quad (89)$$

Moreover, the function  $V$  is a Kantorovich potential for the transport problem at zero temperature.

The convergence of the Kantorovich potentials for the entropic problem to a Kantorovich potential for the original entropy-free problem was recently proved in [NW22]. While the results in [NW22] are more generic, our theorem gives a stronger convergence, although *a priori* only for the Coulomb cost. It follows from the convergence at the level of the Kantorovich potentials that the dual charge at positive temperature converges to a dual charge at zero temperature, as stated in the following theorem.

**Theorem** (Theorem 4 of Chapter 1, page 85). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  be such that  $\int_{\mathbb{R}^d} \rho = N \geq 2$  and such that  $\rho * g_d$  is uniformly bounded. Let  $\rho_\beta^{(e)}$  be a dual charge such that  $\text{supp}(\rho_\beta^{(e)}) \subset \Sigma$ , that is such that  $V_\beta(x) = \rho_\beta^{(e)} * g_d(x)$  is the unique Kantorovich potential of the transport problem at positive temperature with target density  $\rho$ . Then, up to a subsequence, there exists a measure  $\mu$  in  $\mathcal{M}_+(\Sigma)$  such that*

$$\rho_\beta^{(e)} \xrightarrow[\beta \rightarrow \infty]{} \mu \quad \text{for the weak topology on } \mathcal{M}(\Sigma). \quad (90)$$

Moreover, the measure  $\mu$  is a dual charge for the zero temperature problem.

We have seen that the dual charge verifies  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N - 1$ . However, as we explained earlier, we have the intuition that the charge must necessarily be of mass greater or equal to  $N - 1$ , otherwise the system should not be stable. It is in fact possible to show that the charge is of mass  $N - 1$ , at least in the case where the support of the target density  $\rho$  is not bounded, otherwise we can always freely modify the charge outside  $\Omega$  at will, see Remark 2 in Chapter 1, page 84. This equality follows from a conjecture on the asymptotic behaviour of the Kantorovich potential, which we now present, and which will be the subject of Chapter 2.

## §4. Asymptotic of the Kantorovich potential (Chapter 2)

We now present the results of chapter 2, which is a reproduction of our paper [Lel22b]. It has been conjectured [FGGG22] that the Kantorovich potential  $V$  associated to the multimarginal optimal transport with Coulomb cost verify that

$$V(x) \sim -\frac{N-1}{|x|} \quad \text{when } |x| \rightarrow \infty. \quad (91)$$

To the best of our knowledge, although this conjecture was written down explicitly for the first time by Friesecke *et al.* in [FGGG22], the question is older and can be found several times in the work of chemists, see for example [SGGS07; VLGG17]. Until now, the asymptotic was rigorously known in dimension  $d = 1$  for any number of particles  $N$ , using the results of Colombo, De Pascale and Di Marino in [CDPDM15], and for  $N = 2$  in any dimension when the target density  $\rho$  is radial, in which case the transport problem can be explicitly solved as shown by Pass in [Pas15]. In Chapter 2, we prove this conjecture in full generality for the *Riesz costs*.

**Theorem** (Theorem 19 of Chapter 2, page 123). *Let  $\Omega \subset \mathbb{R}^d$  be a connected and unbounded open set. Let  $\rho \in L^1(\Omega, \mathbb{R}_+)$  be a density such that  $\int_{\mathbb{R}^d} \rho = N$  and such that  $\rho(r) > 0$  almost everywhere on  $\Omega$ . Let  $V$  be a Kantorovich potential for multimarginal optimal transport with Riesz cost, that is  $w_s(x) = |x|^{-s}$  for  $s > 0$ . Then, there is a constant  $C$  such that*

$$V(x) + C \sim -\frac{N-1}{|x|^s} \quad \text{when } |x| \rightarrow \infty \text{ and } x \in \Omega. \quad (92)$$

To prove this theorem, we follow a result of [FGGG22], where it is stated that the asymptotic follows more or less immediately from another conjecture. Our main contribution in Chapter 2 is the proof of this conjecture, which reads as follows.

**Theorem** (Theorem 21 of Chapter 2, page 125). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  be such that  $\int_{\mathbb{R}^d} \rho = N$ . If  $\mathbb{P}$  is an optimal (symmetric) transport plan for the multimarginal optimal transport problem with Riesz cost  $w_s(x) = |x|^{-s}$  with  $s > 0$ , then there exists a large enough  $R > 0$  such that*

$$\mathbb{P} \left( (\mathbb{R}^d \setminus B_R) \times (\mathbb{R}^d \setminus B_R) \times \mathbb{R}^{d(N-2)} \right) = 0, \quad (93)$$

where  $B_R \subset \mathbb{R}^d$  denotes the open ball of radius  $R$  centred at the origin

This theorem states that if  $\mathbb{P}$  is an optimal transport plan, then if one particle, say  $x_1$ , is sent to infinity  $|x_1| \rightarrow \infty$ , then all the other particles must remain in a compact subset. Our proof relies on the notion of *c-cyclical monotonicity* [San15; Vil09].

*How does this theorem imply the asymptotic?* As noted by Friesecke *et al.* [FGGG22], given a target density  $\rho$ , we recall that the Kantorovich potential  $V$  is the potential that forces the particles to live on an optimal transport plan  $\mathbb{P}$ . In particular, we have seen that the optimality conditions should be interpreted as equilibrium equation for classical mechanics, which we remind to read

$$\nabla V(x_i) = \sum_{j \neq i}^N \nabla_{x_i} w_s(x_i - x_j) \quad (94)$$

for all  $i = 1, \dots, N$ . Since this equilibrium equation holds over the entire support of  $\mathbb{P}$  and since the support of the density  $\rho$  is not compact, we can send a particle, for example  $x_1$ , to infinity. According to the above theorem, all the



other particles  $x_2, \dots, x_N$  must remain in a compact subset. Consequently, we obtain when  $|x_1| \rightarrow \infty$  that

$$\nabla V(x) \sim (N-1)w'_s(x) \quad \text{when } |x| \rightarrow \infty \quad (95)$$

which essentially allows us to conclude.

The fact that the asymptotic is verified is important in the context of the DFT. Without going into too much detail, the asymptotic ensures that the approximation produced by the SCE formalism allows to account for certain important properties of the real system — properties that are often missed by other approximations, such as the Kohn-Sham formalism. We refer to [VLB94; TH03] and the references given at the end of this section. We also note that this asymptotic, following an argument by Seidl *et al.* in [SVGG16] — see also [LO81, Appendix A] — can be used to obtain information on the minimisers of the *Lieb–Oxford inequality* [LO81; Lie79; Sei+22; LS09; LLS19b]. This inequality is used in DFT to calibrate the exchange-correlation functionals [LP93; PBE96; PS22; Sun+16]. We shall return briefly to this question at the end of Chapter 2, which is the subject of an article in preparation with S. Di Marino [DML].

### Link with the dual charge

We have stated that the dual charge verifies  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N-1$ . Thanks to the asymptotic, we can now affirm, as dictated by our intuition, that this inequality is in fact an equality. In Chapter 2, we prove the following theorem.

**Theorem** (Corollary 20 of Chapter 2, page 124). *Let  $\Omega \subset \mathbb{R}^d$  be a connected and unbounded open set. We assume that  $\Omega$  does not "shrink too much at infinity" in certain directions, that is that  $|A_\Omega| > 0$ , where  $|\cdot|$  is the Lebesgue measure on the sphere  $\mathbb{S}^{d-1}$  and where*

$$A_\Omega := \left\{ \xi \in \mathbb{S}^{d-1} : \text{for all } r_0 \text{ there exists } r \geq r_0 \text{ t.q. } r\xi \in \Omega \right\}.$$

*Let  $\rho \in L^1(\Omega, \mathbb{R}_+)$  be such that  $\int_{\mathbb{R}^d} \rho = N$  and such that  $\rho(x) > 0$  almost everywhere on  $\Omega$ . Let  $\rho^{(e)}$  be a positive measure such that  $|\cdot|^{2-d} * \rho^{(e)}$  is a Kantorovich potential for the multimarginal transport problem with Coulomb cost. Then we have*

$$\int_{\mathbb{R}^d} \rho^{(e)} = N-1. \quad (96)$$

We end this section with a quick comment on the philosophy of the dual charge. We mentioned that this dual charge, while appearing in the framework of optimal transport for the Coulomb cost in [ML13], was also an object found in the DFT literature [Gö99; LAP99]. In DFT, it is sometimes more interesting to consider the charge that generates the exchange-correlation potential rather than the potential itself. The reason is that it is then much easier from a numerical point of view to enforce the correct asymptotic. As noted in [VLGG17] "The main advantage of approximating [the exchange-correlation potential  $V$ ] by



modeling [the dual charge  $\rho^{(e)}$ ] is the fact that if a model for  $[\rho^{(e)}]$  integrates to  $[N - 1]$  and vanishes at infinity, then the corresponding  $[V]$  will have the correct asymptotic behaviour [...] which is usually missed by the standard density functional approximations". We refer to [Tok05; VLGG17; Mar02; LAP99; KS13; Gö99; GL12; AL01; AAG11; AU85] for more details on this issue.

## §5. Phase transition for Riesz gases in dimension one (Chapter 3)

In this section we present the results of Chapter 3. We study there the *Riesz gas* [Lew22a] in dimension  $d = 1$ , that is particles on the real line that interact via the Riesz potential as defined below (97). We are interested in the case of non-singular interactions, that is when the exponent  $s$  in the Riesz potential is negative. In this case, the Riesz potential is given by  $-|r|^{-s}$ . In particular, we study the case where  $s$  lies between the Coulomb potential  $s = -1$  and the logarithmic interaction  $s = 0$ . For  $s = -1$ , we obtain the *Coulomb gas*. This remarkable system is known to be crystallised at any temperature [Kun74; AM80]. On the other hand, in the case of the logarithmic interaction  $s = 0$ , the corresponding model is called the Dyson *log-gas* and it is expected that this system is a fluid at all temperature. In Chapter 3, following a question raised recently in [Lew22a], we show numerically that there exists a phase transition with respect to the temperature and the exponent  $-1 < s < 0$  that interpolates between the phase diagrams of the Coulomb gas and the log-gas.

### §5.1. Definition of Riesz gases in arbitrary dimension

We define the *Riesz gas* [Lew22a]. We consider pointlike classical particles located in  $x_1, \dots, x_N \in \mathbb{R}^d$  which interact with each other through the *Riesz potential*  $w_s$ . This potential is a natural generalisation of the Coulomb potential. For all  $s \in \mathbb{R}$ , it is defined as

$$w_s(x) = \begin{cases} |x|^{-s} & \text{for } s > 0, \\ -\ln|x| & \text{for } s = 0, \\ -|x|^{-s} & \text{for } s < 0. \end{cases} \quad (97)$$

When the exponent  $s$  is negative, the sign is switched so that the potential remains repulsive. We can see that the Coulomb potential in dimension three corresponds to the choice  $s = 1$ . More generally, the choice  $s = d - 2$  yields the Coulomb potential in arbitrary dimension  $d \geq 1$ , the definition of which was recalled in the previous section, see (77). In two space-dimension, we find the logarithmic interaction potential  $-\ln|x - x'|$  corresponding to the case  $s = 0$  in (97). This is a natural convention since it corresponds to the first order of  $w_s$  when  $s \rightarrow 0^+$ .

We fix a domain  $\Omega \subset \mathbb{R}^d$ , and we constrain the particles to live in  $\Omega$ . In what follows, we are interested in the *long-range* case, that is when  $s < d$ . The

long-range case is particularly interesting as of physics, as it allows for the appearance of phase transitions. It is also generally much more complicated to study mathematically than the *short-range* case, that is  $s > d$ . It follows from a consideration of potential theory that in the long-range case  $s < d$ , the electrons, seeking the best possible configuration to compensate for the strong repulsion they exert on each other, will concentrate on the boundary of  $\Omega$ . This has dramatic effect on the energy of the system. To prevent this, we are going to add a compensating *uniform background* of positive charge over  $\Omega$ . This background will then generate an external potential which will bring the particles back inside the interior of  $\Omega$ . We can then define the energy of the system as

$$H_{s,\Omega}(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} w_s(x_i - x_j) - \sum_{i=1}^N \int_{\Omega} w_s(x_i - x') dx' + \frac{1}{2} \iint_{\Omega \times \Omega} w_s(x - x') dx'. \quad (98)$$

The first term corresponds to the interaction of the particles with each other, and the second term to the interaction of the particles with the uniform background. The third and last term corresponds to the interaction energy of the background with itself. This is a constant term that can be neglected when working at fixed  $N$ . However, when we consider the *thermodynamic limit*, that is the limit  $N \rightarrow \infty$ , this term is very important and cannot be neglected in the long-range case  $s < d$ .

The idea of adding a uniform background is well known in physics. In the case of the Coulomb potential  $s = d - 2$ , we find the *Jellium* [Tho04; Wig34; Lew22a], which is very a important model in condensed matter physics. In this context, the uniform background should be seen as a simplified model for the atomic lattice over which electrons are delocalised. In practice, in the laboratory physicists use external potentials that can vary in space and whose associated background charge is not uniform [CI94; Tho+94; HT94]. However, in a region where the potential varies slowly, Jellium is expected to be a good *local* approximation.

At inverse temperature  $\beta \in (0, \infty]$ , our particles are modelled by the *canonical ensemble* [Rue99], that is the Gibbs measure whose density is given by

$$\mathbb{P}_{\beta,s,\Omega}(x_1, \dots, x_N) = \frac{1}{Z(\beta, s, \Omega)} \exp(-\beta H_{s,\Omega}(x_1, \dots, x_N)). \quad (99)$$

Here  $Z(\beta, s, \Omega)$  is the *partition function*, that is the normalisation constant which ensures that  $\mathbb{P}_{\beta,s,\Omega}$  is indeed a probability measure, as encountered previously in the study of the entropic optimal transport. The *free energy* of the canonical ensemble is given by

$$F_s(\beta, \Omega) := -\beta^{-1} \log Z(\beta, s, \Omega). \quad (100)$$

The *Riesz gas* is the object obtained when we consider the thermodynamic limit  $N \rightarrow \infty$  in the preceding quantities. As far as the free energy is concerned,

we need to renormalise by the volume  $|\Omega|$ . We can then define the free energy of the Riesz gas per unit volume as the limit

$$f(s, \beta) := \lim_{\substack{\Omega \nearrow \mathbb{R}^d \\ N \rightarrow \infty \\ \text{and } |\Omega|=N}} \frac{F_s(\beta, \Omega)}{|\Omega|}. \tag{101}$$

For the Riesz gas with short-range interaction  $s > d$ , the existence of this limit is well known, see [Rue99; Geo76; Geo11; DM67]. In the long-range case  $s < d$ , which is generally much more complicated to study, we refer to [Lew22a, Sec. III] and the references mentioned therein. In the specific case of the Coulomb potential  $s = d - 2$ , the existence was proved by Lieb and Nahnofer in dimension  $d = 3$  in a landmark paper [LN75]. The existence was then generalised to arbitrary dimensions  $d \geq 2$  by Sari and Merlini [SM76].

Turning now to the limit of the Gibbs measure  $\mathbb{P}_{\beta,s,\Omega}$ , we expect the object obtained in the limit  $N \rightarrow \infty$  to be a *Gibbsian point process* [Der19; Geo11; Lew22a; Jan]. There exists a whole family of rigorous characterisations/definitions of this object, which are generally believed to be equivalent. For example, we can mention the equations of *Dobrushin–Landford–Ruelle* (DLR) or the hierarchy of *Bogolioubov–Born–Green–Kirkwood–Yvon* (BBGKY) [GLM80]. The limiting process is notably characterised by its *k-point correlation functions*, defined in finite volume  $N < \infty$  by

$$\rho^{(k)}(x_1, \dots, x_k) := \frac{N!}{(N - k)!} \int_{\mathbb{R}^{d(N-k)}} \mathbb{P}_{\Omega,s,\beta}(x_1, \dots, x_k, dx'_{k+1}, \dots, dx'_N). \tag{102}$$

A first fundamental question in the study of the Riesz gases is the existence of this limiting process, which is a rather delicate question. While the case of short-range interactions  $s > d$  is well studied [Rue99; Geo76; Geo11; DM67], the long-range case  $s < d$  is much more complicated and many open problems remain unsolved [Lew22a, Sec. III]. Another fundamental question is that of the uniqueness of this limiting process. In statistical physics, the non-uniqueness of this process is intrinsically linked to the *breaking of symmetry* and therefore to the existence of phase transitions. Finally, there is also the problem of determining the nature of the phase (*e.g.* solid, liquid, gas etc.), which is linked to the properties of the correlation functions as defined above (102), and in particular to that of the two-point correlation function  $\rho^{(2)}(x, x')$ , as we shall see later.

It is also interesting to consider the Riesz gas with periodic boundary conditions [Lew22a, Sec. IV], where the particles interact with each other as well as with their images in the periodised domain  $\Omega$ . In the short-range case  $s > d$ , the periodised Riesz potential  $\widetilde{v}_s$  reads

$$\widetilde{v}_s(x) = \sum_{z \in \mathcal{L}} \frac{1}{|x - z|^s}, \tag{103}$$

where  $\mathcal{L}$  is the periodic lattice. While this series converges when  $s > d$ , this is no longer the case when  $s < d$ . However, it can be shown that the function  $\widetilde{v}_s(x)$ , seen as a function on the complex plane  $s \in \mathbb{C}$ , admits a meromorphic

extension on  $\mathbb{C} \setminus \{d\}$  with a pole at  $s = d$  [BBS89; Bor+88; BBT85; BBS14], see also [Lew22a, Sec. IV], like the Riemann zeta function to which it is tightly related. This periodised potential can in fact be defined by its Fourier transform, given up to a multiplicative constant by

$$\widehat{v}_s(k) = \sum_{k \in \mathcal{L}^* \setminus \{0\}} \frac{\delta_k}{|k|^{d-s}}. \quad (104)$$

Here,  $\mathcal{L}^*$  is the dual lattice of  $\mathcal{L}$ , that is the lattice generated by the dual basis of that of  $\mathcal{L}$ . It turns out that the meromorphic extension to  $d - 2 < \Re(s) < d$  exactly corresponds to adding the uniform background in (103) as introduced earlier. We refer to [Lew22a, Sec. IV] for more details on this matter.

In practice, periodic boundary conditions are particularly useful from a numerical point of view in the long-range case  $s < d$ . Indeed, they can be used to get rid of boundary effect, which can be considerable in the case of long-range interactions. In Chapter 3, our numerical experiments are carried out with these periodic boundary conditions.

## §5.2. Known results in dimension $d = 1$

We now concentrate on the one-dimensional case  $d = 1$ . We present the known theoretical results for the Riesz gas in dimension one, and we discuss two very important integrable models, namely the *Coulomb gas* and the *log-gas*, which correspond respectively to the exponent  $s = -1$  and  $s = 0$ , that is to the potential  $-|x|$  and  $-\ln|x|$ .

The existence of the free energy in the thermodynamic limit (101) was proved by Kunz [Kun74] for the case of the Coulomb potential  $s = -1$ . In the case  $s \geq 0$ , the existence is contained in the works of Serfaty *et al.* We refer to [Ser15] for a self-contained presentation and the references in [Lew22a]. Although these works do not expressively deal with the case of negative exponents  $s < 0$ , they are still covered to some extent by the theory [Ser]. We can also mention the work of physicists [GLM80; GLM81; BGM84] — see the references of [Lew22a]. Hence, the thermodynamic limit of the free energy exists for all  $s \geq -1$ . It is in fact probable that the existence holds up to  $s > -2$  [Lew22a], even if the result is not known at the present time. In the case where we impose periodic boundary conditions, the limit has been proved in [Lew22a] by extending a result of [Har+15].

As of the existence and the uniqueness of the limiting point process in dimension  $d = 1$ , it is known that the limiting process is translation-invariant for  $s > 1$  [FP81] and that it is unique for  $s > 2$  [Pap87]. Consequently, the Riesz gas for  $s > 2$  has no phase transition and is a fluid at all temperature. Since the invariance by translation is not broken for  $s > 1$ , we very much expect that the process is unique for  $s > 1$ . Even more so, it is believed that this is certainly the case for all  $s \geq 0$ . The case  $0 < s < 1$  has been recently studied by Dereudre and Vasseur [DV23], who proved the existence of the DLR equations for the limiting process, and by Boursier [Bou22] who obtained a series of results in the periodic case. Another, quite remarkable, case where the existence and

(non-)uniqueness of the limiting process are known is for the Coulomb potential  $s = -1$ , as we now discuss.

A very specific case is the *Coulomb gas*, which is obtained for  $s = -1$ . This fully integrable model is very well studied in the literature [AM80; AJJ10; BLC63; CGZJ20; Cho75; Kun74]. Kunz [Kun74] has obtained several remarkable results on the Coulomb gas, including the existence of the free energy already mentioned and the existence of the correlations functions (102) of the point process, for which he also obtained an explicit formula. These functions are not unique, which allows to rigorously establish the non-uniqueness of the limiting point process and consequently the breaking of symmetry at any temperature. He also showed that these functions are periodic, which proves that the system is a crystal at any temperature. Yet, these functions must not be constant to ensure this. Kunz proved that this was indeed the case at low enough temperature. This result was eventually generalised to all temperature by Aizenman and Martin [AM80], who also proved that the point processes verified both the BBGKY hierarchy and the DLR equations. We note that the case of the quantum Coulomb gas in dimension one was studied and solved by Brascamp and Lieb [BL02] and Jansen and Jung [JJ14]. We insist that it is quite fortuitous that the Coulomb gas is fully integrable. This is in particular due to the very specific form of the Coulomb potential in dimension one, that is  $-|r|$ . For example, we can see that the force exerted by one particle on another does not depend on their mutual distance, which makes it possible to obtain some very interesting properties that greatly simplify the mathematical analysis.

Another very important case is that of the logarithmic interaction  $s = 0$ , for which we find the *log-gas* model in dimension  $d = 1$ . This model was introduced by Dyson in the works [Dys62a; Dys62b; Dys62c; DM63; MD63] which, along with the work of Wigner [Wig55; Wig67] are at the origin of the field of *random matrix theory* in mathematics [For10; Meh04]. In this context, the log-gas is also called the  $\beta$ -ensemble or the *sine- $\beta$*  process. Recall that  $\beta$  is the inverse temperature  $\beta = T^{-1}$ . For  $\beta = 1, 2$  and  $4$ , the log-gas corresponds to the three well-known *Gaussian ensembles* in the theory of random matrices, which are respectively the GOE (for *Gaussian Orthonormal Ensemble*), the GUE (for *Gaussian Unitary Ensemble*) and the GSE (for *Gaussian Symplectic Ensemble*) [For10]. The log-gas is also a fully integrable model that is widely studied in the literature. We refer the reader to the numerous references in [Lew22a], or to the monograph [For10]. If the uniqueness of the log-gas is not *a priori* known rigorously, it is predicted that there can be no symmetry breaking [RW90]. In fact, the uniqueness has been proved when we restrict ourselves to stationary point processes [EHL21]. Consequently, it is agreed on that the log-gas in dimension one has no phase transition, and that it is a fluid at all temperatures — except at zero temperature where it is crystallised [SS15; Leb15].

Although the log-gas does not exhibit symmetry breaking at positive temperatures, there is a *Berezinskii–Kosterlitz–Thouless* (BKT) transition [Mud14; Her07; KT73] at  $\beta = 2$ . In dimension one, this transition is seen by a qualitative change in the behaviour of the two-point correlation function  $\rho^{(2)}(r, r')$  (102).

Using a fairly general theory of Haldane on one-dimensional quantum fluids [Hal81], Forrester conjectured in [For84] that

$$g(r) \underset{r \rightarrow \infty}{\sim} \begin{cases} 1 - \frac{1}{\pi^2 \beta r^2} & \text{for } \beta < 2, \\ 1 + \frac{\cos(2\pi r)}{2\pi^2 r^2} - \frac{1}{2\pi^2 r^2} & \text{for } \beta = 2, \\ 1 + c \frac{\cos(2\pi r)}{r^{4/\beta}} - \frac{1}{2\pi^2 r^2} & \text{for } \beta > 2 \end{cases} \quad (105)$$

for a universal constant  $c$ . Here, since the two-point correlation function  $\rho^{(2)}(r, r')$  only depends on the distance  $r - r'$ , we let  $g(r) := \rho^{(2)}(0, r)$ . We see that at  $\beta = 2$ , the correlation function transits from an *universal* decay to one whose exponent depends on the temperature. In addition, an oscillating term is seen to appear. However, in clear contrast to that of a crystal, these oscillations are damped and eventually vanish as  $r \rightarrow \infty$ . Forrester’s conjecture is verified in the special cases  $\beta = 1, 2$  and  $4$ , and more generally for any even or rational  $\beta$ , see [For93] and [For10, Chap. 13].

Finally, we mention the case  $s = 2$ , which corresponds to the classical *Calogero–Sutherland–Moser* (CSM) model [Sut03a; Sut04; Sut03b; Cal71], which is fully integrable [Cho00; GM73]. A remarkable fact is that the quantum version of this model is equivalent to the log-gas — see the references previously given. In fact, it was through this analogy and Haldane’s work that Forrester was able to conjecture the asymptotic behaviour of the pair correlation  $g(r)$  as mentioned above.

In this thesis, we are interested in the case of the exponents  $-1 < s < 0$  which interpolate between the Coulomb gas and the log-gas. Following the results that have been stated, Lewin in [Lew22a] asked the following question: *Is there a phase transition for the Riesz gases of exponent  $-1 < s < 0$  that interpolates between the phase diagrams of the Coulomb gas, for which we are always a solid, and the log-gas for which we are (believed to be) always a fluid.* The rest of this introduction is devoted to presenting our numerical results from Chapter 3, which answer this question in the affirmative.

### §5.3. Phase transitions in Riesz gases: numerical results

We therefore seek to determine whether or not there is a phase transition with respect to the temperature  $T$  for the Riesz gases in dimension one for the exponents  $-1 < s < 0$ . We emphasize that the vast majority of results with state the (non-)existence of phase transitions in the Riesz gases are numerical results. To answer our question theoretically, we would first need to define rigorously the Riesz gas, for example by exhibiting a solution to the associated DLR equations or the BBGKY hierarchy, as was done in the case  $0 < s < 1$  by Dereudre and Vasseur [DV23], and then be able to answer the question of the uniqueness or non-uniqueness of this limiting process. These problems are very difficult and very much open at the present time. We recall that the only rigorously known result regarding the (non-)existence of phase transitions in



dimension one is the uniqueness of the Riesz gas for  $s > 2$  [Pap87]. The results presented in Chapter 3 are therefore entirely numerical.

Before coming to these results, we quickly mention what is known in the case of the dimensions  $d = 2, 3$ .

### The case of dimensions $d = 2$ and $d = 3$

The dimension  $d = 3$  is the dimension for which numerical results are the most abundant in the literature. At high temperature, the Riesz gas is a gas — which can be rigorously proved in the short-range case  $s > d$  [Lew22a]. It is then expected that there is a *solid-fluid* transition at small enough temperature for all  $s > 0$ . Within the solid phase, it is also expected from numerical works that there is a *solid-solid* transition where the system transits between a *body-centred cubic* (BCC) crystal and a *face-centred cubic* (FCC) crystal for certain values of  $s$ , but not in the Coulomb case  $s = 1$  for which only the BCC crystal seems to exist. We refer to [Lew22a, Sec. VI. B.] for classical references on this subject.

In dimension  $d = 2$ , things are more subtle. The existence of phase transitions in two-dimensional systems has been the subject of several controversies in the history of contemporary physics, notably because numerical works seemed to highlight the existence of transitions where a well-known mathematical theorem seemed to forbid it. Indeed, it is known that there can be no spontaneous symmetry breaking in dimensions  $d = 1, 2$  for system interacting through short-range potential. This is stated by the famous *Hohenberg–Mermin–Wagner* theorem [Hoh67; Mer67; Mer68; MW66]. However, it should be noted that this theorem only applies to short-range interaction potentials. In particular, the Coulomb potential  $s = 0$  in dimension two or, more generally, the long-range Riesz potentials  $s < d$  for  $d = 1, 2$  do not fall within the scope of this theorem. In fact, as we have indicated, the Coulomb gas in dimension one is crystallised at any temperature [Kun74; AM80]. Alder and Wainwright<sup>15</sup> [AW62] were the first to highlight the existence of a phase transition in a two-dimensional

<sup>15</sup>Pioneers in the use of computers in statistical physics. We refer to [BC18] which traces back the career of the physicist Berni Alder (1925 – 2020) and at the same time the history of the introduction of computers in contemporary physics. A story that was not without some reluctance and controversies in the physics community, and which has been gradually transformed by the arrival of ever more powerful computers on the market. The article [BC18] relates an amusing anecdote about a vote by a show of hands: “*In the round table on statistical mechanics that took place just before Alder’s presentation, Kirkwood summed up the situation as follows: “the most recent evidence for the existence of the liquid-solid transition was provided by Dr. Alder [...] Following his work, Dr. William Wood of Los Alamos again took up the Monte Carlo calculations... it seems that the Rosenbluth calculations were in error at high densities; there is a very definite evidence for a first-order transition, which we would identify as a transition from a solid or crystalline state to a liquid state”. The exchange of viewpoints that followed showed clearly that, in spite of the evidence presented by both methods, the question was still far from being settled. [...] George Uhlenbeck half jokingly put the question [...] on a vote, remembering the audience that a vote taken in a similar occasion the previous fall, as to whether a gas of hard spheres has a transition point or not, had ended up even; “Now of course, with Mr. Kirkwood here, we have weighted the evidence a little bit. May I ask first of the panel to put up their hands if they believe there is a transition in the classical case... Now those who believe there is not a transition... Even, again!”*”

system where the theorem mentioned above seemed to forbid it. A decade later, Berezinskii, Kosterlitz and Thouless [Kos; Ber71; Ber72] brought to light the existence of phase transitions of a very particular type, which are called *topological*, and which we have already encountered in the case of the log-gas in dimension one (see above). Kosterlitz and Thouless were awarded the 2016 Nobel Prize for their work. Kosterlitz, in its acceptance lecture, quoted the work of Alder and Wainwright as inspiration — “*It also helped that there existed some experimental and numerical evidence for transitions to more ordered low temperature phases in 2D crystals*” [Kos]. Concerning our Riesz gases in dimension  $d = 2$ , while several numerical works were mentioning for a long time the existence of a first-order transition in the case  $s < 2$  [Str88], more recent works rather highlight a transition of a topological order *à la* BKT - we refer the reader to [Lew22a, Sec. VI. B.] for a list of references of this matter.

### The case of dimension $d = 1$ and $-1 < s < 0$

We present here the numerical results obtained in Chapter 3 regarding the phase diagram of the Riesz gas in one dimension for  $-1 < s < 0$ . In dimension one, the literature on phase transitions is much scarcer than in dimensions  $d = 2, 3$ . Following a reproduction of [Lew22a, Fig. 5], we summarise in Figure 9 the rigorously known or at least expected results that we have already mentioned. We recall that the Coulomb gas  $s = -1$  is crystallised at all temperature [Kun74; AM80], and that it is predicted that the log-gas  $s = 0$  is always a fluid, with a BKT transition at  $\beta = 2$ .

In Chapter 3, we show numerically that the intuition formulated in [Lew22a] seems correct, *i.e.* there exists a critical temperature  $T_s$  depending on the exponent  $-1 < s < 0$  such that below this temperature the Riesz gas is a solid and above which the system is a fluid. Furthermore, this temperature interpolates well with the rigorous — or at least expected — results for the Coulomb gas and the log-gas, namely that  $T_s \rightarrow \infty$  when  $s \rightarrow -1$ , which is consistent with the Coulomb gas being crystallised at all temperature, and that  $T_s \rightarrow 0$  when  $s \rightarrow 0$ , which is consistent with the log-gas being a fluid at all temperature. In addition, our numerical experiments suggest that there is a second phase transition  $\tilde{T}_s$  corresponding to a BKT-type transition, where the gas transits from a fluid to a *quasi-solid* phase. We observe that  $\tilde{T}_s$  converges to  $1/2$  when  $s \rightarrow 0$ , which is consistent with the BKT transition point of the log-gas. The schematic phase diagram for  $-1 < s < 0$  is then given by Figure 10.

To obtain this diagram, we simulated the Riesz gas with periodic boundary conditions using *Monte Carlo methods* [RC10; RC04]. We then calculated the pair correlation function  $g(r)$ . This function, which expresses how the density of particles of a system varies with respect to a given reference particle, is a very important quantity in statistical mechanics for the study of phase transitions. For example, in the case of a *perfect crystal*, the pair correlation is a periodic function — at zero temperature, it is even a Dirac comb. Indeed, given the position of one particle, the positions of all the other particles are determined by that very particle and the crystal lattice. In a *perfect fluid*, on the other



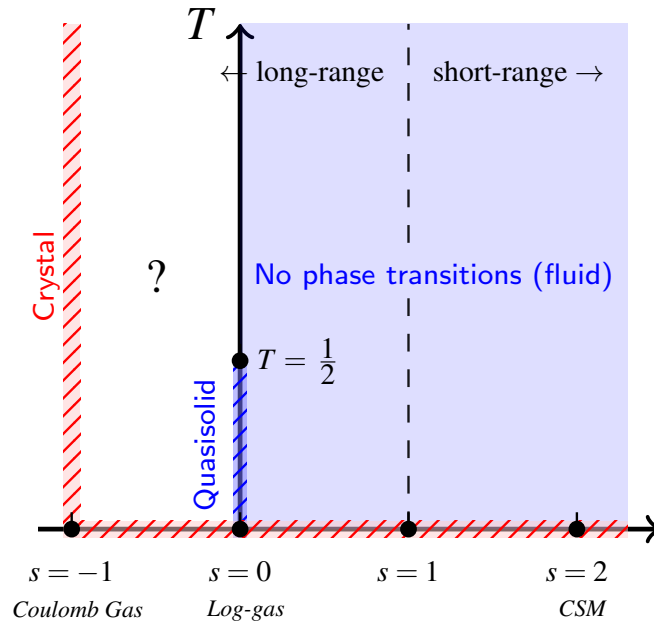


Figure 9: Reproduction of [Lew22a, Fig. 5] depicting the phase diagram of the Riesz gas in one space-dimension as a function of the temperature and the exponent  $s$  of the Riesz potential (97). Our aim in Chapter 3 is to complete the diagram for  $-1 < s < 0$ . Following [Lew22a], we ask ourselves whether or not there is a phase transition that interpolates between the phase diagrams of the Coulomb gas and the log-gas, which correspond to the exponents  $s = -1$  and  $s = 0$ .

hand, the particles are independent of each other, so that  $g(r)$  is a constant function which is equal to the average density of the system. More generally, in the presence of crystalline (quasi-)order,  $g(r)$  is expected to exhibit oscillations and/or a non-integrable long-range decay. On the other hand, in the case of a disordered system,  $g(r)$  is expected to decay rapidly at infinity towards the average density of the system, which is equivalent to saying that two particles that are far apart do not affect each other, as in a gas or a liquid. For the Coulomb gas with periodic boundary conditions, Kunz proved that  $g(r)$  is a properly periodic function, and consequently that the system is crystallised.

Following this phenomenology, we therefore seek to determine whether or not the pair correlation  $g(r)$  of the Riesz gas for  $-1 < s < 0$  becomes periodic at large distance  $r \gg 1$ , which would be a very clear manifestation of the existence of a crystalline order. On the contrary, if  $g(r)$  converges rapidly and monotonically to the average density, we expect the system to be a fluid. In the case of the BKT transition and the associated quasi-solid phase,  $g(r)$  should have oscillations that are gradually damped and eventually

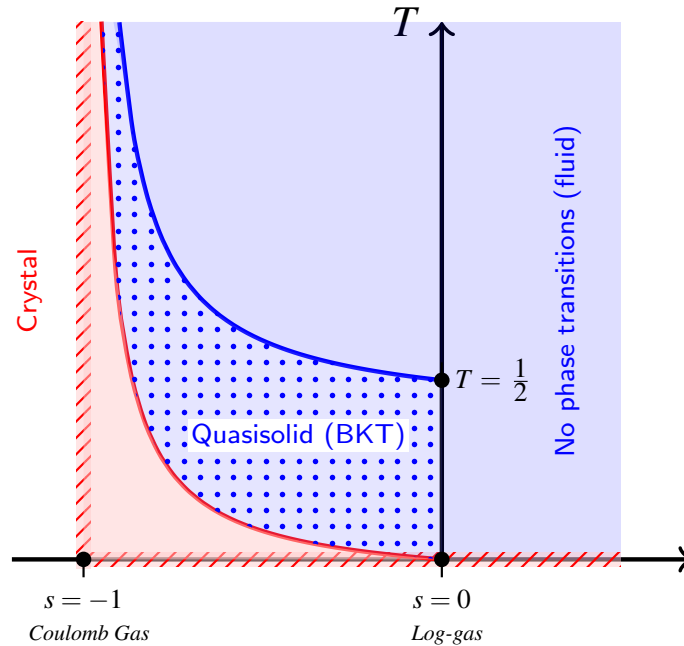


Figure 10: The phase diagram of the one-dimensional Riesz gas for the exponents  $-1 < s < 0$  obtained according to the numerical results presented in Chapter 3. The shape is consistent with the intuition of [Lew22a], namely that there is a critical temperature which separates between a solid phase and a disordered phase, consistent with the phase diagrams of the Coulomb gas  $s = -1$  and the log-gas  $s = 0$ . We also find the existence of a BKT-type transition, corresponding to the formation of a quasi-solid, and which coincide in the limit  $s \rightarrow 0$  with the BKT transition point of the log-gas.

vanish. However, this is in practice difficult to verify in the case of a numerical calculation where  $N < \infty$ . In Figure 11, the image on the left shows the function  $g(r)$  obtained by our algorithm at three different temperatures for  $s = -0.5$ . We see that for the highest temperature, the pair correlation  $g(r)$  converges rapidly towards the average density, which here is set to unity, as in a fluid. For the other two temperatures, on the other hand, we see that  $g(r)$  is an oscillating function. However, it is difficult to determine whether these oscillations eventually disappear when  $r \rightarrow \infty$ , and therefore to decide whether we are in the presence of a solid or quasi-solid phase. This is reminiscent of the problem that gave rise to the numerous historical controversies in dimension  $d = 2$  as discussed above.

To better understand this transition, we therefore study the *structure factor*  $S(k)$  [HM13, Chap. 4]. This quantity is defined as the Fourier transform of the

truncated correlation function  $g(r) - 1$ , that is

$$S(k) := 1 + \frac{1}{2\pi} \int_{\mathbb{R}} e^{ikr} (g(r) - 1) dr. \quad (106)$$

The function  $S(k)$  is often preferred in condensed matter physics and crystallography to  $g(r)$  because it is more easily accessible experimentally. In practice, the pair correlation is calculated as the inverse Fourier transform of  $S(k)$ . The structure factor is linked to the scattering properties of the material [DB08; GH04; Sir+89; Yar+73]. For a (quasi)-solid of characteristic length  $a > 0$ , we can access the geometry of the crystal lattice by scattering waves of wavelength  $\lambda \gg a$ , or equivalently of small wavenumber  $k \ll a^{-1}$ .

If the correlation function has a periodic component, the structure factor should have a Dirac peak associated with the period. In the case where the average density is 1, this peak must be therefore be at  $k = 1$ . In the case of a quasi-solid where the oscillations disappear at large distance, for example in the case of the log-gas, the structure factor should also have a peak at  $k = 1$ , but it should be an integrable singularity rather than a Dirac mass. In fact, in the case of the log-gas, it follows from the behaviour of  $g(r)$  conjectured by Forrester which was recalled above in (105), that there are three different regimes. Above the BKT transition  $\beta < 2$ , the function  $S(k)$  is monotonic near  $k = 1$ . Below the BKT transition and  $2 < \beta < 4$ , the function  $S(k)$  is no longer monotonic in  $k = 1$ , but it is still bounded. Finally, for  $\beta > 4$ , the function  $S(k)$  diverges in  $k = 1$  — logarithmically for  $\beta = 4$  and as  $|1 - k|^{4/\beta - 1}$  for  $\beta > 4$ . What is important to note here is that this singularity remains integrable. This is different from a solid, where we expect  $S(k)$  to have a Dirac mass in  $k = 1$ .

There is a non-trivial link between symmetry breaking and the behaviour of  $S(k)$  near the origin. This link *a priori* only holds for one-dimensional translation-invariant systems. Indeed, Aizenmann, Goldstein and Lebowitz [AGL01] gave a sufficient condition for symmetry breaking to occur. This result, which is related to the notion of *hyperuniformity* [TS03; Tor18], states that if  $S(k)$  behaves like  $C|k|^\eta$  when  $k \rightarrow 0$  for a certain constant  $C > 0$  and for an exponent  $\eta > 1$ , then necessarily invariance by translation must be broken. This is consistent with the Coulomb gas, for which  $\eta = 2$ . For the log-gas, it follows from Forrester's conjecture (105) that  $\eta = 1$ . In general, it seems that the natural exponent is  $\eta = 1 - s$ . This is known in the case  $0 < s < 1$  according to the results obtained recently by Boursier [Bou22]. Forrester has moreover given in [For10, Chap. 11] — see also [AM85] — a heuristic in the case  $s = 0$  which easily extends to the general case  $s$ , and which we reproduce in Chapter 3. However, this argument is based on the premise that is essentially equivalent to crystallisation. Therefore, we conjecture instead that *if the Riesz gas is crystallised, then necessarily  $\eta = 1 - s$ .*

From these different criteria — (non-)oscillations of  $g(r)$ , (non-)monotonicity and/or singularity of  $S(k)$  in  $k = 1$ , and the behaviour of  $S(k)$  near the origin — it is possible to determine numerically the phase diagram that we presented in a schematic manner in Figure 10. If we return to Figure 11, the image on the right shows the structure factor  $S(k)$  for our three different temperatures. For the temperature  $T = 1$ , the function  $S(k)$  behaves linearly at the origin  $k = 0$

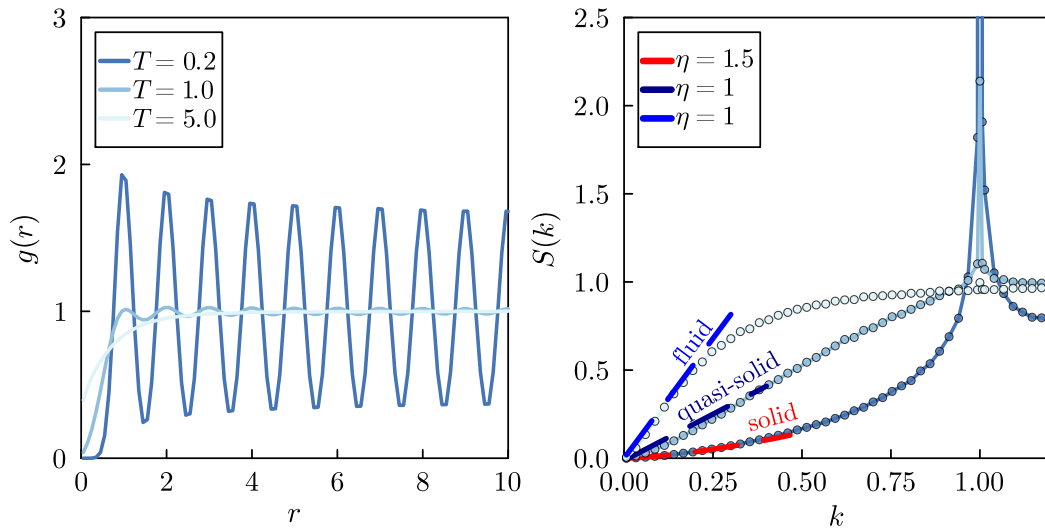


Figure 11: We display the pair correlation  $g(r)$  (left) for the Riesz gas with exponent  $s = -0.5$  obtained by our algorithm, and the structure factor  $S(k)$  determined from the Fourier transform of  $g(r) - 1$  (right). We can see that for the temperature  $T = 5$ , the function  $g(r)$  converges rapidly and monotonically towards the average density  $\rho$  of our system, which here is set to  $\rho = 1$ , as in a fluid. The structure factor does *not* have peak at  $k = 1$ . On the other hand, at lower temperatures, the function  $g(r)$  oscillates and a peak appears at  $k = 1$  for  $S(k)$ . For  $T = 1$ , the function  $S(k)$  behaves linearly when  $k = 0$ , suggesting that the system is a quasi-solid. For  $T = 0.5$ , it flattens near the origin and behaves like  $C|k|^{1-s}$ , and the peak at  $k = 1$  resemble a Dirac mass. This suggest that the system is solid according to Aizenman *et al.* [AGL01].

and has a peak at  $k = 1$ , which seems to indicate that at this temperature the Riesz gas is a quasi-solid. On the contrary, for  $T = 0.2$ , the function  $S(k)$  behaves like  $C|k|^{1-s}$  when  $k \rightarrow 0$  for a certain constant  $C > 0$ , which seems to indicate that the system is a solid at this temperature according to [AGL01].

This leads us to conjecture the existence of two critical temperatures. The first, which we denote  $\tilde{T}_s$ , corresponds to a BKT transition, and the second critical temperature, denoted  $T_s$ , corresponds to the solid transition. Using the various criteria mentioned above, we can draw an approximate phase diagram as a function of the temperature  $T$  and the exponent  $s$ , shown in Figure 12. Determining precisely the two temperatures  $T_s$  and  $\tilde{T}_s$  is complicated by the approximate nature of our numerics. However, regardless of this arbitrariness, it seems to us that the results do qualitatively show the existence of a phase transition for the Riesz gases of exponent  $-1 < s < 0$ , whose behaviour is consistent with the phase diagrams of the Coulomb gas and the log-gas, thus answering in the affirmative the question raised by Lewin in [Lew22a].

To check that our algorithm works, we also tested it on the log-gas. This allows us to confirm Forrester's conjecture (105), in particular for what it implies on the behaviour of the structure factor  $S(k)$  close to  $k = 1$  and the origin  $k = 0$ . These numerics may be of independent interest to some readers.

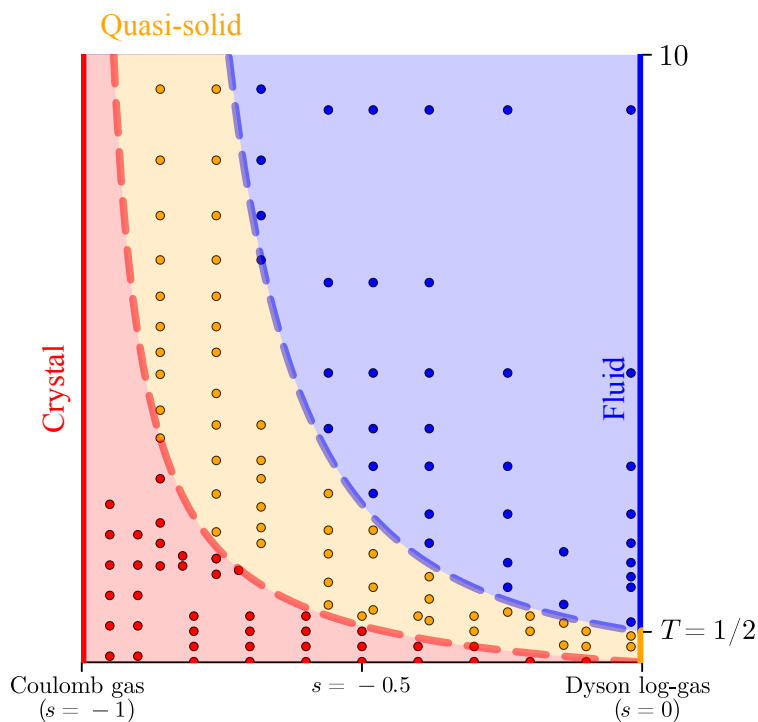


Figure 12: Phase diagram of the Riesz gas in one dimension as a function of the temperature  $T$  and the exponent  $-1 < s < 0$ . The blue dots correspond to the liquid phase. The orange dots correspond to the quasi-solid phase, for which the structure factor has a peak at  $k = 1$  but behaves linearly when  $k \rightarrow 0$ . Finally, the red dots correspond to the solid phase, for which  $S(k)$  behaves sublinearly when  $k \rightarrow 0$ . We then draw  $T_s$  and  $\tilde{T}_s$  schematically.

# CHAPTER 1

## AN EXTERNAL CHARGE APPROACH TO THE OPTIMAL TRANSPORT WITH COULOMB COST

*This chapter is an (slightly) extended version of our paper An external dual charge approach to the optimal transport with Coulomb cost [Lel22a] that was accepted for publication in ESAIM: Control, Optimization and Calculus of Variations. We prove that the Kantorovich potential associated to the multimarginal optimal transport with Coulomb cost is an electrostatic potential induced by an external charge density, which we call the dual charge. We study its properties and use it to discretize the potential in one and three space dimensions.*

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## §1.1. Introduction

In the recent years, *Multimarginal Optimal Transport* (MOT) has begun to attract considerable attention, due to a wide variety of emerging applications outside of mathematics, such as economics, finance, physics and image processing. We refer to [Pas15] for a detailed review and the citations therein. As such, it has become of valuable importance to develop numerical methods to solve this problem, which is plagued with the infamous *curse of dimensionality*.

In physics, MOT appears in a variety of applications, *e.g.* in defining the *Uniform Electron Gas* [LLS18], which in turn serves as a building block for the very important *Local Density Approximation* in *Density Functional Theory* (DFT) [LLS20; LLS19b], a successful computational modeling method in quantum physics. Another example of MOT in physics, and closely related to DFT, is the paradigm of *classical DFT* [YFG76] which exactly reformulates as a MOT problem together with *entropic regularization*.

From a numerical viewpoint, MOT is notoriously hard to solve. Most algorithms require exponential time in the number of marginals and in the discretization size of their supports. While for some specific costs, it is possible to fashion polynomial time methods, in the case of the Coulomb cost the problem is known to be  $\mathcal{NP}$ -hard [ABA21]. Current methods then consist in either adding an entropic regularization [BCN16], which physically translates into adding positive temperature to the system, and which makes the numerics more tractable by heavily exploiting parallel computing; or using ideas from *Linear Programming* (*e.g.* leveraging on the sparsity of optimal plans [FSV22]). One can also mention the approach proposed in [ACE22; Alf+20], namely the *Moment Constrained Optimal Transport*, which relies on a neat relaxation of the primal constraints. We will see that this approach is closely related to our own, see Remark 4.

From the point of view of physics, the Kantorovich dual of the MOT is a very meaningful and interesting object in its own right. In fact, as readily noticed in the literature, the so-called Kantorovich potential can be interpreted as an external potential which forces the particles to live, at equilibrium and zero temperature, on the support of an optimal plan. Moreover, at positive temperature, the dual relates to physics still, and the (unique up to an additive constant) Kantorovich potential is interpreted, from the point of view of statistical mechanics, as the external potential which forces the associated Gibbs



probability measure to have the target density of the MOT as its one-particle density.

In this paper, we suggest to parametrize the Kantorovich potential (both at zero and positive temperature) as an electrostatic potential generated by an external charge distribution, which we call the *dual charge*. While such an approach has already been used numerically at zero temperature in [ML13], here we mainly focus on its theoretical aspects. Moreover, from a numerical viewpoint, this dual charge seems a rather amenable candidate for discretization, as it allows for a natural “smoothing” from the discretization space to the space of potentials.

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## §1.2. Theoretical properties of the dual charge

### §1.2.1. Transport at zero temperature

In what follows, we fix an absolutely continuous density  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  where  $N \geq 2$  and  $d \geq 3$ , representing the expected charge density of a system of  $N$  indistinguishable and classical electrons interacting through the Coulomb potential. Among all symmetric  $N$ -particle probability measures  $\mathbb{P}$  such that  $\rho_{\mathbb{P}} = \rho$ , where

$$\rho_{\mathbb{P}}(x) = N \int_{\mathbb{R}^{d(N-1)}} \mathbb{P}(x, dx_2, \dots, dx_N),$$

we want to determine the one(s) yielding the lowest possible electrostatic energy, that is, we seek to solve the following minimization problem

$$F_{\text{SCE}}(\rho) = \inf_{\rho_{\mathbb{P}} = \rho} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^{d-2}} \mathbb{P}(dx_1, \dots, dx_N) \right\}. \quad (\text{SCE})$$

The Problem SCE is known in the physics literature as the *Strictly-Correlated Electrons* (SCE) [SPL99; Sei99]. The functional  $\rho \mapsto F_{\text{SCE}}(\rho)$  arises in DFT as the semiclassical limit of the celebrated *Levy-Lieb functional*  $F_h(\rho)$  [Lev79; Lie83; Lew18; CFK13; BDP17], and is used in the context of strongly-correlated systems [MGG12; PLK13; Mal+14; MMGG14; GVGG18; Gro+20; Mar+22]. We recall that the Levy–Lieb functional  $F_h(\rho)$ , which is the starting point of DFT, reads

$$F_h(\rho) := \inf_{\rho_{|\Psi|^2} = \rho} \left\{ \hbar^2 \int_{\mathbb{R}^{dN}} \sum_{i=1}^N |\nabla_{x_i} \Psi|^2 + \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^{d-2}} |\Psi|^2(x_1, \dots, x_N) \right\}. \quad (\text{DFT})$$

Here  $\Psi$  is a so-called *wavefunction*, that is a function  $\Psi \in L^2(\mathbb{R}^d)$  normalized such that  $\int_{\mathbb{R}^{dN}} |\Psi|^2 = 1$  and satisfying some additional symmetry conditions. In the formalism of quantum mechanics, the quantity  $\mathbb{P} = |\Psi|^2$  is the probability measure over the space  $(\mathbb{R}^d)^N$  describing the spatial distribution of the particles. The first term in the above minimization corresponds to the kinetic energy of  $\Psi$  while the second corresponds to the interaction energy. We therefore seek to minimize the total kinetic *plus* interaction energy with respect to  $\Psi$  under the constraint  $\rho_{|\Psi|^2} = \rho$ . The **Problem DFT** is similar to **Problem SCE** but with an additional kinetic term accounting for the quantum nature of the system, and the latter is the classical version of the former obtained in the semiclassical limit  $\hbar \rightarrow 0$  [CFK13; BDP17; Lew18]. As such, the SCE approach is to be thought as the exact counterpart to the *Kohn-Sham* (KS) approach [KS65], where one seeks to map a many-body system of interacting electrons into another (fictitious) system of *non-interacting* electrons with the same electronic density, whereas in the SCE approach, the fictitious system is purported to have zero kinetic energy. We refer the reader to the recent survey [FGGG22] for further details regarding **Problem SCE**.

As by now well-established [BDPGG12; CFK13], the SCE problem reformulates as a multimarginal optimal transport problem with all marginals constrained to  $\rho/N$  and cost of transportation given by the *Coulomb cost*

$$c(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^{d-2}}.$$

In particular, **Problem SCE** is equivalent to the following maximization problem [BCDP18], the so-called Kantorovich dual, which reads

$$F_{\text{SCE}}(\rho) = \sup_{\substack{V \text{ s.t.} \\ \int_{\mathbb{R}^d} |V| \rho < +\infty}} \left\{ E_N(V) + \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}, \quad (\text{SCE}_D)$$

where the infimum runs over all continuous functions  $V$  and where  $E_N(V)$  is defined as

$$E_N(V) = \inf_{x_1, \dots, x_N} \left\{ c(x_1, \dots, x_N) - \sum_{i=1}^N V(x_i) \right\}.$$

*Remark 1.* From a numerical viewpoint  $E_N(V)$  is intractable. Instead, one can consider the following problem, which is equivalent to **Problem SCE<sub>D</sub>**:

$$F_{\text{SCE}}(\rho) = \sup_{\substack{V \text{ s.t.} \\ \int_{\Sigma} |V| \rho < +\infty}} \left\{ E_{N,\Sigma}(V) + \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}, \quad (\text{SCE}_{D,\Sigma})$$

where  $E_{N,\Sigma}(V)$  is defined for any continuous  $V$  similarly to  $E_N(V)$  only with the particles constrained to the support  $\Sigma$  of  $\rho$ , that is

$$E_{N,\Sigma}(V) = \inf_{x_1, \dots, x_N \in \Sigma} \left\{ c(x_1, \dots, x_N) - \sum_{i=1}^N V(x_i) \right\}.$$

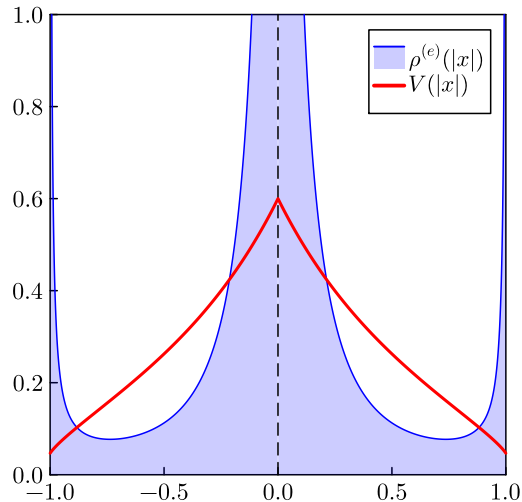


Figure 1.1: Example for Theorem 1: the (radial component of a) Kantorovich potential  $V(x) = \rho^{(e)} * |x|^{-1}$  and its associated dual charge  $\rho^{(e)}$  for a two-electron system ( $N = 2$ ) with density  $\rho = 2|B_1|^{-1}\mathbf{1}_{B_1}$  where  $B_1$  is the unit ball of  $\mathbb{R}^3$ .

A maximizer  $V$  of **Problem SCE<sub>D</sub>** is called a *Kantorovich potential*. Notice that (minus)  $V$  can be regarded as a physical potential, and that  $E_N(V)$  is to be understood as the classical counterpart to the ground-state energy in quantum physics. In fact, in the physics literature,  $V$  is coined as the *SCE potential*, which is to be thought as the effective one-body potential which emulates the SCE system, and which captures the effects of the strongly-correlated regime.

Since **Problem SCE** entirely pertains to electrostatics, one might conjecture that  $V$  shall also relate to electrostatics. More precisely, we ask ourselves whether or not the Kantorovich potential  $V$  can be written as a *Coulomb potential*, that is if there exists a measure  $\rho^{(e)} \in \mathcal{M}(\mathbb{R}^d)$ , which we call the (*external*) *dual charge*, so that  $V(x) = \rho^{(e)} * |x|^{2-d}$ , where  $*$  denotes the usual convolution operator. We would expect that  $\rho^{(e)}$  verifies  $\rho^{(e)} \geq 0$  and  $\text{supp}(\rho^{(e)}) \subset \text{supp}(\rho)$ , in order to attract the electrons into an optimal transport plan. Indeed, in the absence of an external potential landscape, the particles would escape to infinity owing to the repulsive nature of the Coulomb potential. That  $\rho^{(e)}$  be a positive measure ensures that the particles will be bonded to some region near the support of  $\rho^{(e)}$ . Furthermore, because we want the particles to place themselves in such a way that  $\rho_{\mathbb{P}}$  be equal of  $\rho$ , it seems natural to demand that  $\text{supp}(\rho^{(e)}) \subset \text{supp}(\rho)$ . Finally, we expect that  $\int_{\mathbb{R}^d} \rho^{(e)} = N - 1$ , since each fixed electron “sees” the  $N - 1$  other electrons, so as to counterbalance the repulsive force created by those electrons.

Our first theorem confirms this intuition:

**Theorem 1** (Existence of a dual charge at zero temperature). *Given  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$ , there exists a maximizer  $V$  of **Problem SCE<sub>D</sub>** which is an attractive Coulomb potential, that is of the form  $V(x) = \rho^{(e)} * |x|^{2-d}$  for some positive measure  $\rho^{(e)}$  of mass  $\int_{\mathbb{R}^d} \rho^{(e)} \leq N - 1$ . Moreover, if the support*

$\Sigma$  of  $\rho$  is bounded, one can suppose that  $\text{supp}(\rho^{(e)}) \subset \Sigma$  and that  $\int_{\Sigma} \rho^{(e)} = N - 1$ .

The proof of Theorem 1 is consigned in § 1.4.2.

*Remark 2* (Non-uniqueness of dual charges). Let  $\rho^{(e)}$  be a dual charge as in Theorem 1, *i.e.*  $V(x) = \rho^{(e)} * |x|^{2-d}$  is a Kantorovich potential for **Problem SCE<sub>D</sub>**. If the support  $\Sigma$  of  $\rho$  is bounded, *i.e.*  $\Sigma \subset B_R$  for some  $R > 0$  where  $B_R$  denotes the ball of radius  $R$ , then  $\mu^{(e)} = \rho^{(e)} + \sigma_R$  is still an admissible dual charge, where  $\sigma_R$  is the surface measure on the sphere  $\partial B_R$ . Indeed, the charge  $\sigma_R$  generates a constant potential  $c_R = R^{2-d}$  inside of  $B_R$  and  $\sigma_R * |x|^{2-d} \leq c_R$  outside of  $B_R$ , so that  $W(x) = \mu^{(e)} * |x|^{2-d}$  is still a Kantorovich potential, since

$$\int_{\mathbb{R}^d} V\rho + E_N(V) = \int_{\mathbb{R}^d} W\rho + \underbrace{E_N(W + \frac{c_R}{N})}_{\leq E_N(W)}. \quad (1.1)$$

This shows that dual charges need not be unique. Moreover, by considering  $\rho^{(e)} + c\sigma_R$  with  $c > 0$ , the mass of  $\rho^{(e)}$  can be made arbitrarily large. In fact, even restricting our attention to the dual charges supported on  $\Sigma$ , the mass  $\int_{\Sigma} \rho^{(e)}$  can also be made arbitrarily large, by considering  $\rho^{(e)} + c\chi_{\text{eq}}$  where  $\chi_{\text{eq}} \in \mathcal{M}_+(\Sigma)$  is the *equilibrium measure* of  $\Sigma$  (see [Lan72, Chap II.]). We note that generically (*i.e.* for « nice » domains  $\Sigma$ ),  $\chi_{\text{eq}}$  is supported on  $\partial\Sigma$ .

*Remark 3.* In the recent survey [FGGG22], it is mentioned as a conjecture [FGGG22, Eq. (66)] that, when  $\rho$  is supported over the entire space  $\mathbb{R}^d$ , the SCE potential  $V$  shall verify the asymptotic

$$V(x) \sim \frac{N-1}{|x|^{d-2}} \quad \text{as } x \rightarrow \infty.$$

We remark that Theorem 1 implies that  $V$  is lower than the asymptotic — with equality in the case of a compactly supported density  $\rho$ , even though in this case the problem is not well-posed since  $V$  can be freely modified outside of the support of  $\rho$ . In fact, we gave a proof of the above asymptotic in the recent paper [Lel22b], which is reproduced in Chapter 2.

Though the dual charge need not be unique over the entire space, we obtain uniqueness over the support of  $\rho$ .

**Theorem 2** (Uniqueness of the dual charge). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$ . If  $\rho^{(e)}, \mu^{(e)} \in \mathcal{M}(\mathbb{R}^d)$  are two dual charges for **Problem SCE<sub>D</sub>**, then  $\rho^{(e)} = \mu^{(e)}$  over the (topological) interior of  $\Sigma$ , where  $\Sigma$  is the support of  $\rho$ . In particular, when  $\rho$  is supported everywhere on  $\mathbb{R}^d$ , the dual charge is unique.*

The proof of Theorem 2, which follows from the uniqueness of the Kantorovich potential (see § 1.4.1 on the duality theory), is consigned in § 1.4.3.

### §1.2.2. Transport at positive temperature

At positive temperature  $\beta^{-1} > 0$ , we add an entropic term to **Problem SCE** and are led to the following variational problem

$$F_{\text{SCE},\beta}(\rho) = \inf_{\rho_{\mathbb{P}} = \rho} \left\{ \int_{\mathbb{R}^{dN}} c(x_1, \dots, x_N) d\mathbb{P} + \beta^{-1} \text{Ent}(\mathbb{P} | \mu^{\otimes N}) \right\}, \quad (\text{SCE}_{\beta})$$

where  $\mu = \rho/N$  and where  $\text{Ent}(\mathbb{P}|\mu^{\otimes N})$  denotes the relative entropy of  $\mathbb{P}$  with respect to the product measure  $\mu^{\otimes N}$ . The latter is defined for all probability measures  $\mathbb{P}$  which are absolutely continuous with respect to  $\mu^{\otimes N}$  with Radon-Nykodim density  $\frac{d\mathbb{P}}{d\mu^{\otimes N}}$  as

$$\text{Ent}(\mathbb{P}|\mu^{\otimes N}) = \int_{\mathbb{R}^{dN}} \frac{d\mathbb{P}}{d\mu^{\otimes N}}(x_1, \dots, x_N) \log \left( \frac{d\mathbb{P}}{d\mu^{\otimes N}}(x_1, \dots, x_N) \right) d\mu^{\otimes N}.$$

While Problem  $\text{SCE}_\beta$  is an instance of entropy-regularized OT [Lé12; GKR20], which has received growing interest in the past few years, it originally pertains to the statistical mechanics of non-uniform liquids, which shares substantial connections with the DFT of classical systems [Wu09; Sin91; Eva92]. Indeed, the functional  $\rho \mapsto F_{\text{SCE},\beta}(\rho)$  is the Legendre transform of the (canonical) Helmholtz free energy  $\mathcal{F}_\beta(V)$  (see § 1.4.4) with respect to the external potential  $V$ , that is

$$F_{\text{SCE},\beta}(\rho) = \sup_V \left\{ \mathcal{F}_\beta(V) + \int_{\mathbb{R}^d} V \rho \right\}. \quad (\text{SCE}_{D,\beta})$$

The existence of a maximizer  $V_\beta$  for Problem  $\text{SCE}_{D,\beta}$ , which is to be thought as the effective one-body potential which forces the system into the constraint density  $\rho$ , was proved under relatively weak hypotheses through variational techniques in [CCL84, Thm 2.2.]. Note that  $V_\beta$  is unique up to an additive constant by strict concavity, and that to respect the density constraint it must be that  $V_\beta$  is infinite outside of the support of  $\rho$ .

In what follows, we will make use of the following assumption, namely

$$\|\rho * |x|^{2-d}\|_{L^\infty} < \infty. \quad (\text{A})$$

It follows from Hölder's inequality that if there exists  $\varepsilon > 0$  such that  $\rho \in L^{\frac{d}{2}+\varepsilon}(\mathbb{R}^d)$ , then this assumption is verified. The counterpart of Theorem 1 at positive temperature, whose proof is consigned in § 1.4.5, reads:

**Theorem 3** (Existence of a dual charge at positive temperature). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  which verifies Assumption (A). Then, the unique (up to an additive constant) maximizer  $V_\beta$  of Problem  $\text{SCE}_{D,\beta}$  is an attractive Coulomb potential, i.e. there exists a positive measure  $\rho_\beta^{(e)}$  such that  $V_\beta(x) = \rho_\beta^{(e)} * |x|^{2-d}$  (up to an additive constant) on the support  $\Sigma$  of  $\rho$ . Moreover, if  $\Sigma$  is bounded, one can suppose that  $\text{supp}(\rho_\beta^{(e)}) \subset \Sigma$ .*

As the temperature is taken to 0, it is known that one recovers the zero-temperature problem Problem SCE [Car+17]. In the same spirit, we prove that the dual charge at positive temperature converges to a dual charge at zero temperature in the small temperature limit.

**Theorem 4** (Zero-temperature limit of the dual charge). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  which verifies Assumption (A), and suppose that the support  $\Sigma$  of  $\rho$  is bounded. Let  $\rho_\beta^{(e)}$  with  $\text{supp}(\rho_\beta^{(e)}) \subset \Sigma$  be as in Theorem 3, i.e.  $V_\beta(x) = \rho_\beta^{(e)} * |x|^{2-d}$  is the unique (up to an additive constant) maximizer of Problem  $\text{SCE}_{D,\beta}$ . Then, the sequence  $(\rho_\beta^{(e)})_{\beta>0}$  has at least one accumulation point for the vague topology on  $\mathcal{M}(\Sigma)$ , and any of its accumulation point is an external dual charge for Problem  $\text{SCE}_{D,\Sigma}$ .*

The proof of Theorem 4 is consigned in § 1.4.6.

Theorem 4 follows from a more general result, namely the convergence at the level of the Kantorovich potentials, as consigned in Theorem 5 hereafter. While it came to our attention that in [NW22] it is proved a similar (and decidedly more general) result, our convergence is stronger, though *a priori* specific to the Coulomb cost.

**Theorem 5** (Zero-temperature limit of the Kantorovich potential). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  which verifies Assumption (A), and suppose that the support  $\Sigma$  of  $\rho$  is bounded. Let  $V_\beta$  be the unique maximizer of Problem  $SCE_{D,\beta}$  which, up to an additive constant, we can suppose to verify  $\mathcal{F}_\beta(V_\beta) = 0$ . Then, the sequence  $(V_\beta)_{\beta>0}$  is bounded in  $W^{1,\infty}(\mathbb{R}^d)$  uniformly in  $\beta$  in the limit  $\beta \rightarrow \infty$ , and therefore admits at least one accumulation point  $V$ , which is necessarily a Kantorovich potential for Problem  $SCE_{D,\Sigma}$ . By uniqueness of the Kantorovich potential inside the support  $\Sigma$ , we therefore obtain*

$$V_\beta \xrightarrow[\beta \rightarrow \infty]{} V \quad \text{uniformly on every compact set } K \subset \overset{\circ}{\Sigma} .$$

The proof of Theorem 5 is consigned in § 1.4.6.

### §1.3. Numerical investigations

In [ML13], where it is introduced an analogous of our dual charge, the authors solve the multimarginal optimal transport problem with Coulomb cost in three-space dimension by discretizing the dual charge as a combination of few Gaussian functions and performing a nested unconstrained optimization on the dual ???. The functional derivative of the ground-state energy  $E_N(V)$  with respect to the potential  $V$  being intractable, they appealed to derivative-free methods for the outer optimization, while computing  $E_N(V)$  through standard quasi-Newton methods. Nevertheless, as noticed by the authors, «*the derivative-free methods are not suitable for optimizing with respect to a large number of degrees of freedom. More efficient numerical methods need to be developed in order to obtain the Kantorovich dual solution for more general systems*».

In this work, we rather consider the problem with entropy, that is at positive temperature, which is more amenable from a computational standpoint. Indeed, in this case the ground-state energy  $E_N(V)$  is replaced by the Helmholtz free energy  $\mathcal{F}_\beta(V)$  whose functional derivative with respect to the potential  $V$  is tractable. Similarly to [ML13], we solve the Kantorovich dual with entropy by discretizing the dual charge as a combination of a few basis functions. As the temperature is lowered, we will recover the zero-temperature dual charge [Car+17]. Approaching numerically the optimal transport by its entropy-regularized version has gained a lot of popularity in the recent years [PC19; Cut13; Ben+15], thanks to the efficient Sinkhorn–Kopp algorithm. In the context of the multimarginal optimal transport problem with Coulomb cost, this strategy has already been used in [BCN16; Nen16]. Nevertheless, this algorithm still has a problematic computational complexity in  $N$ , especially in space, and it is complicated to go beyond a few electrons in practice.



### §1.3.1. Discretization of the dual charge

We give ourselves a finite set of  $M$  basis functions  $B = \{\rho_i\}_{i=1,\dots,M}$  where  $\rho_i \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  for all  $i = 1, \dots, M$ , and we seek to approximate the external dual charge of **Problem SCE** $_{D,\beta}$  as a linear combination of the elements of  $B$ , that is by  $\rho^{(e)}[\boldsymbol{\omega}]$  for some set of weights  $\boldsymbol{\omega} = (\omega_i)_{i=1,\dots,M}$ , where

$$\rho^{(e)}[\boldsymbol{\omega}] := \sum_{i=1}^M \omega_i \rho_i \quad (\boldsymbol{\omega} \in \mathbb{R}^M).$$

Otherwise stated, we approximate the Kantorovich potential  $V_\beta$  at positive temperature by the potential  $V[\boldsymbol{\omega}]$  generated by the charge distribution  $\rho^{(e)}[\boldsymbol{\omega}]$ , that is by the convolution  $V[\boldsymbol{\omega}] := \rho^{(e)}[\boldsymbol{\omega}] * |x|^{2-d}$ . If we denote by  $\phi_i := \rho_i * |x|^{2-d}$  ( $i = 1, \dots, M$ ) then our approach is equivalent to the following model-reduction

$$\sup_{V \in \text{Span}((\phi_i)_{i=1,\dots,M})} \left\{ \mathcal{F}_\beta(V) + \int_{\mathbb{R}^d} V \rho \right\}. \quad (\text{D}_B)$$

*Remark 4* (Moments Constrained OT). If we denote  $B' = \{\phi_i\}_{i=1}^M$  where as above we define  $\phi_i = \rho_i * |x|^{d-2}$  for all  $i = 1, \dots, M$ , one can show that the following duality principle for **Problem D** $_B$  holds

$$(\text{D}_B) = \inf_{\mathbb{P} \in \Pi_{B'}(\rho)} \left\{ \int_{\mathbb{R}^{dN}} c(x_1, \dots, x_N) d\mathbb{P} + \beta^{-1} \text{Ent}(\mathbb{P} | \mu^{\otimes N}) \right\},$$

where as previously  $\mu = \rho/N$  and where  $\Pi_{B'}(\rho)$  is defined as the set of  $N$ -particle probability measures  $\mathbb{P}$  verifying the following moments constraints for all  $\phi_j \in B'$ :

$$\int_{\mathbb{R}^{dN}} \sum_{i=1}^N \phi_j(x_i) d\mathbb{P}(x_1, \dots, x_N) = \int_{\mathbb{R}^d} \phi_j(x) \rho(x) dx. \quad (1.2)$$

The **Problem D** $_B$  can therefore be regarded as the dual approach to the *Moment Constrained Optimal Transport* as introduced in [Alf+20; ACE22], where the marginal constraint  $\rho_{\mathbb{P}} = \rho$  is relaxed into the above set of moments constraints (1.2).

*Remark 5* (Constraining the mass of the dual charge). According to Theorem 3, the dual charge can be chosen to be a positive measure supported on the support of the marginal constraint  $\rho$ , both at zero and positive temperature. This suggests to consider  $\rho_i \geq 0$  for all  $i = 1, \dots, M$  and to constraint  $\boldsymbol{\omega} \in (\mathbb{R}_+)^M$  in (1.3.1). Moreover, in the small temperature limit the dual charge can be assumed to have total mass no greater than  $N - 1$  according to Theorem 1. In fact, it is exactly equal to  $N - 1$  [Lel22b, Cor. 2]. Therefore, it seems natural to consider the following constrained version of **Problem D** $_B$ :

$$\sup_{\boldsymbol{\omega} \in \Delta_B} \left\{ \mathcal{F}_\beta(V[\boldsymbol{\omega}]) + \int_{\mathbb{R}^d} V[\boldsymbol{\omega}] \rho \right\}$$

where  $\Delta_B$  is the convex set defined as

$$\Delta_B = \left\{ \boldsymbol{\omega} \in (\mathbb{R}_+)^M : \sum_{i=1}^M \omega_i \int_{\mathbb{R}^d} \rho_i \leq N - 1 \right\}.$$



This is particularly interesting as it allows us to restrict our optimization onto a compact convex set, thus ensuring the existence of an optimizer. Nevertheless, the optimum  $\boldsymbol{\omega}^*$  of **Problem D<sub>B</sub>** need not be in  $\Delta_B$ . In fact, one should dismiss this additional constraint when dealing with rather crude discretisations, *i.e.* smalls  $M$ , as it might yield far-from-optimal results.

We denote by  $\mathbb{D}(\boldsymbol{\omega})$  the objective of **Problem D<sub>B</sub>**, where  $\boldsymbol{\omega}$  is the decomposition weights of the dual charge onto the basis  $B = \{\rho_i\}_{i=1,\dots,M}$  as in (1.3.1). We note that  $\mathbb{D}$  is a strictly concave and Lipschitz-smooth function of  $\boldsymbol{\omega}$ . We can solve **Problem D<sub>B</sub>** using classical gradient ascent maximization algorithms with constant stepsize. A straightforward computation shows that for all  $i = 1, \dots, M$ , once again denoting  $\phi_i = \rho_i * |x|^{2-d}$ , we have

$$\partial_{\omega_i} \mathbb{D}(\boldsymbol{\omega}) = \int_{\mathbb{R}^d} \phi_i \rho - \int_{\mathbb{R}^d} \phi_i \rho_{\boldsymbol{\omega}} \quad (1.3)$$

where  $\rho_{\boldsymbol{\omega}}$  denotes the one-particle density of the Gibbs measure  $\mathbb{P}_{\beta, \boldsymbol{\omega}}$  whose density with respect to  $\rho^{\otimes N}/N!$  is given by

$$\mathbb{P}_{\beta, \boldsymbol{\omega}}(x_1, \dots, x_N) = Z_{\beta, \boldsymbol{\omega}}^{-1} \exp \left( -\beta c(x_1, \dots, x_N) + \beta \sum_{i=1}^N V[\boldsymbol{\omega}](x_i) \right) \quad (1.4)$$

where  $Z_{\beta, \boldsymbol{\omega}}$  is the constant ensuring that  $\mathbb{P}_{\beta, \boldsymbol{\omega}}$  is a probability measure. From a numerical perspective, the integral  $\int_{\mathbb{R}^d} \phi_i \rho$  can easily be computed for all  $i = 1, \dots, M$ . In fact, all the computational burden of computing the gradient (1.3) is hidden in the integral involving  $\rho_{\boldsymbol{\omega}}$ . Nevertheless, we notice that, by definition of  $\rho_{\boldsymbol{\omega}}$  as the one-particle density of  $\mathbb{P}_{\beta, \boldsymbol{\omega}}$ , we have

$$\int_{\mathbb{R}^d} \phi_i \rho_{\boldsymbol{\omega}} = \int_{\mathbb{R}^{dN}} \left( \sum_{j=1}^N \phi_i(x_j) \right) d\mathbb{P}_{\beta, \boldsymbol{\omega}}(x_1, \dots, x_N) \quad (1.5)$$

Although intractable analytically, we can appeal to Monte–Carlo methods to compute this quantity. At high temperature  $\beta \ll 1$  and in the compactly-supported case, one can resort to vanilla Monte–Carlo with uniform proposals over the support of  $\rho$ . At low temperature  $\beta \gg 1$ , as the ensemble crystallizes onto a minimizer of **Problem SCE**, one needs to resort to fancier sampling methods since minimizers are believed to be generically supported on low-dimensional manifolds [LRS10; CLS07]

### §1.3.2. Numerical investigations

#### One-dimensional case

We start as a sanity check with a one-dimensional system, *i.e.*  $d = 1$ . This problem is amenable for numerical investigations, as it is entirely solvable for all  $N \in \mathbb{N}$ , as stated in Theorem 6 below. Recall that, in one-dimension, the Coulomb potential is given by  $-|x - x'|$ . We have not considered this specific case (nor the two-dimensional case where the Coulomb potential is given by  $-\ln|x - x'|$ ) in our main theorems to avoid potential problems due to the divergence of both potentials at infinity. Nevertheless, we expect similar results to hold (see Remark 8 below).

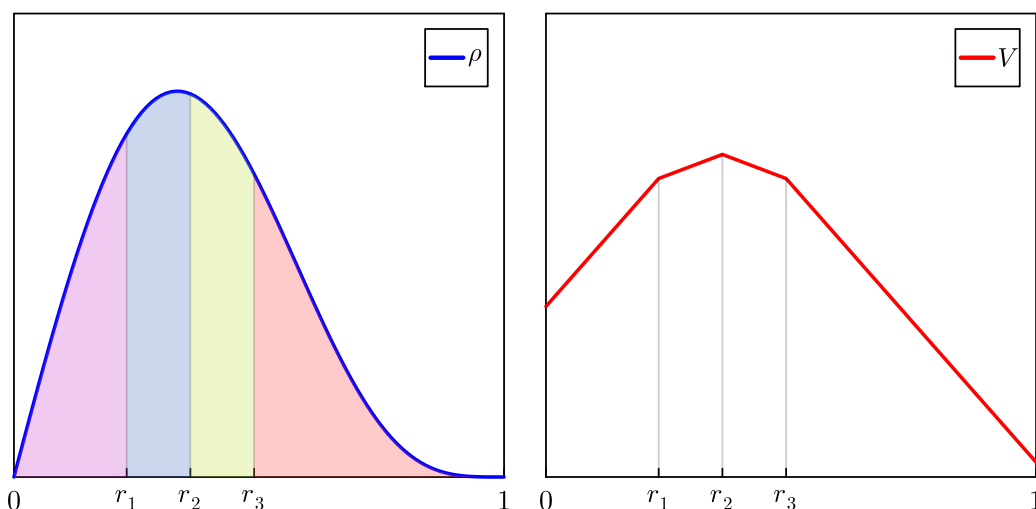


Figure 1.2: Example for Theorem 6 :  $\rho(r) \propto r(1 - r^2)^4$  on  $[0, 1]$  and the corresponding (up to an additive constant) optimal Kantorovich potential  $V$  for  $N = 4$ .

**Theorem 6** ([CDPDM15, Thm. 1.1]). *Let  $\rho \in L^1(\mathbb{R}, \mathbb{R}_+)$  with  $\int_{\mathbb{R}} \rho = N \in \mathbb{N}$ , and suppose that  $\int_{\mathbb{R}} |r|\rho < \infty$ . Then, there exists an explicit measurable map  $t : \mathbb{R} \rightarrow \mathbb{R}$  such that, denoting  $t^{(j)}$  the  $j$ -th fold composition of  $t$  with itself, the probability measure*

$$\mathbb{P}_t(r_1, \dots, r_N) = \frac{\rho(r_1)}{N} \otimes \delta(r_2 - t(r_1)) \otimes \dots \otimes \delta(r_N - t^{(N-1)}(r_1)) \quad (1.6)$$

is a minimizer for the *Problem SCE* in dimension  $d = 1$ , that is

$$\inf_{\rho \# = \rho} \left\{ - \int_{\mathbb{R}^N} \sum_{1 \leq i < j \leq N} |r_i - r_j| d\mathbb{P}(r_1, \dots, r_N) \right\}.$$

Moreover, given  $\ell_0 < \ell_1 < \dots < \ell_{N-1} < \ell_N$  such that  $\int_{\ell_i}^{\ell_{i+1}} \rho = 1$  for all  $i = 0, \dots, N$ , with  $\ell_0 = -\infty$  and  $\ell_N = +\infty$ , the function  $V$  defined as

$$V(r) = - \left( \sum_{i=1}^{N-1} \delta_{\ell_i} \right) * |r|$$

is a Kantorovich potential for the *Problem SCE<sub>D</sub>* in  $d = 1$ . In particular, the comb  $\sum_{i=1}^{N-1} \delta_{\ell_i}$  is, according to our terminology, a dual charge, which only depends on  $\rho$  through the  $\ell_i$ 's.

*Remark 6.* The result in [CDPDM15, Thm. 1.1] does not *per se* apply to the one-dimensional Coulomb potential as claimed above, for it is not strictly convex. Nevertheless, it is straightforward to prove that  $t$  remains an optimal transport plan which in this case need not be unique. In fact, there exist optimal plans

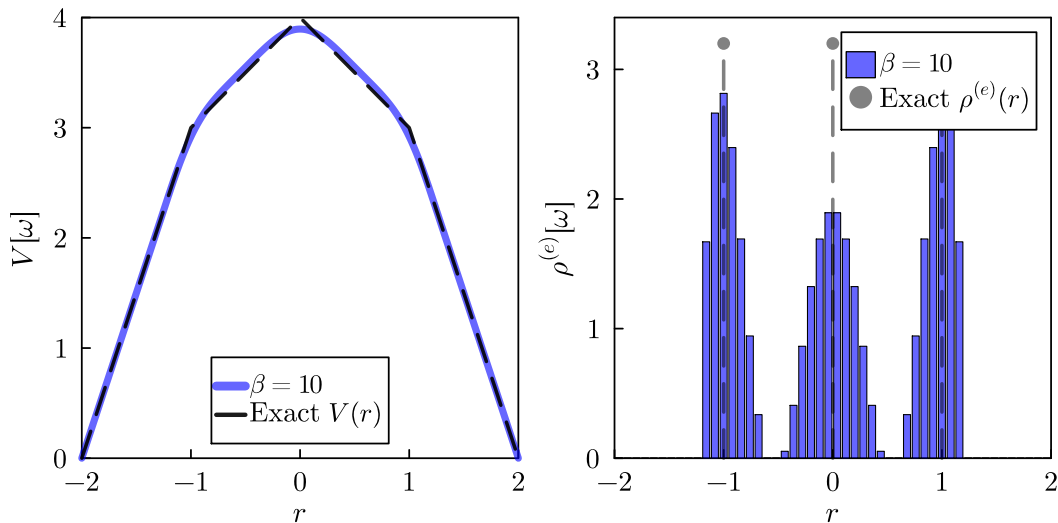


Figure 1.3: We consider the MOT problem in one-dimension with four particles  $N = 4$  where the density  $\rho$  is uniform over  $[-2, 2]$ . On the left, we display the approximate Kantorovich potential  $V[\omega^*]$  for  $\omega^*$  obtained with our algorithm at temperature  $\beta = 10$  compared to the exact Kantorovich potential given by Theorem 6. On the right, we display the associated approximate dual charge  $\rho^{(e)}[\nu^*]$  compared to the exact dual charge given by a (finite) Dirac comb.

which are not of the form (1.6) and which carry entropy. For instance, for any  $N \in \mathbb{N}$  and  $\rho = \mathbf{1}_{[-N/2, N/2]}$ , the following probability measure

$$\mathbb{P}_N = \frac{1}{N!} \sum_{\substack{i_1, \dots, i_N = 0, \dots, N-1 \\ i_k \neq i_l \text{ for } k \neq l}} \mathbf{1}_{[l_{i_1}, l_{i_1+1}]}(r_1) \otimes \dots \otimes \mathbf{1}_{[l_{i_N}, l_{i_N+1}]}(r_N) \quad (1.7)$$

is optimal. In fact, among all minimizers of **Problem SCE** for this given density  $\rho$ ,  $\mathbb{P}_N$  is the one which maximizes the entropy — see Appendix A for a proof of this statement in the general case. Therefore, if one considers the one-dimensional problem at positive temperature  $\beta^{-1}$ , then in the small temperature limit  $\beta \rightarrow \infty$ , one shall recover  $\mathbb{P}_N$ .

*Remark 7.* We see from Theorem 6 that in one-dimension **Problem SCE<sub>D</sub>** is degenerate with respect to  $\rho$ . Indeed, the maximizer  $V$  only depends on the positions  $l_i$ 's, and not on the “shape” of the density  $\rho$  inside each segment  $[l_i, l_{i+1}]$ . This pertains to the special case of the one-dimensional Coulomb force, which does not depend on the distance between the particles — *i.e.* when the electrons are consigned to their respective segment, the potential felt by them is effectively constant, so that their exact positions inside the segments are irrelevant.

We run our algorithm with  $N = 4$  at inverse temperature  $\beta = 10$  where the density  $\rho$  is uniform over  $(-2, 2)$ . We select a simple discretization where the elements of  $B$  are indicator functions of evenly-spaced segments of width  $\delta = N/M$  with  $M = 60$ , that is

$$\rho_i = \mathbf{1}_{[-N/2+i\delta, -N/2+(i+1)\delta]} \quad \text{for all } i = 0, \dots, M-1.$$

We note that, by symmetry with respect to the origin of the density  $\rho$ , we can only care about the segment  $(0, 2)$ . The results obtained, which are displayed at Figure 1.3, are consistent with the results of Theorem 6.

*Remark 8* (Extension of the main theorems to the one-dimensional case). In one-dimension, we see that Theorem 1, which we only stated for  $d \geq 3$ , remains valid according to Theorem 6. Similar uniqueness results as in Theorem 2 holds in one-dimension. At positive temperature  $\beta^{-1}$ , the existence of a dual charge as consigned in Theorem 3 also remains veracious. In fact, we even have a stronger result in the one-dimensional case, namely that the dual charge  $\rho_\beta^{(e)} = -\frac{1}{2}V_\beta''$  at positive temperature has mass  $N - 1$ , as in the zero-temperature case. Indeed, suppose that the support  $\Sigma$  of  $\rho$  is connected, that is of the form  $\Sigma = [a, b]$ . Then, we have

$$-\frac{1}{2} \int_{[a,b]} V_\beta''(r) dr = \frac{1}{2}(V_\beta'(a) - V_\beta'(b)). \quad (1.8)$$

Now, appealing to the equation verified by  $V_\beta$  (see Equation (1.53) below in the case  $d \geq 3$ ), we have

$$V_\beta'(r) = - \int_{\mathbb{R}^{N-1}} \sum_{i=2}^N \text{sgn}(r - r_i) d\overline{\mathbb{P}}_\beta^x(r_2, \dots, r_N), \quad (1.9)$$

where  $\overline{\mathbb{P}}_\beta^x$  is the (normalized) Gibbs measure defined as in (1.54) below. Therefore, we have  $V_\beta'(a) = N - 1$  and  $V_\beta'(b) = -(N - 1)$ , leading to the claimed fact. Finally, regarding the convergence of the Kantorovich potential (Theorem 5) and the dual charge (Theorem 4) at positive temperature in the small temperature limit  $\beta \rightarrow \infty$ , these results also remain veracious in one-dimension (using similar arguments).

### Three-dimensional droplets

We now consider *uniform droplets* in three space-dimension, that is where the target density  $\rho$  is taken to be uniform over a ball, which by scaling and invariance by translation we may assume to be the unit ball  $B_1$ , that is  $\rho = N|B_1|^{-1}\mathbb{1}_{B_1}$ . These uniform droplets were numerically investigated by Räsänen *et al.* in [RSGG11] up to  $N = 30$  using a construction of [SGGS07] providing a trial state  $\mathbb{P}$  for the multimarginal optimal transport problem with Coulomb cost in the case of radially symmetric marginal constraints  $\rho$ . It is known that this trial state does not reach the minimum of the optimal transport problem for all such  $\rho$ , and it is expected that the uniform droplets fall under this category. Nevertheless, it yields an upper bound which has been observed to be near-optimal. In the special two-electron case  $N = 2$ , the problem is analytically solvable using [Pas13]. The unique minimizer of ?? is then known to be

$$\frac{\rho(x_1)}{2} \otimes \delta(x_2 - T(x_1)) \quad \text{where } T(x) = -\frac{x}{|x|} (1 - |x|^3)^{1/3}. \quad (1.10)$$

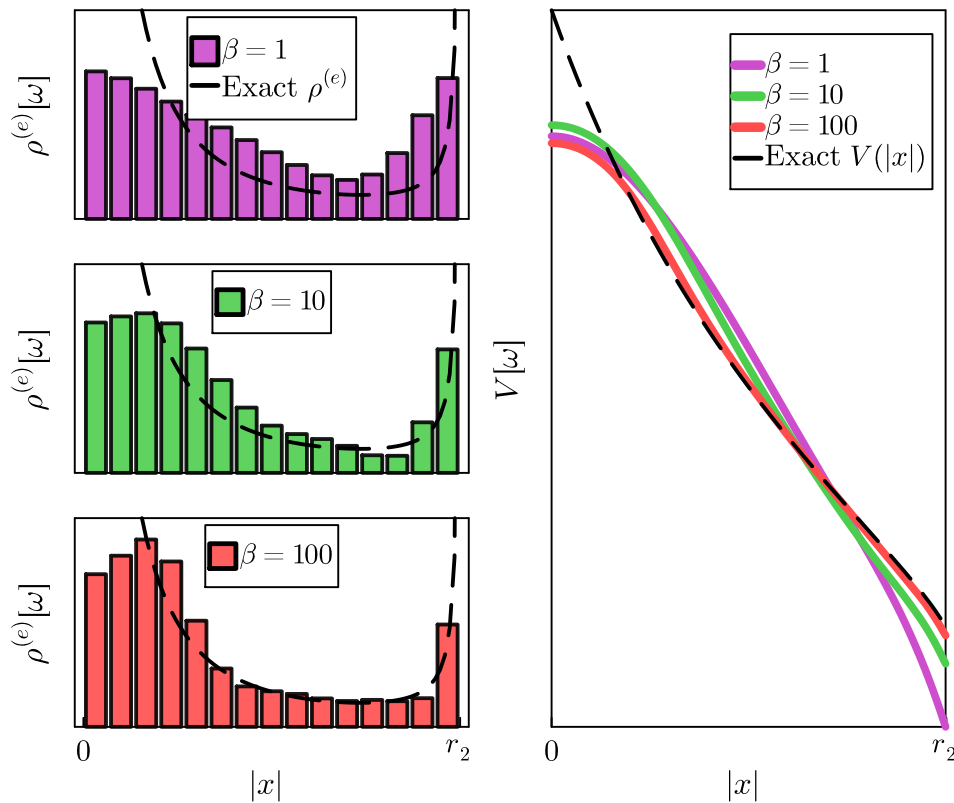


Figure 1.4: Two-electron droplet, that is  $\rho = \mathbb{1}_{B_2}$  where  $B_2 = B(0, r_2)$  is the ball centered at the origin with  $|B_2| = 2$ . The ball  $B_2$  is discretised into  $M = 15$  shells, and the dual charge is taken to be piecewise constant over these shells. We optimize the Kantorovich dual at decreasing temperature  $1/\beta$  over  $\omega = (\omega_i)_{i=1, \dots, 15}$  where  $\omega_i$  is the weight of the  $i$ -th shell. On the left, the (radial component of the) dual charges  $\rho^{(e)}[\omega]$  obtained, and on the right, the associated approximate Kantorovich potentials  $V[\omega]$ . At the temperature is decreased, we recover the exact dual charge  $\rho^{(e)}$  and Kantorovich potential  $V$  at zero temperature (dashed lines).

Given a Lipschitz Kantorovich potential  $V$  — which always exists (see § 1.4.1) — we have for almost every  $x$  in  $B_1$  that the gradient of  $V$  is given by the equation

$$\nabla V(x) = -\frac{x - T(x)}{|x - T(x)|^3}. \quad (1.11)$$

By integrating this equation with respect to  $x$ ,  $V$  can be retrieved up to an additive constant. Conversely, the dual charge  $\rho^{(e)}$  associated to  $V$ , given by  $-\Delta V(x)/4\pi$  where  $\Delta$  denotes the usual Laplacian operator, can be retrieved by taking the divergence with respect to  $x$  in the above equation, see Figure 1.1.

We start by considering the case  $N = 2$  at decreasing temperatures. The dual charge is discretized into  $M = 15$  evenly-spaced concentric shells over which the charge is constant. Otherwise stated, we approximate the dual charge by a piecewise constant function on these shells. The results obtained at

Figure 1.4 are consistent with the theory. We then fix the inverse temperature  $\beta = 50$  and we run our algorithm for an increasing number of particles up to  $N = 30$ . We use the same discretisation as for the two-particle case, except when  $N = 30$  in which case  $M$  is raised up to 25. Given the discretized version of Problem  $D_B$  at zero temperature, that is

$$F_{\text{SCE}}(\boldsymbol{\omega}) := E_N(V[\boldsymbol{\omega}]) + \int_{\mathbb{R}^d} V[\boldsymbol{\omega}]\rho,$$

we can assess the performance of our algorithm by comparing the value  $F_{\text{SCE}}(\boldsymbol{\omega})$  with the values in [RSGG11] which are upper bounds to the true optimal transport cost which are believed to be (near-)optimal. On the other hand, our values are obtained by plugging  $V[\boldsymbol{\omega}^*]$  for the optimized weight vector  $\boldsymbol{\omega}^*$  into the dual formulation of the optimal transport. Therefore, they yield lower bounds on the true cost. The dual charges are displayed in Figure 1.5, and the values of  $F_{\text{SCE}}[\boldsymbol{\omega}]$  are compared with the ones of [RSGG11]. We observe that the gap between those values is small. We notice that, even appealing to rather crude discretisation, our algorithm yields near-optimal values. The case  $N = 30$  was obtained in  $\sim 6 \text{ min}$  on a personal computer using the programming language Julia. The computations of  $F_{\text{SCE}}(\boldsymbol{\omega}^*)$  were made using the solvers of the `Optim.jl` package to determine the ground-state energy  $E_N(V[\boldsymbol{\omega}^*])$ .

### §1.3.3. Implementation details

Given the gradient  $\nabla\mathbb{D}(\boldsymbol{\omega}_t)$  of the objective at the iteration point  $\boldsymbol{\omega}_t$ , the outer optimization in Problem  $D_B$  can be conducted using first-order or quasi-Newton methods. In our experiments, we used a classical gradient ascent algorithm with fixed step-size  $1/L$  where  $L$  is a Lipschitz constant for the gradient of the objective  $\mathbb{B}$ , that is  $\|\nabla\mathbb{D}(\boldsymbol{\omega}) - \nabla\mathbb{D}(\boldsymbol{\omega}')\| \leq L\|\boldsymbol{\omega} - \boldsymbol{\omega}'\|$  for all  $\boldsymbol{\omega}, \boldsymbol{\omega}' \in \mathbb{R}^M$ . The objective  $\mathbb{D}$  being extremely expensive to compute because of the free energy term  $\mathcal{F}_\beta$ , classical line search procedures are impracticable. Note that it would be interesting to implement a procedure which bypasses the usual line searches algorithms to only enforce approximate orthogonality of the gradient with the search direction. We noticed the gradient to be very small for the weights associated to the shells near the center of the ball. This explains why the approximate dual charge does not blow up at the origin, see . Although we observe that this does not affect the quality of  $V[\boldsymbol{\omega}^*]$  as a trial state in the Kantorovich dual at zero temperature, a possible way we found effective to alleviate for this problem is to normalized the ascent directions  $\partial_{\omega_i}\mathbb{D}$  for the concerned dimensions  $\omega_i$ 's.

The inner optimization for Problem  $D_B$  consists in determining the gradient  $\nabla\mathbb{D}(\boldsymbol{\omega}_t)$ . As mentioned above, this can done using many different sampling algorithms. We used the *Metropolis Adjusted Langevin Algorithm* (MALA; see [RT96]). Evidently, an important bottleneck is that, as the temperature is decreased, it becomes harder to sample from the canonical ensemble.

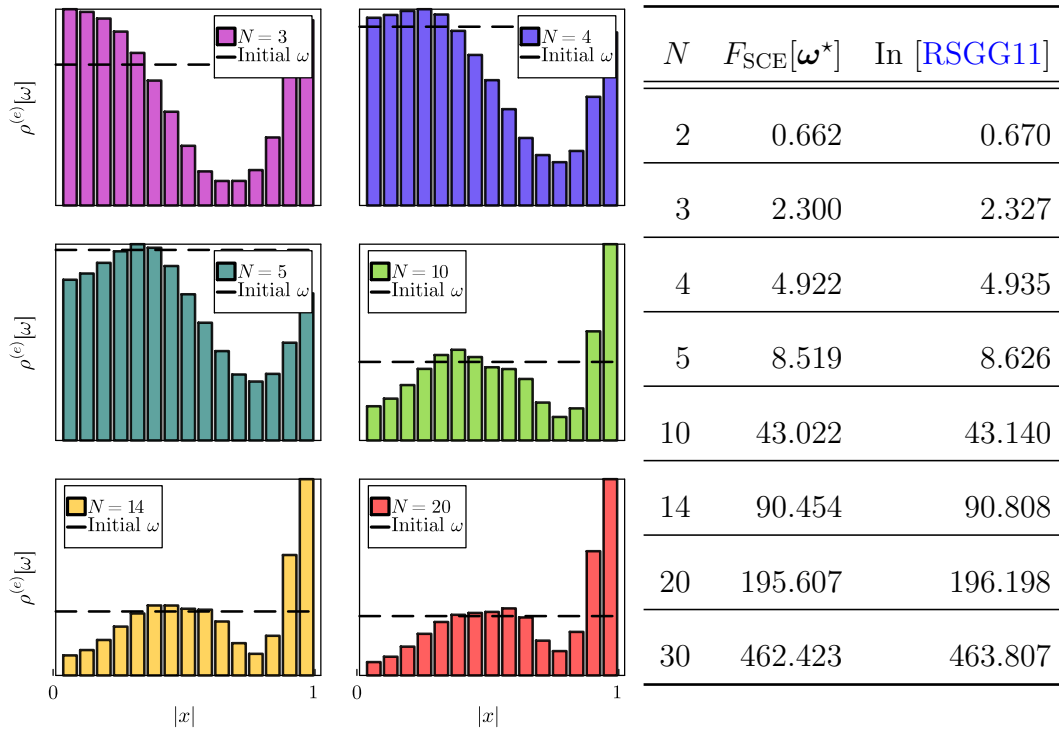


Figure 1.5: Rescaled droplets, *i.e.*  $\rho = N|B_1|^{-1}\mathbb{1}_{B_1}$  where  $B_1$  is the unit ball of  $\mathbb{R}^3$ . The inverse temperature is fixed to  $\beta = 50$ , and the number of electrons is increased up to  $N = 30$ . On the left, we display the dual charges  $\rho^{(e)}[\omega]$  obtained by optimization up to  $N = 20$ . The dashed horizontal lines represent the initial weights of the algorithm, corresponding to the uniform charge of the ball and reminiscent of the so-called *Jellium* in physics. We observe that the dual charge diverges of the boundary of the ball  $B_1$ , as expected from the exactly known dual charge for the droplet with two electrons. A similar singularity may be expected at the origin as for the two-electron case, although our algorithm was not able to detect it. On the right, we compare the total energy of the system at zero temperature obtained with our algorithm with the values in [RSGG11]. The value for  $N = 2$  in the rightmost column is exact.



## §1.4. Proofs

### §1.4.1. On the duality theory at zero temperature

We briefly recall some important facts regarding the duality theory for Problem SCE. In [BCDP18; CDMS19], it is proved that the electrons cannot overlap at optimality, in the sense that there exists  $\alpha > 0$  such that any minimizer  $\mathbb{P}$  of Problem SCE is supported away from  $D_\alpha$ , *i.e.*  $\mathbb{P}(D_\alpha) = 0$ , where

$$D_\alpha = \{(x_1, \dots, x_N) : \exists i \neq j \text{ such that } |x_i - x_j| \leq \alpha\}. \quad (1.12)$$

In particular, one can substitute to  $c$  the *truncated* Coulomb cost  $c_\alpha$ , where

$$c_\alpha(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} \min \left\{ \frac{1}{\alpha^{d-2}}, \frac{1}{|x_i - x_j|^{d-2}} \right\}. \quad (1.13)$$

Using this fact, one can prove [BCDP18, Thm. 2.6] that Problem SCE admits the following dual formulation, which is equivalent to Problem SCE<sub>D</sub>:

$$F_{\text{SCE}}(\rho) = \max_{\substack{V \text{ s.t.} \\ \int_{\mathbb{R}^d} |V| \rho < +\infty}} \left\{ E_{N,\alpha}(V) + \int_{\mathbb{R}^d} V(x) \rho(x) dx \right\}, \quad (\text{D}_\alpha)$$

where  $E_{N,\alpha}(V)$  is defined similarly to  $E_N(V)$ , only with the truncated cost  $c_\alpha$  substituted to  $c$ , that is

$$E_{N,\alpha}(V) = \inf_{x_1, \dots, x_N} \left\{ c_\alpha(x_1, \dots, x_N) - \sum_{i=1}^N V(x_i) \right\}. \quad (1.14)$$

For brevity, we drop the subscript  $\alpha$  in (1.14) from now on. Moreover, according to [BCDP18, Lem. 3.3 and Thm. 3.6], there exists a maximizer  $V$  of Problem D<sub>α</sub> which verifies

$$V(x) = \inf_{x_2, \dots, x_N} \left\{ c_\alpha(x, x_2, \dots, x_N) - \sum_{i=2}^N V(x_i) \right\}. \quad (1.15)$$

Note that, for a Kantorovich potential  $V$  which verifies (2.18), it holds that  $E_N(V) = 0$ . Moreover, we have the following lemma.

**Lemma 7.** *Let  $V$  be a Kantorovich potential for Problem SCE<sub>D</sub> verifying Equation (2.18). Then,  $V$  is Lipschitz and uniformly bounded. Moreover, it satisfies the following limit*

$$\lim_{|x| \rightarrow \infty} V(x) = E_{N-1}(V) < 0 \quad (1.16)$$

where, according to (1.14), we have

$$E_{N-1}(V) = \inf_{x_2, \dots, x_N} \left\{ c_\alpha(x_2, \dots, x_N) - \sum_{i=2}^N V(x_i) \right\}.$$

*Proof of Lemma 7.* From [BCDP18, Thm. 3.4 and Thm. 3.6], we know that  $V$  is uniformly bounded and Lipschitz. Now, using Equation (2.18), for all  $x$  we have

$$V(x) \geq \inf_{x_2, \dots, x_N} \left\{ c_\alpha(x_2, \dots, x_N) - \sum_{i=2}^N V(x_i) \right\} = E_{N-1}(V). \quad (1.17)$$

Moreover, for all  $x$  and  $(x_2, \dots, x_N)$ , we have

$$V(x) \leq c_\alpha(x, x_2, \dots, x_N) - \sum_{i=2}^N V(x_i), \quad (1.18)$$

which entails that

$$\limsup_{|x| \rightarrow \infty} V(x) \leq c_\alpha(x_2, \dots, x_N) - \sum_{i=2}^N V(x_i). \quad (1.19)$$

Taking the infimum with respect to  $x_2, \dots, x_N$  then leads to the conclusion that  $\lim_{|x| \rightarrow \infty} V(x) = E_{N-1}(V)$ . Then, letting  $|x_i| \rightarrow \infty$  for all  $i = 2, \dots, N$  in (1.18), we obtain

$$E_{N-1}(V) \leq -(N-1)E_{N-1}(V), \quad (1.20)$$

which implies that  $E_{N-1}(V) \leq 0$ . If  $E_{N-1}(V) = 0$ , then, once again appealing to (1.17) and (1.18) and letting  $|x_i| \rightarrow \infty$  for  $i = 2, \dots, N$ , we would obtain  $V \equiv 0$ , which is impossible. Therefore  $E_{N-1}(V) < 0$ .  $\square$

In fact, the Kantorovich potential for Problem  $\text{SCE}_D$  is unique [DM] on the support  $\Sigma$  of the target density  $\rho$ :

**Proposition 8.** *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$ , and let  $\Sigma$  be the support of  $\rho$ . Then, if  $V$  and  $W$  are two Kantorovich potentials for Problem  $\text{SCE}_D$ , it holds that  $V = W$  almost everywhere on  $\Sigma$  up to an additive constant (on each connected component of  $\Sigma$ ). In particular, if  $\Sigma = \mathbb{R}^d$ , the Kantorovich potential is unique.*

*Proof.* It is known that if  $V$  and  $W$  are locally Lipschitz, then they must be equal everywhere on the support of  $\Sigma$  — up to an additive constant on each connected component of  $\Sigma$ . Indeed, it follows from [San15, Thm 1.15] that, for any two almost everywhere differentiable Kantorovich potentials  $V$  and  $W$ , we have

$$\nabla V(x) = \nabla W(x) \quad \text{for almost all } x \in \Sigma, \quad (1.21)$$

where the gradients  $\nabla V$  and  $\nabla W$  are to be understood in the *Fréchet* sense. Since Lipschitz functions are differentiable almost-everywhere according to *Rademacher's theorem*, the above equality makes sense. Furthermore, it is known [Eva10] that the gradient of a Lipschitz function identifies with its *distributional* gradient. Therefore, it must be that

$$V(x) = W(x) + c, \quad \text{for all } x \in \Sigma, \quad c \in \mathbb{R}. \quad (1.22)$$

The rest of the argument is due to S. Di Marino [DM]. Let  $V$  be Kantorovich potential for Problem  $\text{SCE}_D$  — *a priori* not necessarily locally Lipschitz. We suppose without loss of generality that  $E_N(V) = 0$ . According to [BCDP18, Lem. 3.3], there always exists another Kantorovich potential  $W$  such that  $V \leq W$  everywhere and such that  $W$  verifies the equation

$$W(x) = \inf_{x_2, \dots, x_N} \left\{ c(x, x_2, \dots, x_N) - \sum_{i=2}^N W(x_i) \right\}. \quad (1.23)$$

By optimality, it must be that  $V = W$  almost everywhere on the support of  $\Sigma$ . If one can prove that  $W$  is locally Lipschitz, then  $V$  would also be locally Lipschitz and uniqueness would therefore follow according to what precedes. So far, we only know for sure that  $W$  is locally Lipschitz when one considers the truncated cost  $c_\alpha$  in the equation (1.23) instead of the singular cost  $c$ , as explained earlier. Nevertheless, as reminded in Lemma 7 above, the potential  $W$  is uniformly bounded [BCDP18, Thm. 3.4]. This entails that  $W$  verifies the above equation with  $c_\alpha$  substituted to  $c$ , and consequently that it is locally Lipschitz. Indeed, let us suppose otherwise. Then for all  $\eta > 0$  there exists  $x_\eta \in \mathbb{R}^d$  such that the infimum in the (1.23) is only attainable when two particles are  $\eta$ -close to each other. Hence

$$V(x_\eta) \geq \frac{1}{\eta^{d-2}} + \inf_{x_2, \dots, x_N} \left\{ - \sum_{i=2}^N V(x_i) \right\}. \quad (1.24)$$

Here, we have simply used the positivity of the Coulomb cost, although it essentially suffices that the cost vanishes at infinity. Finally, using that  $V$  is uniformly bounded, we obtain from the preceding inequality that

$$\frac{1}{\eta^{d-2}} \leq N \|V\|_{L^\infty} < \infty \quad (1.25)$$

so that letting  $\eta \rightarrow 0$  yields a contradiction. Therefore, there exists some threshold value  $\alpha > 0$  such that

$$W(x) = \inf_{x_2, \dots, x_N} \left\{ c_\alpha(x, x_2, \dots, x_N) - \sum_{i=2}^N W(x_i) \right\}. \quad (1.26)$$

As mentioned earlier,  $W$  is then Lipschitz as the pointwise *infimum* of uniformly Lipschitz functions. This concludes the proof of Proposition 8.  $\square$

### §1.4.2. Proof of Theorem 1

Let us turn to the proof of Theorem 1 regarding the existence of a dual charge at zero temperature. As mentioned above, there exists a Kantorovich potential  $V$  for Problem  $\text{SCE}_D$  which verifies

$$V(x) = \inf_{x_2, \dots, x_N} \left\{ c_\alpha(x, x_2, \dots, x_N) - \sum_{i=2}^N V(x_i) \right\}. \quad (1.27)$$

We will show that this particular potential arises from a charge. Indeed, for any  $x_2, \dots, x_N$ , the function

$$x \mapsto c_\alpha(x, x_2, \dots, x_N) - \sum_{i=2}^N V(x_i)$$

is superharmonic (see [Lan72, Chap I.2]) and uniformly Lipschitz in the  $x_i$ 's. Therefore, any  $V$  which verifies (1.27) remains superharmonic as the pointwise infimum of a set of uniformly Lipschitz superharmonic functions. Now, we recall the following theorem:

**Theorem 9** (Riesz's decomposition theorem [Lan72, Thm 1.24]). *Let  $f : \mathbb{R}^d \rightarrow \mathbb{R}$  be a superharmonic function which admits a harmonic minorant, i.e. there exists a harmonic function  $m : \mathbb{R}^d \rightarrow \mathbb{R}$  with  $m(x) \leq f(x)$  for all  $x \in \mathbb{R}^d$ . Then, there exists a positive measure  $\mu \in \mathcal{M}_+(\mathbb{R}^d)$  and a harmonic function  $h : \mathbb{R}^d \rightarrow \mathbb{R}$  such that*

$$f(x) = \mu * |x|^{2-d} + h(x) \quad \text{for all } x \in \mathbb{R}^d. \quad (1.28)$$

We are now ready to provide the proof of Theorem 1.

*Proof of Theorem 1.* Let us consider  $V$  a Kantorovich potential for Problem SCE<sub>D</sub> which verifies (1.27). As noted above,  $V$  is superharmonic. Moreover, according to Lemma 7,  $V$  is uniformly bounded. Therefore, according to Theorem 9 above, there exists a positive measure  $\rho^{(e)} \in \mathcal{M}_+(\mathbb{R}^d)$  and an harmonic function  $h$  such that  $V(x) = \rho^{(e)} * |x|^{2-d} + h(x)$ . But  $h$  must be bounded from above, so that *Liouville's theorem* entails that  $h$  is constant<sup>1</sup>. This provides the existence of a dual charge as stated by Theorem 1. In fact, let us prove that the constant is provided by  $E_{N-1}(V)$ , the limit of  $V$  at infinity (see Lemma 7). This is not entirely trivial, as the limit of a Coulomb potential need not to be zero. Nevertheless, we have the following result, whose proof is given below.

**Lemma 10.** *Let  $\mu \in \mathcal{M}_+(\mathbb{R}^d)$  be any locally finite measure such that  $\mu * |x|^{2-d}$  is bounded at the origin, i.e.  $\int_{\mathbb{R}^d} |x|^{2-d} \mu(dx) < \infty$ . Then*

$$\liminf_{|x| \rightarrow \infty} \mu * |x|^{2-d} = 0. \quad (1.29)$$

In fact, because  $V$  admits a limit at infinity, Lemma 10 implies the stronger result that  $\rho^{(e)} * |x|^{2-d}$  converges to zero everywhere at infinity

$$\lim_{|x| \rightarrow \infty} \rho^{(e)} * |x|^{2-d} = 0. \quad (1.30)$$

Let us prove that the total mass of the dual charge  $\rho^{(e)}$  cannot exceed  $N - 1$ . In what follows, we denote  $W(x) = \rho^{(e)} * |x|^{2-d}$ . Note that, according to (1.30) and Lemma 7, we have for all  $x \in \mathbb{R}^d$

$$V(x) = W(x) + E_{N-1}(V). \quad (1.31)$$

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<sup>1</sup>Liouville's theorem, although usually stated for bounded harmonic functions, is veracious for merely bounded from above or below harmonic functions, see [LL01, Thm. 9.5]

Moreover, for  $E_N(V) = 0$ , we have the following inequality for all  $x$  and  $(x_2, \dots, x_N)$

$$W(x) \leq c_\alpha(x, x_2, \dots, x_N) - \sum_{i=2}^N W(x_i) - E_N(W). \quad (1.32)$$

We first claim that  $E_N(W) = E_{N-1}(W)$ . Indeed, according to (1.31) and still appealing to the fact that  $E_N(V) = 0$ , we have

$$E_N(W) = NE_{N-1}(V). \quad (1.33)$$

Furthermore, we have

$$E_{N-1}(V) = E_{N-1}(W) - (N-1)E_{N-1}(V). \quad (1.34)$$

This entails that  $NE_{N-1}(V) = E_{N-1}(W)$ , and ultimately implies the claim that  $E_N(W) = E_{N-1}(W)$ . Now, because  $W$  vanishes at infinity, we have the following sequence of inequalities

$$E_{N-K}(W) \leq E_{N-K+1}(W) \quad \text{for all } K = 1, \dots, N-1, \quad (1.35)$$

where the limiting case  $K = N-1$  corresponds to the case where all electrons but one are sent to infinity, that is

$$E_1(W) = \inf_x \{-W(x)\}.$$

As the supremum of  $V$  cannot be attained at infinity, we deduce the existence of a smallest  $K \in \{1, \dots, N-1\}$  such that

$$E_{N-1}(W) = E_{N-2}(W) = \dots = E_{N-K}(W) < E_{N-K-1}(W). \quad (1.36)$$

The last inequality implies that the infimum  $E_{N-K}(W)$  is attained inside a compact set for some  $q_1, \dots, q_{N-K}$ . Now, plugging  $q_1, \dots, q_{N-K}$  into (1.32), we obtain

$$W(x) \leq \sum_{k=1}^{N-K} \frac{1}{|x - q_k|^{d-2}}, \quad \text{for all } x \in \mathbb{R}^d, \quad (1.37)$$

which entails, as claimed in Theorem 1, that

$$\int_{\mathbb{R}^d} \rho^{(e)} \leq N - K \leq N - 1. \quad (1.38)$$

Finally, for the last item of Theorem 1, namely that we can suppose  $\text{supp}(\rho^{(e)}) \subset \Sigma$  in the case where  $\Sigma$  is bounded, it suffices to consider the balayage of  $\rho^{(e)}$  onto  $\Sigma$ , as stated in the following technical theorem.

**Theorem 11** (Balayage [Lan72, Thm 4.2, Thm 4.4]). *Let  $G \subset \mathbb{R}^d$  be a region with compact boundary. Given any  $\mu \in \mathcal{M}_+(\mathbb{R}^d)$  such that  $\text{supp}(\mu) \subset \overline{G}$ , there exists a measure  $\nu \in \mathcal{M}_+(\mathbb{R}^d)$ , the so-called balayage measure of  $\mu$  onto  $\partial G$ , which verifies that  $\text{supp}(\nu) \subset \partial G$ , and that  $\nu * |x|^{2-d} = \mu * |x|^{2-d}$  for all  $x \in \mathbb{R}^d \setminus G$  and  $\nu * |x|^{d-2} \leq \mu * |x|^{d-2}$  for all  $x \in \overline{G}$ . Moreover, we have  $\int_{\mathbb{R}^d} \nu \leq \int_{\mathbb{R}^d} \mu$ .*

This concludes the proof of Theorem 1.  $\square$

We now turn to the proof of Lemma 10.

*Proof of Lemma 10.* It suffices to consider the case where  $\mu$  is radial, that is  $\mu(\mathcal{R}A) = \mu(A)$  for all Borel sets  $A \subset \mathbb{R}^d$  and all rotations  $\mathcal{R} \in SO(d)$ . Indeed, we always have

$$0 \leq \liminf_{|x| \rightarrow \infty} \mu * |x|^{2-d} \leq \liminf_{|x| \rightarrow \infty} \tilde{\mu} * |x|^{2-d} \quad (1.39)$$

where  $\tilde{\mu} \in \mathcal{M}_+(\mathbb{R}^d)$  is the radial measure defined as

$$\tilde{\mu}(A) = \int_{SO(d)} \mu(\mathcal{R}A) \nu(d\mathcal{R}). \quad (1.40)$$

where  $\nu$  is the *Haar measure* of  $SO(d)$ . Therefore, let us suppose that  $\mu$  is radial. According to *Newton's theorem* (see, e.g. [LL01, Thm 9.7]), we have

$$\mu * |x|^{2-d} = \frac{1}{|x|^{d-2}} \int_{B_{|x|}} \mu(dx') + \int_{\mathbb{R}^d \setminus B_{|x|}} \frac{\mu(dx')}{|x'|^{d-2}}. \quad (1.41)$$

Let  $(x_n)_{n \geq 0}$  be any sequence such that  $|x_n| \rightarrow \infty$  as  $n \rightarrow \infty$ , and suppose that  $x_0 = 0$ . We write  $r_n = |x_n|$  and  $\mu(r_n) = \mu(B_{r_n})$ . Up to a subsequence, since  $\mu$  is locally finite, we can suppose that

$$\mu(r_{n-1}) \leq \sqrt{r_n}, \quad \text{for all } n \geq 1. \quad (1.42)$$

But, we can write

$$\int_{\mathbb{R}^d} \frac{\mu(dx')}{|x'|^{d-2}} = \sum_{n \geq 1} \int_{B_{r_n} \setminus B_{r_{n-1}}} \frac{\mu(dx')}{|x'|^{d-2}} \geq \sum_{n \geq 1} \frac{1}{r_n} (\mu(r_n) - \mu(r_{n-1})). \quad (1.43)$$

By assumption,  $\int_{\mathbb{R}^d} |x'|^{2-d} \mu(dx') < \infty$ , so it must be that

$$\frac{1}{r_n} (\mu(r_n) - \mu(r_{n-1})) \xrightarrow{n \rightarrow \infty} 0. \quad (1.44)$$

But, according to (1.42), we have

$$\frac{1}{r_n} \mu(r_{n-1}) \leq \frac{1}{\sqrt{r_n}} \xrightarrow{n \rightarrow \infty} 0, \quad (1.45)$$

so that

$$\frac{1}{r_n} \mu(r_n) \xrightarrow{n \rightarrow \infty} 0. \quad (1.46)$$

Now, still appealing to Newton's theorem, we have

$$\mu * |x|^{2-d}(x_n) = \frac{1}{r_n} \mu(r_n) + \int_{\mathbb{R}^d \setminus B_{r_n}} \frac{\mu(dx')}{|x'|^{d-2}} \xrightarrow{n \rightarrow \infty} 0, \quad (1.47)$$

which yields the thesis of Lemma 10.  $\square$

### §1.4.3. Proof of Theorem 2

The uniqueness of the dual charge is intrinsically linked to that of the uniqueness of the Kantorovich potential. We showed at Proposition 8 that the Kantorovich potential is unique up to an additive constant on (each connected component of)  $\Sigma$ . Therefore, if  $\mu^{(e)}$  and  $\rho^{(e)}$  are two dual charges, it must be that (up to an additive constant on each connected component) we have

$$\rho^{(e)} * |r|^{2-d} = \mu^{(e)} * |r|^{2-d} \quad \text{for all } r \in \Sigma. \quad (1.48)$$

Then, by taking the *distributional Laplacian* ( $-\Delta$ ) on each side of the equality yields that  $\rho^{(e)} = \mu^{(e)}$  in the sense of positive measures  $\mathcal{M}(\Sigma)$ , thus yielding the uniqueness over the topological interior of  $\Sigma$ .

### §1.4.4. On the duality theory at positive temperature

We recall important facts regarding the duality theory for Problem  $\text{SCE}_\beta$ . As mentioned above, the SCE problem at positive temperature can be viewed as the Legendre transform

$$F_{\text{SCE},\beta}(\rho) = \sup_V \left\{ \mathcal{F}_\beta(V) + \int_{\mathbb{R}^d} V \rho \right\}, \quad (1.49)$$

where  $\mathcal{F}_\beta(V)$  is the Helmholtz free energy of the canonical ensemble with external potential (minus)  $V$ , that is

$$\mathcal{F}_\beta(V) = -\beta^{-1} \ln Z_\beta(V)$$

where  $Z_\beta(V)$ , the so-called *partition function*, is defined as the volume  $\int_{\mathbb{R}^{dN}} d\mathbb{P}_\beta(V)$  of the configurational canonical ensemble  $\mathbb{P}_\beta(V)$  defined as the *Gibbs–Boltzmann measure* whose density with respect to  $\mu^{\otimes N}$  is given by

$$d\mathbb{P}_\beta(V)(x_1, \dots, x_N) = \exp \left( -\beta c(x_1, \dots, x_N) - \beta \sum_{i=1}^N V(x_i) \right) d\mu^{\otimes N}. \quad (1.50)$$

Appealing to the strict concavity of Problem  $\text{SCE}_{D,\beta}$ , one can formally take the functional derivative of the objective and solve for the first-order optimality condition. One finds

$$\frac{\delta}{\delta V(x)} \left( \mathcal{F}_\beta(V) + \int_{\mathbb{R}^d} V \rho \right) = \rho_{\mathbb{P}_\beta(V)}(x) - \rho(x) \quad (1.51)$$

where  $\rho_{\mathbb{P}_\beta(V)}$  is the one-particle density of the ensemble  $\mathbb{P}_\beta(V)$ , that is

$$\rho_{\mathbb{P}_\beta(V)}(x) = N \int_{\mathbb{R}^{d(N-1)}} \mathbb{P}_\beta(V)(x, dx_2, \dots, dx_N). \quad (1.52)$$

Therefore Problem  $\text{SCE}_{D,\beta}$  amounts to finding the unique (up to an additive constant) potential  $V_\beta$  such that the one-particle density of the associated canonical ensemble is  $\rho$ . We will always write  $\mathbb{P}_\beta$  for  $\mathbb{P}_\beta(V_\beta)$ .



**Convention 1.** *Up to an additive constant, we will always assume that  $V_\beta$  verifies  $Z_\beta(V_\beta) = N$ .*

Note that the first-order optimality condition  $\rho_{\mathbb{P}_\beta} = \rho$  rewrites under the above convention as the fixed-point equation

$$V_\beta(x) = -\beta^{-1} \ln \int_{\mathbb{R}^{d(N-1)}} \mathbb{P}_\beta^x(x_2, \dots, x_N) d\mu^{\otimes(N-1)}(x_2, \dots, x_N) \quad (1.53)$$

for almost all  $x \in \Sigma$ , where  $\Sigma$  is the support of  $\rho$  and where  $\mathbb{P}_\beta^x$  is defined as

$$\mathbb{P}_\beta^x(x_2, \dots, x_N) = \exp \left( -\beta c(x, x_2, \dots, x_N) - \beta \sum_{i=2}^N V_\beta(x_i) \right). \quad (1.54)$$

**Convention 2.** *Since in (1.50) the Gibbs measure  $\mathbb{P}_\beta(V)$  is defined only with respect to  $\rho$ , in the first-order optimality condition (1.51) one can freely modify  $V_\beta$  outside of the support  $\Sigma$  of  $\rho$ . Furthermore, although Equation (1.53) is only valid inside of  $\Sigma$ , the right-hand side is well-defined for all  $x$  and can be used to extend  $V_\beta$  over the entire space. In what follows we will always suppose that  $V_\beta$  is defined everywhere according to (1.53).*

Finally, let us indicate that the equality at (1.49) between **Problem SCE $_\beta$**  and **Problem SCE $_{D,\beta}$**  follows from a classical theorem of Convex Analysis, see *e.g.* [Sim11, Thm 5.17], and that the above argument leading to the equation (1.53) was made rigorous in [CCL84] under the assumption that (*i.e.* see Assumption (A))

$$D(\rho) = \iint_{\mathbb{R}^d \times \mathbb{R}^d} \frac{\rho(dx)\rho(dx')}{|x - x'|^{d-2}} < \infty. \quad (1.55)$$

### §1.4.5. Proof of Theorem 3

Let us start with the following lemma:

**Lemma 12.** *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  be such that the assumptions Assumption (A) are verified. Then, the unique maximizer  $V_\beta$  of **Problem SCE $_{D,\beta}$**  is a twice continuously differentiable superharmonic function, *i.e.*  $V_\beta \in C^2(\mathbb{R}^d)$  and  $-\Delta V_\beta \geq 0$ .*

*Proof of Lemma 12.* It follows from dominated convergence that  $V_\beta$  is twice continuously differentiable. To prove that  $V_\beta$  is superharmonic, it then suffices to check that  $-\Delta V_\beta \geq 0$  [Lan72, p.56-57]. We denote by  $\overline{\mathbb{P}}_\beta^x$  the Gibbs-Boltzmann measure  $\mathbb{P}_\beta^x$  normalized to unity. We have

$$\nabla V_\beta(x) = - \int_{\mathbb{R}^{d(N-1)}} \nabla_x c(x, x_2, \dots, x_N) d\overline{\mathbb{P}}_\beta^x(x_2, \dots, x_N) \quad (1.56)$$

and, taking the divergence with respect to  $x$  above, we obtain

$$\begin{aligned} -\beta^{-1} \Delta V_\beta(x) &= \int_{\mathbb{R}^{d(N-1)}} |\nabla_x c(x, x_2, \dots, x_N)|^2 d\overline{\mathbb{P}}_\beta^x \\ &\quad - \left| \int_{\mathbb{R}^{d(N-1)}} \nabla_x c(x, x_2, \dots, x_N) d\overline{\mathbb{P}}_\beta^x \right|^2. \end{aligned} \quad (1.57)$$

The thesis is then obtained by appealing to Jensen's inequality. Note that for any  $x$ , the function  $(x_2, \dots, x_N) \mapsto \nabla_x c(x_2, \dots, x_N)$  is integrable against the Gibbs measure  $\mathbb{P}_\beta^x$  for the latter vanishes exponentially fast as  $x_j \rightarrow x$  for all  $j = 2, \dots, N$ . Therefore, the above equation makes sense for all  $x$ .  $\square$

**Lemma 13.** *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  be such that Assumption (A) are verified. Then, the unique maximizer  $V_\beta$  of **Problem SCE<sub>D,β</sub>** under Conventions 1 to 2 is bounded in  $L^\infty(\mathbb{R}^d)$  independently in  $\beta$  in the limit  $\beta \rightarrow \infty$ . More precisely, we have*

$$\sup_{x \in \mathbb{R}^d} V_\beta(x) \leq \|\rho * |x|^{2-d}\|_{L^\infty} + \frac{1}{2}D(\rho). \quad (1.58)$$

and

$$\inf_{x \in \mathbb{R}^d} V_\beta(x) \geq -(N-1)(\|\rho * |x|^{2-d}\|_{L^\infty} + \frac{1}{2}D(\rho)). \quad (1.59)$$

*Proof of Lemma 13.* The upper bound (1.58) follows by appealing to Jensen's inequality in Equation (1.53). Indeed, we have

$$V_\beta(x) \leq \int_{\mathbb{R}^{d(N-1)}} \left( c(x, x_2, \dots, x_N) - \sum_{i=2}^N V_\beta(x_i) \right) d\mu^{\otimes N-1}, \quad (1.60)$$

which implies

$$V_\beta(x) \leq (N-1)\mu * |x|^{2-d} + \binom{N-1}{2}D(\mu) - (N-1) \int_{\mathbb{R}^d} V_\beta(x) \mu(dx). \quad (1.61)$$

Now, recalling that  $\mu = \rho/N$ , we have by definition

$$F_{\text{SCE},\beta}(\rho) = -\beta^{-1} \ln Z_\beta(V_\beta) + N \int_{\mathbb{R}^d} V_\beta(x) \mu(dx). \quad (1.62)$$

Therefore, using the above equality and the fact that  $Z_\beta(V_\beta) = N$  by Convention 1, we obtain

$$V_\beta(x) \leq (N-1)\mu * |x|^{2-d} + \binom{N-1}{2}D(\mu) - \frac{(N-1)}{N}(F_{\text{SCE},\beta}(\rho) + \beta^{-1} \ln N). \quad (1.63)$$

Finally, from the fact that  $F_{\text{SCE},\beta}(\rho)$  is positive and converges to  $F_{\text{SCE}}(\rho)$  as  $\beta \rightarrow \infty$ , we obtain the upper bound (1.58) as claimed. Now using the positivity of the Coulomb cost, we have

$$V_\beta(x) \geq -\beta^{-1}(N-1) \ln \left( \int_{\mathbb{R}^d} e^{\beta V_\beta(x')} d\mu(x') \right) \quad (1.64)$$

and plugging the upper bound (1.58) into (1.64) we obtain the lower bound (1.59).  $\square$

We can now provide the proof of Theorem 3.

*Proof of Theorem 3.* From Lemma 12 and Lemma 13, we know that  $V_\beta$  (under Conventions 1 to 2) is superharmonic and bounded from below. Therefore, the proof of Theorem 3, regarding the existence of a dual charge at positive temperature, is identical to that of the proof of Theorem 1 regarding the existence of a dual charge at zero temperature.  $\square$

### §1.4.6. Proofs of Theorem 4 and Theorem 5

We start with the proof of Theorem 5 regarding the convergence of the Kantorovich potential  $V_\beta$  to a Kantorovich potential for Problem  $\text{SCE}_{D,\Sigma}$  in the limit  $\beta \rightarrow \infty$ .

**Lemma 14.** *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  be such that Assumption (A) are verified, and such that the support  $\Sigma$  of  $\rho$  is bounded. Then, the unique maximizer  $V_\beta$  of Problem  $\text{SCE}_{D,\beta}$  under Conventions 1 to 2 is bounded in  $W^{1,\infty}(\mathbb{R}^d)$  independently in  $\beta$  in the limit  $\beta \rightarrow \infty$ .*

*Proof of Lemma 14.* According to Lemma 13, we know that  $V_\beta$  is bounded in  $L^\infty(\mathbb{R}^d)$  uniformly in  $\beta$  in the small temperature limit, so that it only remains to prove that  $\nabla V_\beta$  is also bounded in  $L^\infty(\mathbb{R}^d)$  uniformly in  $\beta$  as  $\beta \rightarrow \infty$ . Recall that

$$\nabla V_\beta(x) = - \int_{\mathbb{R}^{d(N-1)}} \nabla_x c(x, x_2, \dots, x_N) d\overline{\mathbb{P}}_\beta^x(x_2, \dots, x_N) \quad (1.65)$$

where  $\overline{\mathbb{P}}_\beta^x$  is the Gibbs-Boltzmann measure  $\mathbb{P}_\beta^x$  normalized to unity. Using the fact that  $\mathbb{P}_\beta^x$  is symmetric, we have

$$|\nabla V_\beta(x)| \leq (N-1) \int_{\mathbb{R}^{d(N-1)}} \frac{d\overline{\mathbb{P}}_\beta^x(x_2, \dots, x_N)}{|x - x_2|^{d-1}}. \quad (1.66)$$

For all  $\beta > 0$  and all  $x \in \mathbb{R}^d$ , we have  $\text{supp}(\mathbb{P}_\beta^x) \subset \Sigma$ , where  $\Sigma$  is the support of  $\rho$ . Therefore, since  $\Sigma$  is by hypothesis bounded, we have

$$\int_{\mathbb{R}^{d(N-1)}} \frac{d\overline{\mathbb{P}}_\beta^x(x_2, \dots, x_N)}{|x - x_2|^{d-1}} = \frac{1}{|x|^{d-1}} + o_{|x| \rightarrow \infty}(1),$$

where the  $o_{|x| \rightarrow \infty}(1)$  is independent of  $\beta$ . Therefore, for a large enough compact set  $K$ , the gradient  $\nabla V_\beta$  is bounded in  $L^\infty(\mathbb{R}^d \setminus K)$  uniformly in  $\beta$  in the limit  $\beta \rightarrow \infty$ . It remains to control  $|\nabla V_\beta|$  inside of  $K$ .

Let us define the (unnormalized) Gibbs-Boltzmann measure  $\mathbb{Q}_\beta^x$  as

$$d\mathbb{Q}_\beta^x(x_2, \dots, x_N) = \frac{\mathbb{P}_\beta^x(x_2, \dots, x_N)}{|x - x_2|^{d-1}} d\mu^{\otimes(N-1)}(x_2, \dots, x_N).$$

and let  $\overline{\mathbb{Q}}_\beta^x$  be the associated probability measure, *i.e.*  $\mathbb{Q}_\beta^x$  normalized to unity. We denote by  $g_\beta(x)$  the volume of  $\mathbb{P}_\beta^x$ , *i.e.*  $g_\beta(x) = \int_{\mathbb{R}^{d(N-1)}} d\mathbb{P}_\beta^x$ , and by  $h_\beta(x)$  the volume of  $\mathbb{Q}_\beta^x$ . We can then rewrite

$$A(x) := \int_{\mathbb{R}^{d(N-1)}} \frac{d\overline{\mathbb{P}}_\beta^x(x_2, \dots, x_N)}{|x - x_2|^{d-1}} = \frac{h_\beta(x)}{g_\beta(x)} \quad (1.67)$$

According to the *Gibbs Variational Principle*, the free energies associated with  $\mathbb{P}_\beta^x$  and  $\mathbb{Q}_\beta^x$  can be rewritten as

$$-\beta^{-1} \ln g_\beta(x) = F(\overline{\mathbb{P}}_\beta^x) = \min_{\mathbb{P}} F(\mathbb{P}) \quad (1.68)$$

$$\text{and } -\beta^{-1} \ln h_\beta(x) = F'(\overline{\mathbb{Q}}_\beta^x) = \min_{\mathbb{P}} F'(\mathbb{P}), \quad (1.69)$$

where the minimum runs over all probability measures  $\mathbb{P}$  on  $\mathbb{R}^{d(N-1)}$  and where the functionals  $F$  and  $F'$  are defined as the total energies

$$F(\mathbb{P}) = \int_{\mathbb{R}^{d(N-1)}} \mathcal{H}^x(x_2, \dots, x_N) d\mathbb{P} + \beta^{-1} \mathbf{Ent}(\mathbb{P} | \mu^{\otimes(N-1)}) \quad (1.70)$$

$$\text{and } F'(\mathbb{P}) = F(\mathbb{P}) + \beta^{-1} \int_{\mathbb{R}^{d(N-1)}} \ln |x - x_2|^{d-1} d\mathbb{P}, \quad (1.71)$$

where  $\mathcal{H}^x$  is the Hamiltonian defined as

$$\mathcal{H}^x(x_2, \dots, x_N) = c(x, x_2, \dots, x_N) - \sum_{i=2}^N V_\beta(x_i).$$

Therefore, we obtain

$$-\beta^{-1} \ln A(x) = F'(\overline{\mathbb{Q}}_\beta^x) - F(\overline{\mathbb{P}}_\beta^x) \geq F'(\overline{\mathbb{Q}}_\beta^x) - F(\overline{\mathbb{Q}}_\beta^x), \quad (1.72)$$

and the preceding inequality rewrites as

$$A(x) \leq \exp \left( - \int_{\mathbb{R}^{d(N-1)}} \ln |x - x_2|^{d-1} d\overline{\mathbb{Q}}_\beta^x \right). \quad (1.73)$$

Using the inequality  $-\log t^{d-1} \leq t^{2-d}$  for  $t > 0$ , we then obtain

$$A(x) \leq \exp \left( \int_{\mathbb{R}^{d(N-1)}} \frac{d\overline{\mathbb{Q}}_\beta^x}{|x - x_2|^{d-2}} \right). \quad (1.74)$$

Now, let us appeal to the Gibbs Variational Principle once again to further the inequality (1.74). Indeed, for any probability measure  $\mathbb{P}$ , we have (by definition) that  $F'(\overline{\mathbb{Q}}_\beta^x) \leq F'(\mathbb{P})$ . Using the fact that the entropy and the Coulomb cost are positive, we are led to

$$\begin{aligned} \int_{\mathbb{R}^{d(N-1)}} \frac{d\overline{\mathbb{Q}}_\beta^x}{|x - x_2|^{d-2}} &\leq F'(\mathbb{P}) \\ &+ \int_{\mathbb{R}^{d(N-1)}} \left( \sum_{i=2}^N V_\beta(x_i) - \beta^{-1} \log |x - x_N|^{d-1} \right) d\overline{\mathbb{Q}}_\beta^x. \end{aligned} \quad (1.75)$$

Using that  $V_\beta$  is uniformly bounded in  $\beta$  in the limit  $\beta \rightarrow \infty$ , say  $\|V_\beta\|_{L^\infty(\mathbb{R}^d)} \leq M$  for all  $\beta$  as  $\beta \rightarrow \infty$ , we further obtain

$$(1 - \beta^{-1}) \int_{\mathbb{R}^{d(N-1)}} \frac{d\overline{\mathbb{Q}}_\beta^x}{|x - x_2|^{d-2}} \leq F'(\mathbb{P}) + (N-1)M. \quad (1.76)$$

Now, it suffices to consider the trial state  $\mathbb{P} = |\Sigma|^{-(N-1)} \mathbf{1}_\Sigma^{\otimes(N-1)}$  in (1.76). One can check that the bound then obtained is bounded in  $L^\infty(K)$  uniformly in the temperature  $\beta$ , yielding the thesis of Lemma 14.  $\square$

**Lemma 15.** *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$  be such that Assumption (A) are verified, and let  $V_\beta$  be the unique maximizer of Problem  $SCE_{D,\beta}$  under Conventions 1 to 2. Then there exists  $V_\infty \in W^{1,\infty}(\mathbb{R}^d)$  such that, up to a subsequence  $\beta' \rightarrow \infty$ , we have*

$$V_\beta \xrightarrow{\beta \rightarrow \infty} V_\infty \quad \text{uniformly on every compact set.} \quad (1.77)$$

Moreover,  $v_\infty$  is a Kantorovich potential for Problem  $SCE_{D,\Sigma}$ .

*Proof of Lemma 15.* The Kantorovich potential  $V_\beta$  being bounded in  $W^{1,\infty}(\mathbb{R}^d)$  uniformly in  $\beta$  in the limit  $\beta \rightarrow \infty$ , it follows that there exists  $V_\infty \in W^{1,\infty}(\mathbb{R}^d)$  such that (up to a subsequence  $\beta' \rightarrow \infty$ )

$$V_\beta \rightarrow V_\infty \text{ weakly in } W^{1,\infty}(\mathbb{R}^d) \text{ and locally uniformly in } L^\infty(\mathbb{R}^d). \quad (1.78)$$

We claim that  $V_\infty$  is a Kantorovich potential for **Problem SCE** $_{D,\Sigma}$ . Since  $F_{\text{SCE},\beta}(\rho) \rightarrow F_{\text{SCE}}(\rho)$  as  $\beta \rightarrow \infty$  [Car+17], we have

$$F_{\text{SCE},\beta}(\rho) = -\beta^{-1} \ln N + \int_\Sigma V_\beta \rho \xrightarrow{\beta \rightarrow \infty} \int_\Sigma V_\infty \rho = F_{\text{SCE}}(\rho). \quad (1.79)$$

Now, by duality we have

$$E_{N,\Sigma}(V_\infty) + \int_\Sigma V_\infty \rho \leq F_{\text{SCE}}(\rho) = \int_\Sigma V_\infty \rho, \quad (1.80)$$

where the last equality follows from (1.79). Therefore, we will have proved that  $V_\infty$  is a Kantorovich potential for **Problem SCE** $_{D,\Sigma}$  if we can prove that  $E_{N,\Sigma}(V_\infty) \geq 0$  — which will eventually imply that  $E_{N,\Sigma}(V_\infty) = 0$ . Let us then define

$$A_\varepsilon = \left\{ (x_1, \dots, x_N) \in \Sigma^N : \sum_{i=1}^N V_\infty(x_i) > c_\alpha(x_1, \dots, x_N) + \varepsilon \right\}.$$

By convention, we have  $Z_\beta(V_\beta) = N$  for all  $\beta > 0$ . Appealing to *Fatou's lemma*, we have

$$N \geq \int_{A_\varepsilon} \liminf_{\beta \rightarrow \infty} \mathbb{P}_\beta(x_1, \dots, x_N) d\mu^{\otimes N}(x_1, \dots, x_N) \quad (1.81)$$

But, by definition of  $A_\varepsilon$ , for all  $(x_1, \dots, x_N) \in A_\varepsilon$  we have

$$\liminf_{\beta \rightarrow \infty} \mathbb{P}_\beta(x_1, \dots, x_N) = +\infty. \quad (1.82)$$

Therefore, in regards of (1.81) and (1.82), it must be that

$$\mu^{\otimes N}(A_\varepsilon) = 0.$$

Since  $A_\varepsilon$  is open in  $\Sigma^N$ , we necessarily have that  $A_\varepsilon$  is empty for all  $\varepsilon > 0$ . Therefore, we obtain  $E_{N,\Sigma}(V_\infty) \geq 0$  as wanted, yielding the thesis that  $V_\infty$  is a Kantorovich potential for **Problem SCE** $_{D,\Sigma}$ .  $\square$

*Proof of Theorem 5.* The proof of Theorem 5 now follows entirely from Lemma 14 and Lemma 15.  $\square$

We now turn to the proof of Theorem 4. Given two measures  $\mu, \nu \in \mathcal{M}(\mathbb{R}^d)$ , their *Coulomb energy* is defined as

$$D(\mu, \nu) = \iint_{\mathbb{R}^d \times \mathbb{R}^d} \frac{\mu(dx)\nu(dx')}{|x - x'|^{d-2}},$$

and we use the shorthand notation  $D(\mu) := D(\mu, \mu)$  to denote the *self-energy* of  $\mu$ . If we define the space  $\mathcal{E}(\mathbb{R}^d) \subset \mathcal{M}(\mathbb{R}^d)$  as

$$\mathcal{E}(\mathbb{R}^d) = \left\{ \mu \in \mathcal{M}(\mathbb{R}^d) : D(\mu) < \infty \right\},$$

then  $(\mu, \nu) \mapsto D(\mu, \nu)$  defines an inner product which endows  $\mathcal{E}(\mathbb{R}^d)$  with a Hilbert space structure [Lan72, Thm 1.18]. In particular, the weak topology on  $\mathcal{E}(\mathbb{R}^d)$  is defined as follows: given  $(\mu_n)_n \subset \mathcal{E}(\mathbb{R}^d)$ , we say that the sequence  $(\mu_n)_n$  weakly converge (in energy) to  $\mu$  if, for all  $\nu \in \mathcal{E}(\mathbb{R}^d)$ , we have

$$D(\mu_n, \nu) \xrightarrow{n \rightarrow \infty} D(\mu, \nu).$$

This weak topology is stronger than that of the vague topology on  $\mathcal{M}(\mathbb{R}^d)$  [Lan72, Lem 1.3]. In particular,  $\{\mu \in \mathcal{E}(\mathbb{R}^d) : D(\mu) < c\}$  is compact for the vague topology for any  $c > 0$ .

*Proof of Theorem 4.* Let  $\rho_\beta^{(e)}$  be the dual charge associated with  $V_\beta$  which is not yet “swept” onto  $\Sigma$  using Theorem 11. We claim that  $D(\rho_\beta^{(e)})$  is bounded uniformly in  $\beta$  in the limit  $\beta \rightarrow \infty$ . Integrating by parts, we have

$$D(\rho_\beta^{(e)}) = \frac{1}{c_d} \int_{\mathbb{R}^d} |\nabla V_\beta(x)|^2 dx \quad \text{where } c_d = \frac{d(d-2)\pi^{d/2}}{\Gamma(\frac{d}{2} + 1)}.$$

Now, as in the proof of Theorem 5, we have

$$|\nabla V_\beta(x)|^2 \leq (N-1)^2 \int_{\mathbb{R}^{d(N-1)}} \frac{d\mathbb{P}_\beta^x(x_2, \dots, x_N)}{|x - x_2|^{2(d-1)}}.$$

Outside a large enough compact set  $K$ , the above integral is bounded in  $L^2(\mathbb{R}^d \setminus K)$  uniformly in  $\beta$  in the limit  $\beta \rightarrow \infty$ . It then remains to control the  $L^2$ -norm of  $\nabla V_\beta$  inside  $K$ . The strategy to do so is completely analogous to that of the proof of Theorem 5.

Now, if  $\rho_\beta^{(e)}$  is swept onto  $\Sigma$ , its self-energy does not increase [Lan72, Thm. 4.4.]. Therefore, according to what precedes, the self-energy of the balayage measure remains uniformly bounded in  $\beta$  in the limit  $\beta \rightarrow \infty$ . As such, there exists  $\rho_\infty \in \mathcal{E}(\mathbb{R}^d)$  such that

$$\rho_\beta^{(e)} \xrightarrow{*} \rho_\infty \quad \text{in } \mathcal{M}(\mathbb{R}^d), \tag{1.83}$$

and we have that  $\rho_\infty$  is a dual charge for Problem  $\text{SCE}_{D,\Sigma}$ . Indeed, we know that (up to a subsequence; see Theorem 5)

$$\rho_\beta^{(e)} * |x|^{2-d} \xrightarrow{\beta \rightarrow \infty} V_\infty \quad \text{uniformly on } \Sigma, \tag{1.84}$$

where  $V_\infty$  is a Kantorovich potential for Problem  $\text{SCE}_{D,\Sigma}$ . Therefore, using (1.83), we obtain  $\rho_\infty * |x|^{2-d} = V_\infty$ , yielding that  $\rho_\infty$  is a dual charge for Problem  $\text{SCE}_{D,\Sigma}$ .  $\square$

## §A. Vanishing temperature limit of the optimal transport plan in one-dimension

We discuss the limit in the vanishing temperature limit  $\beta \rightarrow \infty$  of the multimarginal optimal transport with Coulomb cost at positive temperature in

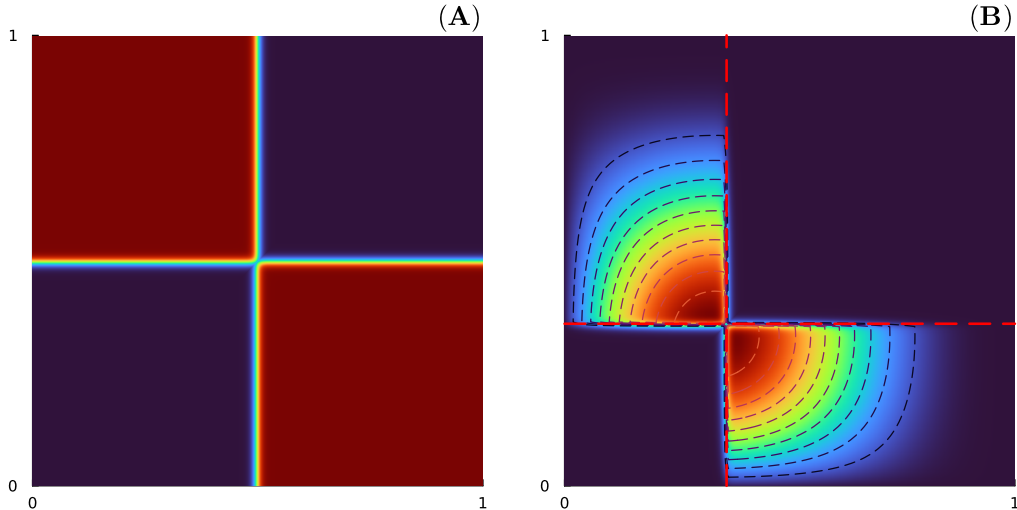


Figure 1.6: We compute the optimal transport plan at low enough temperature  $\beta \gg 1$  using *Sinkhorn's algorithm* as implemented in the Julia package `OptimalTransport.jl`. We consider two particles  $N = 2$  for the uniform density  $\rho$  over  $[-1, 1]$  (left) and for the density  $\rho \propto r(1 - r^2)^4$  on  $[0, 1]$  (right). On the right, the contour plot of the target density is added onto the heatmap. The results are in accordance with that of Theorem 16.

the one-dimensional case. It is well-known result from the theory of optimal transport with entropy that, if  $\mathbb{P}_\beta$  is the (unique) optimal transport plan for the problem with entropy — *i.e.* at positive temperature in our language — then  $\mathbb{P}_\beta$  should converge to an optimal transport plan  $\mathbb{P}$  for the original problem with no entropy — *i.e.* at zero temperature [PC19, Prop. 4.1]. Moreover, since the minimizer at zero temperature need not be unique, the limiting  $\mathbb{P}$  can be characterized as the one which minimizes the entropy — or maximizes depending on the sign convention. Here, we prove the following theorem:

**Theorem 16.** *Let  $\rho \in L^1(\mathbb{R}, \mathbb{R}_+)$  be an absolutely continuous density with  $\int_{\mathbb{R}} \rho = N \in \mathbb{N}$ . Then, the multimarginal optimal transport problem with Coulomb cost in one-dimension  $d = 1$ , that is*

$$\inf_{\rho_{\mathbb{P}} = \rho} \left\{ - \int_{\mathbb{R}^N} \sum_{1 \leq i < j \leq N} |r_i - r_j| d\mathbb{P}(r_1, \dots, r_N) \right\}. \quad (\text{SCE}_{d=1})$$

has an infinite number of optimizers. In fact, if we let the sequence  $-\infty = \ell_0 < \ell_1 < \dots < \ell_{N-1} < \ell_N = \infty$  be such that  $\int_{\ell_i}^{\ell_{i+1}} \rho = 1$  for all  $i = 0, \dots, N - 1$ , then any admissible transport plan  $\mathbb{P}$  such that

$$\text{supp}(\mathbb{P}) \subset \bigcup_{\substack{i_1, \dots, i_N = 0, \dots, N-1 \\ i_k \neq i_l \text{ for } k \neq l}} [\ell_{i_1}, \ell_{i_1+1}] \times \dots \times [\ell_{i_N}, \ell_{i_N+1}] \quad (1.85)$$

is optimal for Problem  $\text{SCE}_{d=1}$ . In particular, if we denote  $\rho_i$  the restriction of  $\rho$  on the segment  $[\ell_{i-1}, \ell_i]$ , the optimal transport plan which minimizes the



relative entropy with respect to  $(\rho/N)^{\otimes N}$  is given by

$$\mathbb{P}_N(r_1, \dots, r_N) = \frac{1}{N!} \sum_{\substack{i_1, \dots, i_N = 0, \dots, N-1 \\ i_k \neq i_l \text{ for } k \neq l}} \rho_{i_1}(r_1) \otimes \dots \otimes \rho_{i_N}(r_N). \quad (1.86)$$

*Proof of Theorem 16.* Let  $\mathbb{P}$  be an admissible transport plan for Problem SCE<sub>d=1</sub>, and suppose that the support  $\Gamma$  of  $\mathbb{P}$  is contained in the aforementioned set, that is

$$\Gamma \subset \Sigma := \bigcup_{\substack{i_1, \dots, i_N = 0, \dots, N-1 \\ i_k \neq i_l \text{ for } k \neq l}} [\ell_{i_1}, \ell_{i_1+1}] \times \dots \times [\ell_{i_N}, \ell_{i_N+1}]. \quad (1.87)$$

Without loss of generality, given  $(r_1, \dots, r_N) \in \Gamma$ , we may always rearrange the particles such that  $r_1 \leq \dots \leq r_N$ . Therefore, we can suppose that  $r_i \in [\ell_{i-1}, \ell_i]$  for all  $i = 1, \dots, N$ . We let  $V$  be the Kantorovich potential defined in Theorem 6, that this

$$V(r) = - \sum_{i=1}^{N-1} \delta_{\ell_i} * |r|. \quad (1.88)$$

It follows from the expression of  $V$  that for all  $r_i \in [\ell_{i-1}, \ell_i]$ , we have

$$V(r_i) = (N - 2i + 1)r_i + \sum_{j=1}^{i-1} \ell_j - \sum_{j=i}^{N-1} \ell_j. \quad (1.89)$$

Therefore, summing over the  $i$ 's we obtain

$$\sum_{i=1}^N V(r_i) = \sum_{i=1}^N (N - 2i + 1)r_i + \sum_{j=1}^N (N - 2j)\ell_j. \quad (1.90)$$

On the other hand, once the particles are rearranged, we can write

$$- \sum_{1 \leq i < j \leq N} |r_i - r_j| = \sum_{i=1}^N (N - 2i + 1)r_i. \quad (1.91)$$

Therefore, whenever each particles is consigned to its segment, and irrespective of its position inside it, we have

$$V(r_1) + \dots + V(r_N) = - \sum_{1 \leq i < j \leq N} |r_i - r_j| + C \quad (1.92)$$

where  $C = \sum_{j=1}^N (N - 2j)\ell_j$  is a constant. Furthermore, it can be checked by the triangular inequality that the equality above becomes an inequality over all configurations in  $\mathbb{R}^N$ . Therefore, it follows from the classical results of optimal transport that  $\mathbb{P}$  is optimal for Problem SCE<sub>d=1</sub>. Among all the transport plan which verifies (1.87), the one which minimizes the relative entropy with respect to the product measure  $(\rho/N)^{\otimes N}$  is given by

$$\mathbb{P}_N(r_1, \dots, r_N) = \frac{1}{N!} \sum_{\substack{i_1, \dots, i_N = 0, \dots, N-1 \\ i_k \neq i_l \text{ for } k \neq l}} \rho_{i_1}(r_1) \otimes \dots \otimes \rho_{i_N}(r_N). \quad (1.93)$$

Indeed, given  $\mathbb{P}$  supported on  $\Gamma$  such that  $\Gamma \subset \Sigma$ , then

$$\text{Ent}(\mathbb{P}|\mu) = \frac{1}{N!} \sum_{\substack{i_1, \dots, i_N = 0, \dots, N-1 \\ i_k \neq i_l \text{ for } k \neq l}} \text{Ent}(\mathbb{P}_{i_1, \dots, i_N}|\mu), \quad (1.94)$$

where  $\mu = (\rho/N)^{\otimes N}$  and where  $\mathbb{P}_{i_1, \dots, i_N}$  denotes the restriction of  $\mathbb{P}$  on the hypercube  $I_{i_1, \dots, i_N} := [\ell_{i_1}, \ell_{i_1+1}] \times \dots \times [\ell_{i_N}, \ell_{i_N+1}]$ . It follows from Jensen's inequality that

$$\text{Ent}(\mathbb{P}_{i_1, \dots, i_N}|\mu) \geq \mathbb{P}(I_{i_1, \dots, i_N}) \log \mathbb{P}(I_{i_1, \dots, i_N}) \quad (1.95)$$

The equality is obtained when  $\mathbb{P}_{i_1, \dots, i_N} = \rho_{i_1} \otimes \dots \otimes \rho_{i_N}$ . Furthermore, by the case of equality in the Jensen's inequality, it can be checked that this choice is the only one which achieves the equality above. For the transport plan  $\mathbb{P}_N$ , we therefore have

$$\text{Ent}(\mathbb{P}_N|\mu) = N \log N. \quad (1.96)$$

Nevertheless, it might *a priori* be that there exists an optimal transport plan  $\mathbb{P}$  for **Problem SCE<sub>d=1</sub>** whose support  $\Gamma$  is not supported on the set  $\Sigma$  as defined in (1.87). We claim that if such  $\mathbb{P}$  exists, it must have infinite entropy. Indeed, let us show that if  $(r_1, \dots, r_N)$  is a configuration in  $\Gamma$  such that particles do not overlap, *i.e.*  $r_i \neq r_j$ , and such that  $r_i \neq \ell_k$  for any  $k = 1, \dots, N-1$ , then necessarily — up to reordering of the particles such that  $r_1 < \dots < r_N$  — each particle  $r_i$  is consigned in its segment, *i.e.*  $r_i \in [\ell_{i-1}, \ell_i]$  for all  $i = 1, \dots, N$ . Indeed, differentiating the optimality condition with respect to  $r_i$  — which is allowed from the fact that particles do not overlap and that  $V$  is differentiable away from the  $\ell_k$ 's — we obtain that

$$V'(r_i) = N - 2i + 1 \quad (1.97)$$

Necessarily, according to (1.89), we have  $r_i \in (\ell_{i-1}, \ell_i)$ . Therefore, if the support of  $\Gamma$  is not contained in  $\Sigma$ , then necessarily we have  $\Gamma \cap \Sigma^c \subset \mathcal{A}$  where the set  $\mathcal{A}$  is

$$\begin{aligned} \mathcal{A} := & \{(r_1, \dots, r_N) \in \mathbb{R}^N : \exists i \neq j \text{ s.t. } r_i = r_j\} \\ & \cup \{(r_1, \dots, r_N) \in \mathbb{R}^N : \exists k = 1, \dots, N-1 \text{ s.t. } r_i = \ell_k \text{ for some } i\}. \end{aligned} \quad (1.98)$$

The set  $\mathcal{A}$  has zero Lebesgue measure. Therefore, if  $\mathbb{P}(\mathcal{A}) > 0$ , then  $\mathbb{P}$  must have singular part and therefore its relative entropy with respect to  $(\rho/N)^{\otimes N}$  is infinite.  $\square$

## §B. Convergence of the entropy-regularized optimal transport to the unregularized problem

In this short appendix, we comment of the convergence of the entropy-regularized optimal transport problem towards the original unregularized problem in the

vanishing limit of the regularization parameter, hereafter denoted  $\varepsilon$  — and corresponding to the temperature  $1/\beta$  in this chapter. We set ourselves in the generic setting of a  $N$ –marginal optimal transport problem with cost of transportation  $c(x_1, \dots, x_N)$  and marginal constraints given by  $\mu_1, \dots, \mu_N$ , where for simplicity we assume that the  $\mu_i$ 's are absolutely continuous probability measures on  $\mathbb{R}^d$ . We let

$$F_\varepsilon(\mathbb{P}) := \int_{\mathbb{R}^{dN}} c(x_1, \dots, x_N) d\mathbb{P}(x_1, \dots, x_N) + \varepsilon \text{Ent}(\mathbb{P}). \quad (1.99)$$

Here, we denote  $\text{Ent}(\mathbb{P})$  the entropy of a probability measure  $\mathbb{P}$  with respect to the Lebesgue measure. We then consider the multimarginal optimal transport problem

$$C_0 \equiv C_0(\mu_1, \dots, \mu_N) := \inf_{\mathbb{P} \in \Pi(\mu_1, \dots, \mu_N)} F_0(\mathbb{P}) \quad (\text{MOT})$$

and its entropy-regularized version

$$C_\varepsilon \equiv C_\varepsilon(\mu_1, \dots, \mu_N) := \inf_{\mathbb{P} \in \Pi(\mu_1, \dots, \mu_N)} F_\varepsilon(\mathbb{P}). \quad (\text{MOT}_\varepsilon)$$

An important question is the convergence of **Problem MOT $_\varepsilon$**  to **Problem MOT** in the limit  $\varepsilon \rightarrow 0$ . In [Car+17], Carlier *et al.* proved in the case of the classical two-marginal optimal transport with quadratic cost — *i.e.*  $N = 2$  and  $c(x, y) = |x - y|^2$  — that the (unique) optimal transport plan  $\mathbb{P}_\varepsilon$  of the regularized problem converges to the optimal transport  $\mathbb{P}$  to the unregularized problem in the limit  $\varepsilon \rightarrow 0$  — which is also unique in this case, at least under good assumptions on the marginals  $\mu_1$  and  $\mu_2$ . The main difficulty is that the optimal transport plan  $\mathbb{P}$  will generically be supported onto a low-dimensional manifold and will therefore have infinite entropy. To cope with this problem, the idea of [Car+17] is to slightly regularize  $\mathbb{P}$  using the so-called *block approximation*  $\mathbb{P}^{(\ell)}$  so that its entropy is finite and its cost of transportation remains very close to the minimum. We refer to [Car+17] for a concise definition this approximation. Here  $0 < \ell \ll 1$  is some scale parameter, where morally the approximation gets finer as  $\ell \rightarrow 0$ . The key properties proved are that for all  $\ell > 0$  the block approximation  $\mathbb{P}^{(\ell)}$  remains an admissible transport plan, that is  $\mathbb{P}^{(\ell)} \in \Pi(\mu, \nu)$  [Car+17, Prop. 2.10], that  $\mathbb{P}^{(\ell)}$  converges to  $\mathbb{P}$  for the Wasserstein distance  $W_2$  [Car+17, Cor. 2.12], and finally that the entropy of  $\mathbb{P}^{(\ell)}$  is controlled from above by  $-\log(\ell)$  as  $\ell \rightarrow 0$  [Car+17, Prop. 2.14].

The rationale of [Car+17] can actually be extended at no costs to the multimarginal setting for rather general costs of transport  $c(x_1, \dots, x_N)$ . Indeed, the block approximation can be trivially extended to a multimarginal setting. The control obtained in [Car+17, Prop. 2.14] on the entropy of the block approximation  $\mathbb{P}^{(\ell)}$  is a general statement that has nothing to do either with the multimarginal framework nor the cost of transportation  $c$ . Now, the remaining ingredient is the convergence of the cost of the regularized optimal transport  $\mathbb{P}_\varepsilon$  — that is, without the entropy term, *i.e.*  $F_0(\mathbb{P}_\varepsilon)$  — to the original optimal transport cost, that is  $C_0$ . In fact, Carlier *et al.* prove the much stronger — in their setting — statement that the block approximation  $\mathbb{P}^{(\ell)}$  converges in the Wasserstein distance  $W_2$  — which entails that  $F_0(\mathbb{P}_\varepsilon) \rightarrow F_0(\mathbb{P})$  by classical

functional analysis arguments, see [Vil09, Def. 6.8, Thm. 6.9]. In fact, a close look at their proof shows that this remains true for any Wasserstein distance  $W_p$  with  $p \geq 1$ , so that their rationale extends easily to cost of transportations  $c(x_1, \dots, x_N)$  which are continuous and do not grow faster than polynomially at infinity.

Indeed, let us prove the following generalisation of [Car+17] to the multi-marginal case:

**Theorem 17** (Convergence of the entropy-regularized problem in the limit  $\varepsilon \rightarrow 0$  [Car+17]). *Let  $c : (\mathbb{R}^d)^N \rightarrow \mathbb{R}$  be a continuous cost of transportation such that there exists  $p \geq 1$  so that the cost  $c$  does not grow faster than  $x \mapsto |x|^p$ . Suppose that all the marginals  $\mu_i$  ( $i = 1, \dots, N$ ) have both finite entropy and  $p$ -moment, that is*

$$\int_{\mathbb{R}^d} |x|^2 \mu_i(x) dx < \infty \quad \text{and} \quad \int_{\mathbb{R}^d} \mu_i(x) \log \mu_i(x) dx < \infty. \quad (1.100)$$

Then, it holds that

$$C_\varepsilon(\mu_1, \dots, \mu_N) \rightarrow C_0(\mu_1, \dots, \mu_N) \quad \text{as } \varepsilon \rightarrow 0. \quad (1.101)$$

Futhermore, letting  $\mathbb{P}_\varepsilon$  be the unique optimal transport plan of the entropic-regularized optimal transport problem, then any accumulation point of the sequence  $(\mathbb{P}_\varepsilon)_\varepsilon$  as  $\varepsilon \rightarrow 0$  is an optimal transport plan for the original unregularized optimal transport problem.

*Proof of Theorem 17.* Let us first prove the convergence of the costs (1.101). By positivity of the entropy, we have  $C_\varepsilon \geq C_0$  for all  $\varepsilon \geq 0$ . Now, let us consider a (not necessarily optimal) transport plan  $\mathbb{P}_{\text{fin}} \in \Pi(\mu_1, \dots, \mu_N)$  that has finite entropy. Such a plan always exists as soon as the marginals  $\mu_i$ 's have finite entropy as assumed in (1.100), by considering the tensor product  $\mathbb{P} = \mu_1 \otimes \dots \otimes \mu_N$  as a trial state for Problem MOT. Then, we have by definition

$$C_\varepsilon \leq F_\varepsilon(\mathbb{P}_{\text{fin}}). \quad (1.102)$$

By the fact that  $\mathbb{P}_{\text{fin}}$  has finite entropy, we obtain

$$\limsup_{\varepsilon \rightarrow 0} C_\varepsilon \leq \limsup_{\varepsilon \rightarrow 0} F_\varepsilon(\mathbb{P}_{\text{fin}}) = F_0(\mathbb{P}_{\text{fin}}). \quad (1.103)$$

From this, we obtain

$$\limsup_{\varepsilon \rightarrow 0} C_\varepsilon \leq \inf_{\substack{\mathbb{P} \in \Pi(\mu_1, \dots, \mu_N) \\ \text{Ent}(\mathbb{P}) < \infty}} F_0(\mathbb{P}) := C'(\mu_1, \dots, \mu_N) \equiv C'. \quad (1.104)$$

Now, it remains to prove that  $C' = C_0$ . Let  $\mathbb{P}^*$  be an optimal transport plan for the unregularized problem, that is  $C_0 = F_0(\mathbb{P}^*)$ . As stated earlier, it need not be that  $\mathbb{P}^*$  has finite entropy. If it were — although it will generically never be the case — then we would have immediately that  $C' = C_0$ , thus proving (1.101). The idea is then to regularize  $\mathbb{P}^*$  so that it has finite entropy and a transport cost very close to the original one, while still ensuring the marginal constraints.

This is where comes into play the block approximation  $\mathbb{P}^{(\ell)}$  mentioned above — see [Car+17] for a detailed definition. Then, we have  $C' \leq F_0(\mathbb{P}^{(\ell)})$  for all  $\ell > 0$ , and letting  $\ell \rightarrow \infty$  yields that  $C' \leq C_0$  according to [Car+17, Cor. 2.12], which eventually yields the equality  $C' = C_0$ , whence (1.101).

Finally, from the convergence of the costs, we can prove that any accumulation point of the sequence  $(\mathbb{P}_\varepsilon)_\varepsilon$  is an optimal transport plan for the unregularized problem. Let us suppose that (up to a subsequence which we do not relabel)  $\mathbb{P}_\varepsilon \rightarrow \mathbb{P}$ , and let us prove that  $\mathbb{P}$  is an optimal transport plan for Problem MOT. By lower semicontinuity of the functional  $F_0$ , we have

$$F_0(\mathbb{P}) \leq \liminf_{\varepsilon \rightarrow 0} F_0(\mathbb{P}_\varepsilon) \leq \liminf_{\varepsilon \rightarrow 0} F_\varepsilon(\mathbb{P}_\varepsilon) = \liminf_{\varepsilon \rightarrow 0} C_\varepsilon = C. \quad (1.105)$$

Therefore  $\mathbb{P}$  is an optimal transport plan for the unregularized optimal transport problem. This terminates the proof of Theorem 17.  $\square$

*Remark 9.* In the above proof, the convergence of the costs  $F_0(\mathbb{P}^{(\ell)})$  to  $F_0(\mathbb{P}) = C_0$  was given by the convergence of the block approximation  $\mathbb{P}^{(\ell)}$  to  $\mathbb{P}$  in the Wasserstein sense  $W_p$  for some  $p \geq 1$ . *A priori*, when the cost of transportation  $c(x_1, \dots, x_N)$  no longer verifies the assumptions of Theorem 17, the convergence needs to be proved at the level of the cost — that is, not by appealing to the convergence of  $\mathbb{P}^{(\ell)}$  to  $\mathbb{P}$ . That is, one needs to prove  $F_0(\mathbb{P}^{(\ell)}) \rightarrow F_0(\mathbb{P})$  directly on the quantities. In the case of the Coulomb cost (at least in dimension  $d = 2$ ) this was done by more direct computations by Gerolin *et al.* in [GKR20].

Now, we have the following theorem:

**Theorem 18.** *Let us suppose that any accumulation point of  $(\mathbb{P}_\varepsilon)_\varepsilon$ , where  $\mathbb{P}_\varepsilon$  is the unique optimal transport for Problem  $\text{MOT}_\varepsilon$ , is an optimal transport plan for Problem MOT. Suppose that there exists an optimal transport plan for the unregularized optimal transport problem that has finite entropy. Then, the sequence  $(\mathbb{P}_\varepsilon)_\varepsilon$  converges to the optimal transport plan for the unregularized problem which has the largest possible entropy.*

*Proof of Theorem 18.* The proof goes as in the discrete setting, see [PC19]. Let us suppose there exists  $\mathbb{P}_{\text{fin}}$  an optimal transport plan for Problem MOT that has finite entropy, that is  $C_0 = F_0(\mathbb{P}_{\text{fin}})$  and  $\text{Ent}(\mathbb{P}_{\text{fin}}) < \infty$ . Let  $\mathbb{P}_\varepsilon$  be as usual the (unique) optimal transport plan to the regularized Problem  $\text{MOT}_\varepsilon$ . Then, by definition we have  $C_0 = F_0(\mathbb{P}_{\text{fin}}) \leq F_0(\mathbb{P}_\varepsilon)$  and  $F_\varepsilon(\mathbb{P}_\varepsilon) \leq F_\varepsilon(\mathbb{P}_{\text{fin}})$ . Comparing these two inequalities yield that

$$\text{Ent}(\mathbb{P}_\varepsilon) \leq \text{Ent}(\mathbb{P}_{\text{fin}}). \quad (1.106)$$

Therefore, if  $\mathbb{P}_\varepsilon$  converges (up to a subsequence  $\varepsilon \rightarrow 0$ ) to an optimal transport plan  $\mathbb{P}$  for Problem MOT, then it follows from the preceding inequality that  $\mathbb{P}$  must maximize the entropy. Because the set of solutions to Problem MOT is convex and the entropy is a strictly convex functional, we also have that  $\mathbb{P}$  is unique. This also entails that the entire sequence  $\mathbb{P}_\varepsilon$  converges to this specific  $\mathbb{P}$  — a stronger statement to that of Theorem 17.  $\square$

*Remark 10* (Unicity of the limit  $(\mathbb{P}_\varepsilon)_\varepsilon$ ). In the case where the minimizers to Problem MOT are not unique, then *a priori* the sequence of regularized optimal transport plan  $\mathbb{P}_\varepsilon$  can have several accumulation points, all of them being optimal for the unregularized problem. From what has been said previously at the end of the proof of Theorem 18, we know nonetheless that if there exists an optimal transport plan  $\mathbb{P}_{\text{fin}}$  for Problem MOT that has finite entropy, then necessarily the sequence  $(\mathbb{P}_\varepsilon)_\varepsilon$  actually converges as a whole to the unique optimal transport plan  $\mathbb{P}$  for Problem MOT which has maximum entropy. This is very clear from Theorem 16 in the case of the one-dimensional MOT problem with Coulomb.

## §C. The Uniform Electron Gas at the boundary

When introducing the dual charge and its discretization, we had in mind one particular numerical application among others, which has to do with the *Uniform Electron Gas*. In this appendix, we introduce and present some very preliminary results on this problem, which we plan to study more in depth in the near future.

### §C.1. The Uniform Electron Gas

The *Uniform Electron Gas* (UEG) [LLS18; LLS19b] is an important model which is notably used in the *Local Density Approximation* [LLS20], a very important class of approximations for the exchange-correlation functionals in DFT. It is also used to obtain bounds on the best constant of the *Lieb–Oxford inequality* [Lie79; LO81; Sei+22; LLS22], which is a universal bound on the electrostatic interaction energy of quantum electron systems. We refer to Appendix A in Chapter 2 for more details on this inequality. The UEG is defined as follows. We consider  $N$  classical particles interacting with each others through the Riesz potential  $w_s$ , that is

$$w_s(x) = \begin{cases} |x|^{-s} & \text{for } s > 0, \\ -\ln|x| & \text{for } s = 0, \\ -|x|^{-s} & \text{for } s < 0. \end{cases} \quad (1.107)$$

Given a domain  $\Omega \subset \mathbb{R}^d$  such that  $|\Omega| = N$ , we constrain the electrons to be inside  $\Omega$  and distributed in such a way that their density is constant on  $\Omega$ , that is  $\rho_{\mathbb{P}} = \mathbf{1}_\Omega$  where  $\mathbb{P}$  is the distribution of the electrons. Among those distributions, we seek for the one(s) yielding the smallest *indirect energy* possible, that is we

want to solve the following minimization problem

$$E_{\text{UEG}}(\Omega, s) := \inf_{\rho_{\mathbb{P}} = \mathbb{1}_{\Omega}} \left\{ \int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} w_s(r_i - r_j) d\mathbb{P}(r_1, \dots, r_N) \right\} - \frac{1}{2} \iint_{\Omega \times \Omega} w_s(r - r') dr dr'. \quad (1.108)$$

The first term is nothing but the multimarginal optimal transport problem with Riesz cost where the target marginal is given by the uniform density on the domain  $\Omega$ , that is  $\mathbb{1}_{\Omega}/|\Omega|$ . The second term represents the interaction of this density with itself, and is sometimes called the *Hartree energy* in the context of quantum physics. Once subtracted, we obtain the so-called *indirect energy*. The UEG energy per unit volume is then defined in [LLS18, Thm. 2.6] as the thermodynamic limit

$$e_{\text{UEG}}(d, s) := \lim_{\substack{N \rightarrow \infty \\ \Omega \nearrow \mathbb{R}^d \text{ and } |\Omega|=N}} \frac{E_{\text{UEG}}(\Omega, s)}{N}. \quad (1.109)$$

It may appear to some readers that the name *Uniform Electron Gas* actually refers to another model named *Jellium*. Nevertheless, we will adopt here the convention of [LLS18; LLS20; LL15; LLS19a] where the UEG is *a priori* a distinct model different to Jellium, and which is defined as in (1.108) and (1.109). In fact, in this appendix, we are interested in investigating a question which is related to the work Lewin, Lieb and Seiringer [LLS19a] where it is proved that the UEG and the Jellium are actually equivalent models at the level of their energies. Before explaining this, let us recall the definition of Jellium.

## §C.2. The Jellium

*Jellium* — also called *one-component plasma* (OCP) — is a very important model in condensed matter physics. Although an accurate historical account of this model is delicate, it seems that the classical version of this model was first proposed by Thomson [Tho04] at the very beginning of the last century, while its quantum formulation was introduced by Wigner [Wig34] in 1934. Jellium is a simplified model of delocalised electrons in metals and semiconductors. Although rather rudimentary, it can qualitatively reproduce the characteristics of some real materials, such as plasmons and crystallisation. The idea behind Jellium is to replace the atomic lattice on which the electrons are located with a positively charged *uniform background*, similar to a "jelly" — hence the name of the model, which was given by Herring [Her52]. Despite its apparent simplicity, many conjectures on the Jellium remain largely open at the present time. If we denote, as previously,  $\Omega$  a domain of  $\mathbb{R}^d$  with  $|\Omega| = N$ , the Jellium energy is



defined as

$$E_{\text{Jel}}(\Omega, s) := \inf_{x_1, \dots, x_N \in \mathbb{R}^d} \left\{ \sum_{i=1 \leq i < j \leq N} w_s(x_i - x_j) - \sum_{i=1}^N \int_{\Omega} w_s(x_i - x') dx' + \frac{1}{2} \iint_{\Omega \times \Omega} w_s(x - x') dx dx' \right\} \quad (1.110)$$

The first term corresponds to the interaction energy of the electrons with each other, while the second corresponds to the interaction of each electron with the uniform background. The third and last term corresponds to the interaction of the background with itself. This model was already introduced in the Introduction of this thesis, see (98). We also refer to Chapter 3, where we study this model in one space-dimension. The Jellium energy per unit volume is then defined in the thermodynamic limit as

$$e_{\text{Jel}}(s, d) := \lim_{\substack{N \rightarrow \infty \\ \Omega \nearrow \mathbb{R}^d \text{ and } |\Omega|=N}} \frac{E_{\text{Jel}}(\Omega, s)}{N} \quad (1.111)$$

The existence of this thermodynamic limit was proved by Lieb and Narhnofer [LN75] in the Coulomb case  $s = 1$  in dimension  $d = 3$  in a landmark paper, and generalised to the Coulomb case  $s = d - 2$  in any dimension by Sari and Merlini [SM76]. For general exponent  $s \geq 0$ , we refer to the long line of works initiated by Serfaty *et al.* see [Ser15] or the references in [Lew22a]. The case of the Coulomb gas in dimension one, for which  $s = -1$ , was settled by Kunz [Kun74].

An important conjecture on Jellium concerns the nature of the minimiser of this energy. It is expected — although a proof is still lacking — that the electrons are crystallised onto a periodic lattice at optimality [BL15, Sec. 2.6]. This conjecture was first formulated by Wigner [Wig34] in the Coulomb case  $s = d - 2$  for dimensions  $1 \leq d \leq 3$ . In dimension  $d = 3$ , this lattice should be a *body-centred cubic* (BCC) lattice, and a hexagonal lattice in dimension  $d = 2$ . The conjecture extends for all  $s \geq d - 2$ . We note that in dimension  $d = 1$ , it is known that Jellium is crystallised on  $\mathbb{Z}$  in the Coulomb case  $s = -1$  [Kun74; AM80] — as well as in the quantum case [BL02]. There is no rigorous proof in the general Riesz case, and the problem is still largely open. These conjectures are one of many examples of *crystallisation conjectures* in mathematical physics. These conjectures are very complicated problems that usually require sophisticated techniques. We refer to [BL15] for an overview of the subject.

### §C.3. Equivalence of Jellium and Uniform Electron Gas

Let us now come to the question of the equivalence of the UEG and the Jellium, that is the equality of the two energies (1.111) and (1.109). It had been assumed for a long time that the two models should be equivalent, although a rigorous proof was only made available very recently, first by Cotar and Petrache [CP19]

and shortly afterwards by Lewin, Lieb and Seiringer [LLS19a]. The reason for this is as follows. Jellium and UEG are *dual* to each other. It is believed that the external charge associated to the UEG — that is, the charge which generates the potential which forces the particles into the UEG ground-state, and which exactly corresponds to the dual charge of the associated multimarginal optimal transport as introduced in this chapter — should be constant in space, and therefore that UEG is equivalent to Jellium. As mentioned above, Jellium is believed to be crystallised onto a BCC crystal, so that its density should not be constant in space. Nevertheless, because the crystal has no preferred orientation nor position, one may freely average it in space in such a way as to obtain a state whose density is effectively constant. Such a state is called a *floating Wigner crystal* [BL82; Dru+04; MZ02], and it was believed for a long time that this trial state should yield the equality of both models. It turns out that this is wrong [LL15; BBS89; BBS14; Bor+13]. We will now explain the strategy of [LLS19a] to modify this trial state in such a way as to obtain the equivalence, without dwelling on the mathematical and computational details. We note that the equivalence is *not* true in dimension  $d = 1$  [LLS19a].

First, let us note that  $e_{\text{UEG}}(s, d) \geq e_{\text{Jel}}(s, d)$  for all  $s$  and all dimension  $d \geq 1$ , as can be shown by a direct bound. It remains to find a well-chosen trial state  $\mathbb{P}$  for the UEG to obtain the converse inequality. Assuming that Jellium is crystallised onto a lattice  $\mathcal{L}$ , a perfectly legitimate idea would be to start from such a minimiser in finite volume  $|\Omega| = N < \infty$ , that is where the particles  $x_1, \dots, x_N$  are located on the vertices of the lattice  $\mathcal{L} \cap C$ , where  $C$  is a box of volume  $|C| = N$ , and to average the configuration over the *Wigner-Seitz cell*  $Q$  of the lattice. We obtain a finite piece of the so-called *floating Wigner crystal* as mentioned above, and it can be checked by a direct calculation that the resulting state  $\mathbb{P}$  satisfies the constraint  $\rho_{\mathbb{P}} = \rho_{\Omega}$ . It is then tempting to use  $\mathbb{P}$  as a trial state for our variational problem. A calculation then shows that this trial state is *bad* [LL15; BBS89; BBS14; Bor+13]. In dimension  $d = 3$  for  $s = 1$ , we find that in the limit  $N \rightarrow \infty$  the energy of the floating crystal is given by

$$\int_{\mathbb{R}^{dN}} \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|} - \frac{1}{2} \iint_{\mathbb{R}^3 \times \mathbb{R}^3} \frac{\rho_{\Omega}(x)\rho_{\Omega}(x')}{|x - x'|} \xrightarrow{N \rightarrow \infty} \zeta_{\text{BCC}}(1) + \frac{2\pi}{3} \int_Q |x|^2 dx. \quad (1.112)$$

Here the quantity  $\zeta_{\text{BCC}}(1)$  corresponds to the energy of the BCC lattice and is expressed from the *Epstein function* which is the generalisation of the Riemann's *zeta function*  $\zeta$  to regular lattices [BL15, Sec. 2.5]. Therefore, under the conjecture that Jellium is crystallised onto a BCC lattice, we have  $e_{\text{Jel}}(1, 3) = \zeta_{\text{BCC}}(1)$ . Consequently,  $\mathbb{P}$  does not provide the equality of the two models. Indeed, we see the appearance of a positive shift in the right-hand side of (1.112) which corresponds to the trace of the quadrupole on  $Q$ . The physical explanation is as follows [LL15]. When we make the crystal float, we create a gradient of charge which is responsible for a dipole whose energy remains at the thermodynamic limit. For example, when the crystal is close to the top

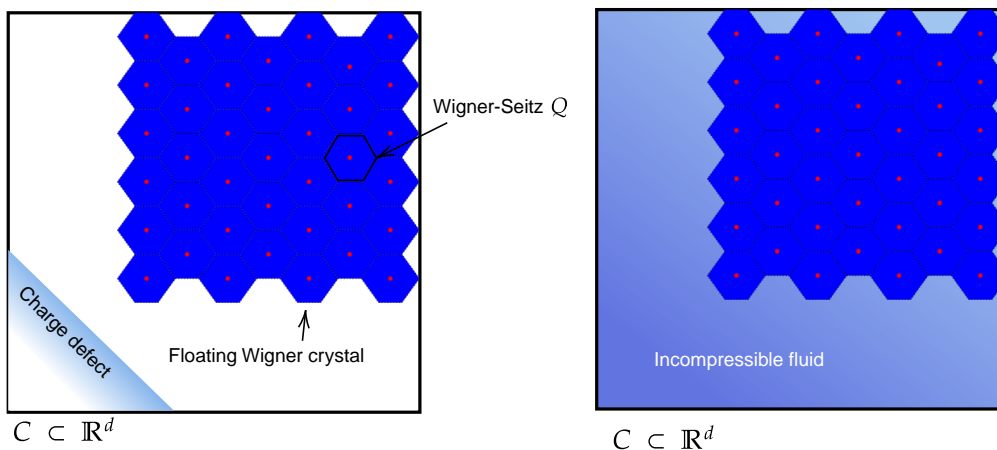


Figure 1.7: When the floating crystal is at the top left corner of the  $C$ , a significant defect of charge is created in the opposite corner. This gradient of charge is responsible for the quadrupole term in (1.112). The idea is [LLS19a] is to immerse the crystal in an incompressible fluid. When the crystal moves, the fluid ensures that local neutrality is maintained and prevents the creation of a this gradient of charge.

left-hand corner of  $C$ , a charge defect is created in the opposite corner. This gradient of charge is responsible for the said dipole in (1.112).

The idea of [LLS19a], to avoid the formation of this dipole, is to immerse the floating crystal in a perfect charged fluid, whose *raison d'être* is to ensure the conservation of local neutrality. Indeed, in the presence of this fluid, when the crystal moves as before into a corner of  $C$ , the fluid that previously occupied this space is displaced into the opposite corner, preventing the energy shift seen in (1.112). With this new test state, the equivalence of the two models is obtained.

#### §C.4. The Uniform Electron Gas near its boundary

The equivalence of the two models at the level of the energy is thus obtained. This equivalence was obtained by constructing a judicious trial state which provides equality of the two models in the thermodynamic limit  $N$ . An interesting question now arises. *In finite volume, that is at a fixed  $N < \infty$ , is the ground state of the Uniform Electron Gas also a fluid near its boundary?* We plan on answering this question in the near future using the dual charge. Here, we only discuss the general strategy and we give a very preliminary numerical result.

To answer this question, we seek to compute the *two-point correlation function*  $\rho^{(2)}(x, y)$  — or *pair correlation* — of the UEG in the finite volume  $N < \infty$ . That is, if  $\mathbb{P}_N$  is the measure which achieves the *infimum* in (1.110), we look at the quantity

$$\rho^{(2)}(x, y) = N(N-1) \int_{\mathbb{R}^{d(N-2)}} \mathbb{P}_N(x, y, dx_3, \dots, dx_N). \quad (1.113)$$

In the case of a perfect crystal, the function  $\rho^{(2)}$  is a periodic function with sharp peaks at the crystal sites. On the other hand, in the case of a perfect fluid, the function  $\rho^{(2)}$  should be constant. Therefore, we wonder whether or not we can see a qualitative change of behaviour in the function  $y \mapsto \rho^{(2)}(x_0, y)$  according to this phenomenology, where the particle  $x_0$  is fixed well inside  $\Omega$  and when it is located near its boundary. As a very preliminary test, we ran our algorithm as defined in this chapter for the UEG in dimension  $d = 2$ . More precisely, using our algorithm we approximate the Kantorovich potential  $V$  for the multimarginal optimal transport at positive temperature  $\beta = 50$  with  $N = 15$ . Here, we set the target density to be the uniform measure on the unit disk  $D$  of  $\mathbb{R}^2$ . We then compute the pair correlation of the associated Gibbs measure  $\mathbb{P}_\beta$ .

In Figure 1.8, we observe rather clearly that the system seems to be crystallised. Nevertheless, as we get  $x_0$  closer to the boundary, it is unclear that a change of behaviour happens. We do observe that the region near  $x_0$  is getting darker and closer to the boundary, but this is not clear. It is in fact of no surprise that we need to consider bigger  $N$ 's, but this is very challenging numerically. We will go back to this question in the near future.

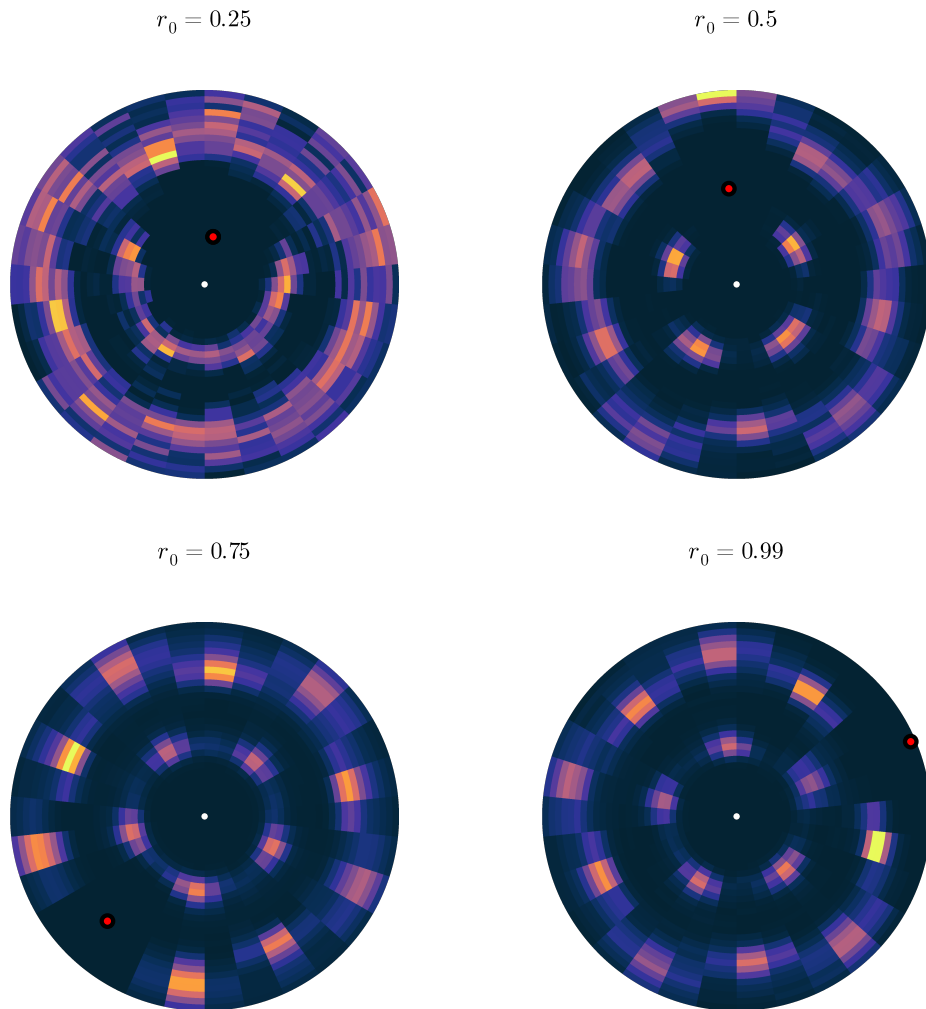


Figure 1.8: We compute the pair correlation of the UEG in two dimension where  $\Omega$  is taken to be the unit disk. More precisely, we run our algorithm to obtain an approximate Kantorovich potential for  $\beta = 50$ , and we compute the pair correlation  $\rho^{(2)}(x, y)$  of the associated Gibbs measure. We then fix a reference particle  $x_0$  (red point) and display the heatmap of  $y \mapsto \rho^{(2)}(x_0, y)$ . Here,  $r_0 = |x_0|$ . We clearly observe what seems to be a crystal. Nevertheless, the number of particles is too small to infer any specific change of behaviour near the boundary.

## CHAPTER 2

# ASYMPTOTIC OF THE KANTOROVICH POTENTIAL FOR THE MULTIMARGINAL OPTIMAL TRANSPORT WITH REPULSIVE COSTS

*This chapter is an extended version of our paper Asymptotic of the Kantorovich potential for the optimal transport with Coulomb cost [Lel22b], that has been submitted to a peer-reviewed journal. We prove a conjecture regarding the asymptotic of the Kantorovich potential for the optimal transport with repulsive costs. To the best of our knowledge, this conjecture was only written down explicitly for the first time in the recent survey of Friesecke et al. in [FGGG22], although it had been an open problem for a longer time. The proof relies on the concept of  $c$ -cyclical monotonicity in optimal transportation.*

*Among its several application, we may use this asymptotic to gain some knowledge of the minimizers (if any) in the Lieb–Oxford inequality. This was noticed in [SVGG16] — see also the original paper of Lieb and Oxford [LO81, Appendix A]. In the appendix of this chapter, we briefly explain the argument of Seidl et al. in [SVGG16]. This argument is only formal. In a work in preparation with S. Di Marino (Universita di Genova), which we do not comment here, we make this argument rigorous [DML].*

### Summary of the present chapter

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## §2.1. Introduction

Given a function  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N$ , we study the following *Multimarginal Optimal Transport* (MOT) problem

$$F_{\text{SCE}}(\rho) = \min_{\mathbb{P} \mapsto \rho} \left\{ \int_{(\mathbb{R}^d)^N} \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^s} \mathbb{P}(dx_1, \dots, dx_N) \right\}, \quad (\text{SCE})$$

where  $s > 0$ . Here, the notation “ $\mathbb{P} \mapsto \rho$ ” means that the minimum runs over all (symmetric) probability measures  $\mathbb{P}$  on  $(\mathbb{R}^d)^N$  such that for every  $i = 1, \dots, N$  the marginal of  $\mathbb{P}$  over the  $i$ -th copy of  $\mathbb{R}^d$  in  $(\mathbb{R}^d)^N$  is given by  $\rho/N$ . In quantum chemistry, when  $s = 1$  and  $d = 3$ , **Problem SCE** is coined as the *Strictly-Correlated Electrons* functional [Sei99; SPL99]. This functional is of great interest in *Density-functional Theory* (DFT), an important computational modeling method in quantum physics and chemistry. We refer to [FGGG22; VLGG17] for recent surveys on **Problem SCE** and its applications in DFT.

As any MOT problem, **Problem SCE** admits the following dual formulation

$$F_{\text{SCE}}(\rho) = \sup_{V \text{ s.t. } \int_{\mathbb{R}^d} |V| \rho < \infty} \left\{ E_N(V) - \int_{\mathbb{R}^d} V \rho \right\}. \quad (\text{SCE}_D)$$

Here, the supremum runs over all continuous functions  $V : \mathbb{R}^d \rightarrow \mathbb{R}$  which are integrable with respect to  $\rho$ , and the quantity  $E_N(V)$  is defined as

$$E_N(V) = \inf_{x_1, \dots, x_N} \left\{ \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^s} + \sum_{i=1}^N V(x_i) \right\}. \quad (2.1)$$

Maximizers of **Problem SCE<sub>D</sub>** are known to exist, as shown in [BCDP18; CDMS19], and are called *Kantorovich potentials*. Furthermore, they can be chosen (locally) Lipschitz. Note that  $V$  should be regarded as an external potential applied to the  $N$  classical particles and  $E_N(V)$  as the classical ground-state energy of the resulting system. It has been argued [SGGS07; VLGG17; FGGG22] on physical ground that, when  $\rho$  is supported over the entire space  $\mathbb{R}^d$  and in the case of the Coulomb potential in three space-dimension ( $s = 1$  and  $d = 3$ ), any (regular enough) Kantorovich potential  $V$  should verify the asymptotic

$$\boxed{V(x) \sim -\frac{N-1}{|x|} \text{ as } |x| \rightarrow \infty.} \quad (2.2)$$

This conjecture is rigorously known to hold either when  $N = 2$  and  $\rho$  is spherically symmetric using [Pas13, Cor. 3.1.1], or for an arbitrary number of particles  $N$  in one space-dimension appealing to the results of [CDPDM15], using that in both cases an explicit formula is available for the SCE system.



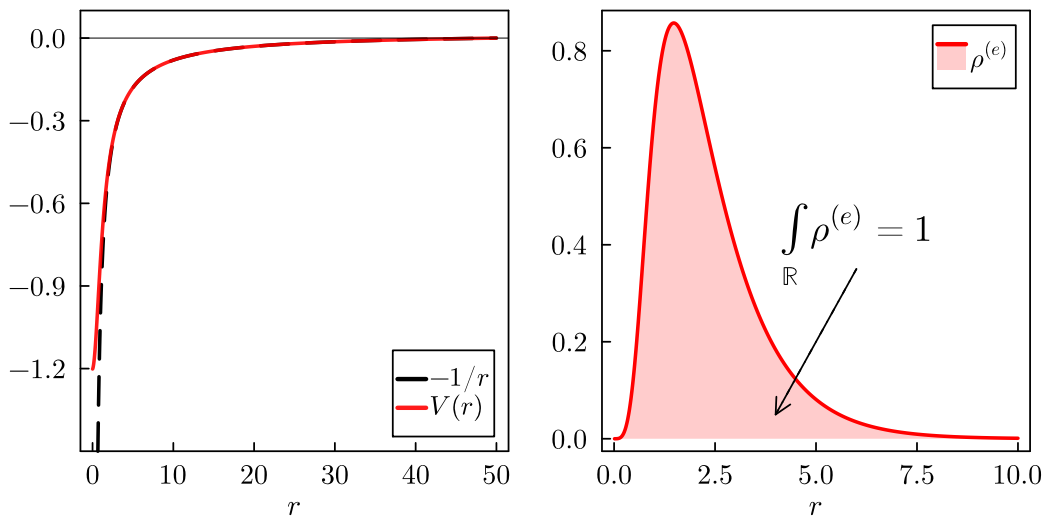


Figure 2.1: Example of a Kantorovich potential for  $N = 2$  with target density  $\rho(x) = ce^{-|x|}$  for some constant  $c$  in three space-dimension. In this case all the computations can be carried [Pas13]. We find that the Kantorovich potential  $V$  verifies the asymptotic (2.2), and that the dual charge has mass  $N - 1 = 1$ , see Corollary 20. We plot here the radial components.

It is also known to hold when  $\rho$  is compactly supported for one well-chosen Kantorovich potential, even though in this specific case, the problem is not entirely well-posed, see Remark 3 in Chapter 1, page 84, and also Remark 12 below.

In the paper, we prove this conjecture in full generality:

**Theorem 19** (Asymptotic of the Kantorovich potential). *Let  $\Omega \subset \mathbb{R}^d$  be an unbounded and connected open set, and let  $\rho \in L^1(\Omega, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N$  be such that  $\rho > 0$  almost everywhere on  $\Omega$ . Let  $V$  be a Kantorovich potential for Problem  $SCE_D$  which is locally Lipschitz on  $\Omega$ . Then, there exists some constant  $C$  such that*

$$V(x) + C \sim -\frac{N-1}{|x|^s} \quad \text{in the limit } |x| \rightarrow \infty \text{ and } x \in \Omega. \quad (2.3)$$

It is known [San15, Prop. 1.15] that the gradients (in the *Fréchet* sense) of two Kantorovich potentials coincide  $\rho$  almost-everywhere. When those potentials are furthermore locally Lipschitz, these gradients identify with their *weak* counterparts (in the *Sobolev*  $W_{\text{loc}}^{1,\infty}$  sense; see, e.g. [Eva10, Thm. 5.8.5]). Since  $\Omega$  is connected, they must then be equal up to an additive constant on  $\Omega$ . Therefore, it is enough to prove Theorem 19 for one such Kantorovich potential. It is moreover known that such a Kantorovich potential exists, see Lemma 22 below.

We also note that the asymptotic (2.3) is of theoretical importance in computational quantum chemistry. Indeed, in the context of strongly-correlated materials, the functional defined by Problem SCE has been proved useful to fashion *exchange-correlation functionals* in DFT [MGG12; Mal+14; MMGG14].

From this perspective, the Kantorovich potential corresponds to the sum of the *exchange-correlation potential* and the *Hartree potential*. Now, it is well-known that those potentials shall verify some requirements, in particular having the asymptotic (2.3) at infinity [TH03].

*Remark 11.* We will see from the proof of Theorem 19 that the specific form of the pair interaction potential  $w$  does not really matter, as long as  $w(x, x') = \tilde{w}(|x - x'|)$  where  $\tilde{w} : [0, \infty) \rightarrow (0, \infty)$  behaves like  $r^{-s}$  at infinity. Here, we have exactly taken the Riesz potential with positive exponent  $s$ , that is  $\tilde{w}(r) = r^{-s}$ .

In [Lel22a] and as reported in Chapter 1, we proved that, in the Coulomb case  $s = d - 2$  in dimension  $d > 2$ , there always exists a Kantorovich potential  $V$  for Problem SCE which is an attractive Coulomb potential induced by some positive charge distribution, coined as the *dual charge*. That is,  $V$  rewrites as  $V(x) = -|\cdot|^{2-d} * \rho^{(e)}(x)$  where  $\rho^{(e)}$  is a positive measure. Using Theorem 19, we were able to prove the following statement.

**Corollary 20** (Total mass of the dual charge). *We suppose that  $s = d - 2$  with  $d > 2$ . Let  $\Omega \subset \mathbb{R}^d$  be an unbounded and connected open set, which we assume to not “shrink in all directions at infinity”, in the sense that  $|A_\Omega| > 0$ , where  $|\cdot|$  is the Lebesgue measure on the sphere  $\mathbb{S}^{d-1}$  and*

$$A_\Omega := \left\{ \xi \in \mathbb{S}^{d-1} : \text{for all } r_0 \text{ there exists } r \geq r_0 \text{ s.t. } r\xi \in \Omega \right\}.$$

Let  $\rho \in L^1(\Omega, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N$  be such that  $\rho > 0$  almost everywhere  $\Omega$ . Let  $\rho^{(e)}$  be a positive measure such that  $-|\cdot|^{2-d} * \rho^{(e)}$  is a Kantorovich potential for Problem SCE<sub>D</sub> which is locally Lipschitz on  $\Omega$ . Then, it must be that

$$\int_{\mathbb{R}^d} \rho^{(e)} = N - 1. \quad (2.4)$$

The intuition for Corollary 20 is that, an electron being fixed, the dual charge needs to exert a force which counterbalances the repulsion due to the other  $N - 1$  electrons, thus leading to the conjectured equality (2.4). The hypothesis regarding the shape of  $\Omega$  at infinity is mainly technical and will become transparent in the proof of the corollary.

There always exists a measure  $\rho^{(e)}$  as in the statement of the corollary, see [Lel22a] or Theorem 1 in Chapter 1, page 83. In fact, such a measure is uniquely determined on  $\Omega$  as  $\rho_{\text{ext}} := -c_d \Delta V$ , where  $V$  is a specific Lipschitz Kantorovich potential of Problem SCE, *i.e.* one that verifies Equation (2.18) below. Here

$$c_d := \frac{d(d-2)\pi^{d/2}}{\Gamma\left(\frac{d}{2} + 1\right)}$$

and  $-\Delta$  is to be understood as (minus) the distributional Laplacian on  $\mathbb{R}^d$ . We prove in Theorem 1 of Chapter 1 that the distribution  $\rho^{(e)}$  defined as such is actually a positive measure.

*Remark 12.* In the case where  $\rho$  is compactly supported, we can always *assume* that (2.4) holds. Indeed, when the support  $\Omega$  of  $\rho$  is compact, we can always

add to  $\rho^{(e)}$  some charge uniformly distributed over a sphere of radius  $R$ , where  $\Omega \subset B_R$ , since the potential induced inside of  $\Omega$  by this additional charge is constant.

The proofs of Theorem 19 and Corollary 20 are consigned in § 2.3.

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## §2.2. Dissociation at infinity

In order to prove Theorem 19, we will appeal to the following theorem.

**Theorem 21** (Dissociation at infinity). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N$ . Let  $\mathbb{P}$  be a symmetric minimizer of Problem SCE. Then, there exists a large enough ball  $B_R \subset \mathbb{R}^d$  of radius  $R$  such that*

$$\mathbb{P}\left((\mathbb{R}^d \setminus B_R) \times (\mathbb{R}^d \setminus B_R) \times \mathbb{R}^{d(N-2)}\right) = 0. \quad (2.5)$$

The above theorem (which is evidently true when  $\rho$  is compactly supported) was mentioned as a conjecture in [FGGG22, Eq. (65)]. It says that, at optimality, as one particle is sent to infinity, all the other remaining particles shall remain in a bounded domain  $\mathbb{P}$  almost-surely. Otherwise stated, “dissociation at infinity” only occurs for one particle at a time. We will provide later in § 2.3.3 a stronger version of this theorem.

The proof of Theorem 21 only relies on the notion of cyclical monotonicity, which we now recall. One can prove [San15, Thm. 1.43] that, given any minimizer  $\mathbb{P}$  of Problem SCE, its support  $\Gamma$  is concentrated on a set  $\Gamma_0$  which is *c-cyclically monotone*, in the sense that for all  $k \in \mathbb{N}$ , all families  $(x_1^i, \dots, x_N^i)$  with  $i = 1, \dots, k$  of points in  $\Gamma_0$ , and all set of permutations  $\sigma_1, \dots, \sigma_N \in \mathfrak{S}_k$  we have

$$\sum_{i=1}^k c(x_1^i, \dots, x_N^i) \leq \sum_{i=1}^k c(x_1^{\sigma_1(i)}, \dots, x_N^{\sigma_N(i)}), \quad (2.6)$$

where

$$c(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|^s}.$$

Here, the meaning of (2.6) is that exchanging the positions of any particles at optimality necessarily leads to an increase of the energy.

In [BCDP18; CDMS19], it is proved that, at optimality, the particles cannot get too close to one another, in the sense that there exists some distance  $\eta > 0$  such that, for any minimizer  $\mathbb{P}$  of **Problem SCE**, we have

$$\mathbb{P} \left( \min_{1 \leq i < j \leq N} |x_i - x_j| < \eta \right) = 0.$$

By continuity of  $c$  away from the diagonals, this entails that (2.6) actually holds on the closure of  $\Gamma_0$ , and hence on the entire support  $\Gamma$  of  $\mathbb{P}$ . Furthermore, in the formulation of **Problem SCE**, one can substitute to the Riesz potential  $|x|^{-s}$  its truncated version, *i.e.*  $\min\{|x|^{-s}, \eta^{-s}\}$ . In what follows, we will write

$$|x|_\eta := \max\{|x|, \eta\},$$

so that the truncated Riesz potential reads  $|x|_\eta^{-s}$ .

*Proof of Theorem 21.* We start with the two-marginal case  $N = 2$  as an illustration of the general argument. Let  $\mathbb{P}$  be a minimizer of **Problem SCE** with support  $\Gamma$ . We proceed *reductio ad absurdum* by assuming that, for all radius  $R > 0$ , we have

$$\mathbb{P} \left( (\mathbb{R}^d \setminus B_R) \times (\mathbb{R}^d \setminus B_R) \right) > 0. \quad (2.7)$$

By definition of the support  $\Gamma$  of  $\mathbb{P}$ , we have

$$\mathbb{P} \left( \left( (\mathbb{R}^d \setminus B_R) \times (\mathbb{R}^d \setminus B_R) \right) \cap \Gamma \right) > 0. \quad (2.8)$$

Therefore, there must exist a sequence  $(x_1^{(k)}, x_2^{(k)}) \in \Gamma$  such that  $x_i^{(k)} \rightarrow \infty$  as  $k \rightarrow \infty$  ( $i = 1, 2$ ). Given any  $(x_1, x_2) \in \Gamma$ , and appealing to the  $c$ -cyclical monotonicity, we have

$$\frac{1}{|x_1 - x_2|^s} \leq \frac{1}{|x_1 - x_2|^s} + \frac{1}{|x_1^{(k)} - x_2^{(k)}|^s} \leq \frac{1}{|x_1 - x_1^{(k)}|^s} + \frac{1}{|x_2 - x_2^{(k)}|^s}. \quad (2.9)$$

We now let  $k \rightarrow \infty$  to obtain the contradiction that for all  $(x_1, x_2) \in \Gamma$

$$0 < \frac{1}{|x_1 - x_2|^s} \leq 0. \quad (2.10)$$

Let us now consider the general case  $N \geq 2$ . Let  $\mathbb{P}$  be a minimizer of **Problem SCE** with support  $\Gamma$ . Note that we can (and will) suppose that  $\mathbb{P}$  is symmetric. Indeed, if  $\mathbb{P}$  is not symmetric, it suffices to consider

$$\tilde{\mathbb{P}}(x_1, \dots, x_N) = \frac{1}{N!} \sum_{\sigma \in \mathfrak{S}_N} \mathbb{P}(x_{\sigma(1)}, \dots, x_{\sigma(N)}).$$

Let us assume that there exists a sequence of configurations in  $\Gamma$  such that exactly  $\mathcal{J} \in \{2, \dots, N\}$  particles escape to infinity. That is, there exists a sequence  $(x_1^{(k)}, \dots, x_N^{(k)}) \in \Gamma$  such that  $x_i^{(k)} \rightarrow \infty$  as  $k \rightarrow \infty$  for  $i = 1, \dots, \mathcal{J}$ , and such that the remaining particles remain in some bounded region. Up to a subsequence and by compactness, we may assume that there exists  $\bar{x}_i \in \mathbb{R}^d$

such that  $x_i^{(k)} \rightarrow \bar{x}_i$  as  $k \rightarrow \infty$  for all  $i = \mathcal{J} + 1, \dots, N$ . Once again appealing to the  $c$ -cyclical monotonicity, for all  $(x_1, \dots, x_N) \in \Gamma$  we have

$$\begin{aligned} & \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{1 \leq i < j \leq N} \frac{1}{|x_i^{(k)} - x_j^{(k)}|_\eta^s} \\ & \leq \sum_{i=2}^N \frac{1}{|x_1^{(k)} - x_i|_\eta^s} + \sum_{2 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} \\ & \quad + \sum_{i=2}^N \frac{1}{|x_1 - x_i^{(k)}|_\eta^s} + \sum_{2 \leq i < j \leq N} \frac{1}{|x_i^{(k)} - x_j^{(k)}|_\eta^s}. \end{aligned} \quad (2.11)$$

In the definition of  $c$ -cyclical monotonicity as given in (2.6), the above corresponds to the choice  $k = 2$  with  $\sigma_1 = (12)$  and  $\sigma_i = \text{id}$  for all  $i = 2, \dots, N$ . Note that, contrary to (2.9), we used the truncated cost above. This is mainly for convenience, to emphasize that all quantities are finite. In particular, after subtracting the interactions between the  $x_2, \dots, x_N$  (resp.  $x_2^{(k)}, \dots, x_N^{(k)}$ ) on both sides of (2.11), we can legally write

$$\sum_{i=2}^N \frac{1}{|x_1 - x_i|_\eta^s} + \sum_{i=2}^N \frac{1}{|x_1^{(k)} - x_i^{(k)}|_\eta^s} \leq \sum_{i=2}^N \frac{1}{|x_1^{(k)} - x_i|_\eta^s} + \sum_{i=2}^N \frac{1}{|x_1 - x_i^{(k)}|_\eta^s}. \quad (2.12)$$

We now let  $k \rightarrow \infty$  to obtain that for all  $(x_1, \dots, x_N) \in \Gamma$

$$\sum_{i=2}^N \frac{1}{|x_1 - x_i|_\eta^s} \leq \sum_{i=\mathcal{J}+1}^N \frac{1}{|x_1 - \bar{x}_i|_\eta^s}. \quad (2.13)$$

Let us comment on the transition between (2.12) and (2.13). We remark that from the fact  $|x_i^{(k)}| \rightarrow \infty$  for all  $i = 1, \dots, \mathcal{J}$ , it does not *a priori* follow that the mutual distances between the particles should diverge, that is  $|x_i^{(k)} - x_j^{(k)}| \rightarrow \infty$ . Nevertheless, we recall that the particles live on the support of an optimal transport plan. Therefore, they minimize the following energy, namely

$$(x_1^{(k)}, \dots, x_N^{(k)}) \in \arg \min \left\{ \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{i=1}^N V(x_i) \right\} \quad (2.14)$$

Because the Kantorovich potential  $V$  admits a limit at infinity — see Lemma 22 below — we see that it is always energetically favorable for two particles which run to infinity while remaining in the minimizing set of the energy to be as far away from each other as possible. In fact, their distance will always diverge to infinity. This justifies the transition between (2.12) and (2.13).

Now, we let  $x_i = x_i^{(k)}$  for  $i = 1, \dots, N$  above. Letting  $k \rightarrow \infty$ , and by the assumption that  $\mathcal{J} > 1$ , we see from the above inequality that the escaping particles cannot contribute to the first-order in (2.13), meaning that

$$\frac{|x_1^{(k)}|_\eta^s}{|x_1^{(k)} - x_i^{(k)}|_\eta^s} \rightarrow 0 \quad (i = 2, \dots, \mathcal{J}). \quad (2.15)$$

Since

$$\frac{|x_1^{(k)}|}{|x_1^{(k)} - x_i^{(k)}|} \geq \frac{|x_1^{(k)}|}{|x_1^{(k)}| + |x_i^{(k)}|} = \frac{1}{1 + \frac{|x_i^{(k)}|}{|x_1^{(k)}|}}, \quad (2.16)$$

it must be that  $|x_1^{(k)}| = o(|x_i^{(k)}|)$ . But, by symmetry of  $\mathbb{P}$ , meaning that for any  $(x_1, \dots, x_N) \in \Gamma_0$ , we have  $(x_{\sigma(1)}, \dots, x_{\sigma(N)}) \in \Gamma_0$  for all  $\sigma \in \mathfrak{S}_N$ , we can switch the indices 1 and  $i$  in (2.15). Reproducing the above argument, we are led to  $|x_i^{(k)}| = o(|x_1^{(k)}|)$ , and therefore to a contradiction. Hence, *reductio ad absurdum*, the thesis of Theorem 21 is proved.  $\square$

## §2.3. Proof of the main results

In this section, we prove that any Kantorovich potential  $V$  which is locally Lipschitz has the conjectured asymptotic behavior at infinity. We start by briefly recalling some important facts regarding the duality theory for Problem SCE.

### §2.3.1. On the duality theory

Given a minimizer  $\mathbb{P}$  of Problem SCE with support  $\Gamma$  and a Kantorovich potential  $V$ , there exists  $\Gamma_1 \subset \Gamma$  with  $\mathbb{P}(\Gamma \setminus \Gamma_1) = 0$  such that

$$\sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{i=1}^N V(x_i) = E_N(V) \quad \text{on } \Gamma_1. \quad (2.17)$$

When  $V$  is continuous, the above equality is valid on the closure  $\overline{\Gamma_1}$ , and therefore on the entire support  $\Gamma$ . Among all the Kantorovich potentials of Problem SCE<sub>D</sub>, one can specify a very special potential, namely one that verifies the following equation

$$V(x_1) = \sup_{x_2, \dots, x_N} \left\{ - \sum_{i=2}^N V(x_i) - \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} \right\}. \quad (2.18)$$

For such a  $V$ , we always have  $E_N(V) = 0$ . We have the following lemma.

**Lemma 22** ([BCDP18, Thm. 3.4 and Thm. 3.6] and Lemma 7 of Chapter 1, page 95). *Let  $\rho \in L^1(\mathbb{R}^d, \mathbb{R}_+)$  with  $\int_{\mathbb{R}^d} \rho = N$ , and let  $V$  be a Kantorovich potential for Problem SCE<sub>D</sub> verifying Equation (2.18). Then,  $V$  is Lipschitz and it satisfies the following limit*

$$\lim_{|x| \rightarrow \infty} V(x) = -E_{N-1}(V) > 0 \quad (2.19)$$

where, following the notation of (2.1), we have

$$E_{N-1}(V) = \inf_{x_2, \dots, x_N} \left\{ \sum_{2 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{i=2}^N V(x_i) \right\}.$$

Recall that two Lipschitz Kantorovich potentials are equal (up to an additive constant) on (the connected components) of the support of  $\rho$  [San15, Prop. 1.15]. Therefore, it follows from the above lemma that the Kantorovich potential  $V$  which verifies Equation (2.18) is unique on  $\Omega$ , where  $\Omega$  verifies the assumptions of Theorem 19. Furthermore, it also follows that any Kantorovich potential which is locally Lipschitz on  $\Omega$  is actually Lipschitz and equal (up to an additive constant) to the  $V$  which verifies Equation (2.18).

### §2.3.2. Proof of Theorem 19

Let  $V$  be the Kantorovich potential which verifies Equation (2.18), and let  $\mathbb{P}$  be a symmetric minimizer of Problem SCE with support  $\Gamma$ . Let us first prove the lower bound asymptotic, that is

$$W(x) := V(x) + E_{N-1}(V) \geq -\frac{N-1}{|x|^s} + o\left(\frac{1}{|x|^s}\right). \quad (2.20)$$

We argue as in (the proof of) Theorem 1 of Chapter 1. Let  $(x_1^{(k)}, \dots, x_N^{(k)}) \in \Gamma$  be such that  $x_1^{(k)} \rightarrow \infty$  as  $k \rightarrow \infty$ . Such a sequence exists as the support  $\Gamma$  of  $\rho$  is unbounded by hypothesis. According to Theorem 21 regarding the dissociation at infinity, all the other particles must stay in a bounded region  $\mathbb{P}$  almost-surely as  $x_1^{(k)}$  goes to infinity. Up to a subsequence and by compactness, we may therefore assume that  $x_i^{(k)} \rightarrow \bar{x}_i$  for some  $\bar{x}_i \in \mathbb{R}^d$  ( $i = 2, \dots, N$ ). We have

$$\sum_{1 \leq i < j \leq N} \frac{1}{|x_i^{(k)} - x_j^{(k)}|_\eta^s} + \sum_{i=1}^N W(x_i^{(k)}) = E_N(W) \quad (2.21)$$

According to Lemma 22,  $w$  is continuous and  $W(x) \rightarrow 0$  as  $x \rightarrow \infty$ . Hence, we obtain by letting  $k \rightarrow \infty$  above that

$$\sum_{2 \leq i < j \leq N} \frac{1}{|\bar{x}_i - \bar{x}_j|_\eta^s} + \sum_{i=2}^N W(\bar{x}_i) = E_N(W). \quad (2.22)$$

We now select  $x_i = \bar{x}_i$  ( $i = 2, \dots, N$ ) in Equation (2.18) as verified by  $V$ . We obtain using the above equality that for all  $x \in \mathbb{R}^d$

$$W(x) \geq -\sum_{i=2}^N \frac{1}{|x - \bar{x}_i|_\eta^s} = -\frac{N-1}{|x|^s} + o\left(\frac{1}{|x|^s}\right). \quad (2.23)$$

Therefore, the lower bound asymptotic (2.20) is proved. Now, the upper bound asymptotic is obtained as follows. Since  $W$  vanishes at infinity, we have  $E_N(W) \leq E_{N-1}(W)$ . Furthermore, by definition of  $E_{N-1}(W)$ , for all  $x_1, \dots, x_N \in \mathbb{R}^d$  we have

$$E_{N-1}(W) \leq \sum_{2 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{i=2}^N W(x_i). \quad (2.24)$$



According to (2.17), we have

$$\sum_{1 \leq i < k \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{i=1}^N W(x_i) = E_N(W) \quad \text{on } \Gamma. \quad (2.25)$$

From (2.24) and the above equality, we finally obtain

$$W(x_1) \leq \sum_{i=2}^N \frac{1}{|x_1 - x_i|_\eta^s} \quad \text{for all } (x_1, \dots, x_N) \in \Gamma. \quad (2.26)$$

By the assumption that  $\rho > 0$  almost everywhere on  $x \in \Omega$ , and since the above inequality holds over the support  $\Gamma$  of  $\mathbb{P}$ , we then obtain the upper bound asymptotic on  $W$  as sought-after.  $\square$

### §2.3.3. Dissociation and binding inequalities

From our main theorem regarding the asymptotic behavior at infinity of the Kantorovich potential, we deduce the following result, which is of independent interest.

**Corollary 23** (Dissociation and binding inequalities). *Let  $V$  be Kantorovich potential for Problem  $SCE_D$  which is Lipschitz, vanishes at infinity, i.e.  $\lim_{x \rightarrow \infty} V(x) = 0$ , and satisfies the asymptotic*

$$V(x) \sim -\frac{N-1}{|x|^s} \quad \text{in the limit } |x| \rightarrow \infty \text{ and } x \in \Omega,$$

where  $\Omega$  is an unbounded and connected open set. Such a  $V$  exists according to Theorem 19 and Lemma 22. Let  $E_K(V)$  be the infimum defined over  $(\mathbb{R}^d)^K$  as in (2.1), with the convention that  $E_1(V)$  is the infimum of  $V$  over  $\mathbb{R}^d$ . Then, the following items are veracious:

1. We have the following set of inequalities:

$$E_N(V) = E_{N-1}(V) < E_{N-2}(V) < \dots < E_1(V); \quad (2.27)$$

2. The ground-state energy  $E_K(V)$  is attained in a compact set for all number of particles  $K = 1, \dots, N$ ;
3. There exists a large enough radius  $R > 0$  such that

$$\Omega \cap \left( (\mathbb{R}^d \setminus B_R) \times (\mathbb{R}^d \setminus B_R) \times \mathbb{R}^{d(N-2)} \right) = \emptyset. \quad (2.28)$$

Here the set  $\Omega \subset (\mathbb{R}^d)^N$  is the set of minimizers of  $E_N(V)$  defined as

$$\Omega := \arg \min_{x_1, \dots, x_N} \left\{ \sum_{1 \leq i < j \leq N} \frac{1}{|x_i - x_j|_\eta^s} + \sum_{i=1}^N V(x_i) \right\}. \quad (2.29)$$

A potential  $V$  which behaves like  $-(N-1)|x|^{-s}$  at infinity can bind  $N-1$  or less particles. The equality  $E_N(V) = E_{N-1}(V)$  in (2.27) is rather remarkable, as it implies that the Kantorovich potential  $V$  can bind one more additional electron, leading to a total of  $N$  (or less) particles. This shows that the minimizer(s) and the Kantorovich potential for **Problem SCE** are rather subtle objects.

The Item 3 in the above corollary is a refinement of our first dissociation theorem, namely Theorem 21. Recall that if  $V$  is a Kantorovich potential, then it verifies the optimality condition (2.17). It follows that the support of  $\mathbb{P}$  is contained in  $\Omega$  as defined in (2.29). Nevertheless, we emphasize that it need not be true that the support is equal to the whole of  $\Omega$ . For instance, in one space-dimension with the Coulomb potential  $-|r|$ , the support can be a proper subset of  $\Omega$ , see Appendix A of Chapter 1.

Finally, the strict inequalities in (2.27) say that placing one particle at infinity always increases the energy of the system with  $N-1$  particles. The physical intuition is that, if  $K > 1$  particles are placed at infinity where the potential  $V$  behaves like  $-(N-1)|x|^{-s}$ , upon those particles is exerted an attractive potential which eventually pull them back in the finite vicinity, since the remaining electrons induce a potential  $-(N-K)|x|^{-s}$  at infinity, and that  $(N-1) - (N-K) > 0$ . This is the classical argument of Zhislin–Sigalov in [ZS65], where the authors proved a stability result for neutral or positively-charged atoms and molecules using the so-called *Hunzinker–van Winter–Zhislin* (HVZ) theorem, which is an important result in quantum physics [Lew11; Fri03] but is an easy fact for classical systems like ours. We now reproduce this argument to prove the corollary.

*Proof of Corollary 23.* We note that  $E_N(V)$  is attained by definition, since  $V$  is a Kantorovich potential, and that the equality  $E_N(V) = E_{N-1}(V)$  follows from the fact that the support  $\Omega$  of  $\rho$  is unbounded. We note that  $E_K(V) < E_{K-1}(V)$  implies that  $E_K(V)$  is attained. Otherwise, given any minimizing sequence  $x_1^{(k)}, \dots, x_K^{(k)}$  for  $E_K(V)$ , up to a permutation on the indices we could extract a subsequence such that the first  $K_0 \in \{1, \dots, K-1\}$  particles  $x_1^{(k)}, \dots, x_{K_0}^{(k)}$  converge to a minimizer of  $E_{K_0}(V)$  by compactness. This would imply the contradiction  $E_{K_0}(V) = E_K(V) < E_{K-1}(V) \leq E_{K_0}(V)$  since  $V$  vanishes at infinity. Since  $V$  is a continuous function which vanishes at infinity and which is negative somewhere according to its asymptotic behavior at infinity, the infimum of  $V$  must be attained on some compact set, *i.e.*  $E_1(V)$  is attained. Therefore, if we can prove the set of strict inequalities in (2.27), the Item 2 will immediately follow. We proceed by induction on the number of particles  $K$ . We start by proving that  $E_2(V) < E_1(V)$ . Let  $x^*$  be a minimizer of  $E_1(V)$ , that is  $V(x^*) = \min V$ . By definition of  $E_2(V)$ , for all  $x \in \mathbb{R}^d$  we have

$$E_1(V) + V(x) + \frac{1}{|x - x^*|^s} \geq E_2(V). \quad (2.30)$$

By the asymptotic behavior of  $V$ , we have

$$V(x) + \frac{1}{|x - x^*|^s} = -\frac{N-2}{|x|^s} + o\left(\frac{1}{|x|^s}\right) \quad \text{in } \Omega. \quad (2.31)$$

Therefore, for large enough  $|x| \gg 1$  in  $\Omega$ , we obtain  $E_2(V) < E_1(V)$ . As mentioned above, this implies that  $E_2(V)$  is attained. We then proceed by induction, *i.e.*  $E_{K-1}(V)$  is attained for some  $\bar{x}_1, \dots, \bar{x}_{K-1}$ , and we have

$$E_{K-1}(V) + V(x) + \sum_{i=1}^{K-1} \frac{1}{|x - \bar{x}_i|^s} \geq E_K(V). \quad (2.32)$$

Once again, by the asymptotic behavior of  $V$ , we have

$$V(x) + \sum_{i=1}^{K-1} \frac{1}{|x - \bar{x}_i|^s} = -\frac{N-K}{|x|^s} + o\left(\frac{1}{|x|^s}\right) \quad \text{in } \Omega. \quad (2.33)$$

Therefore, for large enough  $|x| \gg 1$  in  $\Omega$ , we obtain  $E_K(V) < E_{K-1}(V)$ , and therefore that  $E_K(V)$  is attained. Hence, the Item 2 is proved by induction, and consequently the Item 1. The Item 3 follows immediately from the Item 1.  $\square$

### §2.3.4. Proof of Corollary 20

Finally, we conclude this paper with the proof that if  $\rho^{(e)}$  is a measure such that  $U^{\rho^{(e)}} := -|\cdot|^{d-2} * \rho^{(e)}$  is a Kantorovich potential for Problem SCE<sub>D</sub> which is locally Lipschitz on the support  $\Omega$  of  $\rho$ , where  $\Omega$  verifies the assumptions mentioned in the corollary, then  $\rho^{(e)}(\mathbb{R}^d) = N - 1$ .

The upper bound  $\rho^{(e)}(\mathbb{R}^d) \leq N - 1$  was already proved in Theorem 1 of Chapter 1, where it follows from the fact that, given any compactly supported (finite) measure  $\mu$ , we have

$$U^\mu(x) \sim -\frac{\mu(\mathbb{R}^d)}{|x|^{d-2}} \quad \text{in the limit } x \rightarrow \infty. \quad (2.34)$$

Nonetheless, this asymptotic need not be true for a non-compactly supported measure  $\mu^1$ . Nevertheless, one can prove [KM79, Cor. 3.4] that, under the assumption

$$\int_{\mathbb{R}^d} \frac{\mu(dx)}{|x|^{d-2}} < \infty, \quad (2.35)$$

there exists a “small” Borel set  $E \subset \mathbb{S}^{d-1}$  (in the sense that  $C_d(E) = 0$ , where  $C_d(E)$  is the *capacity* of  $E$ , see [Lan72, Chap. II]) such that

$$\lim_{r \rightarrow \infty} r^{2-d} U^\mu(r\xi) = -\mu(\mathbb{R}^d). \quad (2.36)$$

for all  $\xi \in \mathbb{S}^{d-1} \setminus E$ . Notice that the hypothesis (2.35) is verified for  $\rho^{(e)}$  as  $U^{\rho^{(e)}}(0) < \infty$ , since  $U^{\rho^{(e)}}$  is continuous. Now, by the assumption on  $\Omega$  stated in Corollary 20, we have  $|A_\Omega| > 0$ , where  $|\cdot|$  denotes the Lebesgue measure on the sphere  $\mathbb{S}^{d-1}$  and where

$$A_\Omega := \left\{ \xi \in \mathbb{S}^{d-1} : \text{for all } r_0 \text{ there exists } r \geq r_0 \text{ s.t. } r\xi \in \Omega \right\}.$$

---

<sup>1</sup>For instance, consider  $\mu = \sum_i a_i \delta_{x_i}$  where  $(x_i)_i$  is a sequence of points such that  $x_i \rightarrow \infty$  as  $i \rightarrow \infty$ , and  $(a_i)_i$  is a sequence of positive reals such that  $\sum_i a_i < \infty$ .

In particular, since any measurable set  $B \subset \mathbb{S}^{d-1}$  of null capacity verifies  $|B| = 0$  [Lan72, Thm. 2.1], we have  $|A_\Omega \setminus E| > 0$ . Therefore, there exists some direction  $\xi_0 \in A_\Omega \setminus E$  and a sequence  $(r_i)_{i \geq 0}$  of positive reals such that  $\lim_i r_i = \infty$  and

$$\lim_{i \rightarrow \infty} r_i^{2-d} U^{\rho^{(e)}}(r_i \xi_0) = -\rho^{(e)}(\mathbb{R}^d), \quad (2.37)$$

and such that  $r_i \xi_0 \in \Omega$  for all  $i \geq 0$ . Using Theorem 19, we have

$$\lim_{i \rightarrow \infty} r_i^{2-d} U^{\rho^{(e)}}(r_i \xi_0) = -(N - 1). \quad (2.38)$$

The thesis of Corollary 20 is therefore proved.  $\square$

## §A. Application: minimizers of the Lieb–Oxford inequality

In this Appendix, we briefly mention how the asymptotic that we have proved in this chapter can be used to gain some knowledge on the minimizers (if any) of the *Lieb–Oxford inequality*. We reproduce here a heuristical argument due to Seidl *et al.* in [SVGG16] — see also the original paper of Lieb and Oxford [LO81, Appendix A]. In a joint work in preparation with S. Di Marino, we make this argument rigorous [DML]. We do not comment here on our proof.

The Lieb–Oxford inequality [Lie79; LO81; Sei+22; LS09] is an important result in quantum physics, which states that there exists a constant  $C_{\text{LO}}(d, s)$  for all dimension  $d \geq 1$  such that

$$F_{\text{SCE}}(\rho) - \frac{1}{2} \iint_{\mathbb{R}^d \times \mathbb{R}^d} \frac{\rho(x)\rho(x')}{|x - x'|^s} \geq -C_{\text{LO}}(d, s) \int_{\mathbb{R}^d} \rho^{1+s/d} \quad (2.39)$$

for all  $\rho \in L^1 \cap L^{1+s/d}(\mathbb{R}^d)$ . Here  $F_{\text{SCE}}(\rho)$  is the SCE functional as defined at the very beginning of this chapter — that is, the multimarginal optimal transport with Riesz cost. Although it was originally proved in dimension  $d = 3$  in the case of the Coulomb potential  $s = 1$ , the inequality holds in all dimension  $d$  for all  $0 < s < d$  as proved in [LNP16], as well as for the logarithmic interaction  $s = 0$  in dimension  $d = 2$ , as shown in [Lew+15]. It should be emphasized that the constant  $C_{\text{LO}}(d, s)$ , which we take to be *smallest* possible constant for which the above inequality is verified, is *universal*, which is remarkable. The search for this optimal constant has been an active field of research [SVGG16; KLCH99; Sei+22; LL15; LLS22]. The knowledge of this constant is important in the context of DFT, where the inequality is used for the calibration of exchange-correlation functionals [LP93; PBE96; PS22; Sun+16]. So far, the best bounds on this constant in the case  $s = 1$  and  $d = 3$  are given by

$$1.4442 \leq C_{\text{LO}}(3, 1) \leq 1.5765.$$

The upper bound was obtained very recently in [LLS22]. The lower bound was conjectured in [LP93] and proved recently in [LLS19a; CP19]. It has been conjectured [LP93; OC07; R +09] that  $C_{\text{LO}}(3, 1) = -\zeta_{\text{BCC}}(1)$ , where  $\zeta_{\text{BCC}}$  is

the *Epstein zeta function* of the body-centered cubic lattice in  $\mathbb{R}^3$ , and that the associated trial state is the *Uniform Electron Gas*. We refer to the Appendix C of Chapter 1 for details on this matter.

We note that the inequality was stated above for any density  $\rho$ , and in particular it holds for any number of particles, *i.e.*  $\int_{\mathbb{R}^d} \rho = N \in \mathbb{N}$ . If we constraint the number of particles  $N$  in the inequality, the constant  $C_{\text{LO}}(d, s)$  will depend on  $N$ . A question is the nature of the density  $\rho$  which achieves this optimal constant in the inequality (2.39) at fixed  $N$ . Although it is not known at the present that a minimizer exists in full generality, in [SVGG16] it was shown heuristically that minimizers are necessarily compactly supported. The argument, also found in [LO81, Appendix A] in the case  $N = 1$ , which uses the asymptotic of the Kantorovich potential as proved above, is the following. If we write the Euler–Lagrange equation for the optimal density  $\rho$  in the Lieb–Oxford inequality (2.39), we obtain formally

$$V_{\text{SCE}}(x) - \int_{\mathbb{R}^d} \frac{\rho(x')}{|x - x'|^s} dx' = C_{\text{LO}}(d, s) \left(1 + \frac{s}{d}\right) \rho^{s/d}(x) \quad \text{for all } x \in \text{supp}(\rho). \quad (2.40)$$

Here, the quantity  $V_{\text{SCE}}$  is the functional derivative of the functional  $F_{\text{SCE}}$  with respect to the density  $\rho$ . The quantity  $V_{\text{SCE}}$  should therefore be given, at least formally, by the Kantorovich potential for the associated optimal transport [San15]. Using the asymptotic that was proved in this chapter, and the fact that

$$\int_{\mathbb{R}^d} \frac{\rho(x')}{|x - x'|^s} dx' = \frac{N}{|x|^s} + o\left(\frac{1}{|x|^s}\right), \quad (2.41)$$

we obtain from the Euler–Lagrange equation (2.40) that the density  $\rho$  behaves as

$$\rho(x) \sim \frac{C}{|x|^d} \quad (2.42)$$

for some constant  $C$  in the large  $|x|$  limit. If the support of  $\rho$  is not compact, then it follows that  $\rho$  cannot be integrable, which is a contradiction.

## CHAPTER 3

# PHASE TRANSITIONS IN ONE-DIMENSIONAL RIESZ GASES WITH LONG-RANGE INTERACTION

*In this chapter, which reproduces our paper Phase transitions in one-dimensional Riesz gases with long-range interaction<sup>1</sup>, we provide numerical evidence for the existence of a phase transition with respect to the temperature in the one-dimensional Riesz gases with non-singular pair interaction, that is particles on the line interacting via the potential  $-|r|^{-s}$ , where  $s \in (-1, 0)$ . This work was initiated following a question asked in [Lew22a] concerning the existence of a transition curve interpolating between the phase diagrams of the Coulomb gas  $s = -1$  and that of the Dyson log-gas  $s = 0$ . Our results confirm this prediction.*

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### §3.1. Introduction

It is well-known from the celebrated theorem of Hohenberg–Mermin–Wagner [Hoh67; MW66; Mer67; Mer68], as well as from an earlier result due to Van Hove [VH50], that in one and two space-dimensions continuous symmetries can never be spontaneously broken at finite temperature, because long-range correlations are destroyed by thermal fluctuations. Nevertheless, a crucial assumption to these results is the short-range nature of the pair interaction. When the interaction remains strong at long distance, long-range fluctuations can persist in the thermodynamic limit, thus allowing for the existence of phase transitions. A seminal example was given by Dyson [Dys69] who proved the existence of a phase transition in the 1D *Ising Ferromagnet* with low-decaying interactions between spins. Overall, many examples of 1D statistical lattice models where some breaking of symmetry occurs at finite temperature are known [Kit69; CW81; GSS03; SHK17; Sar+18; CS04]. However, similar examples in the continuum seem much more scarcer in the literature. The 1D *Coulomb gas*, which is a remarkable and completely integrable model, is an example of a one-dimensional system of classical particles in the continuum for which the translational-symmetry is broken at all temperature [Kun74; AM80] — see also [BL02; JJ14] in the quantum case. One can also mention the works [Joh91; Joh95] — although the nature of the transition is different than that of the one here sought.

In this chapter, we consider 1D *Riesz gases*, that is particles on the line interacting through the pair potential  $v_s(r) = \pm|r|^{-s}$ , together with a uniform neutralizing background in the spirit of Jellium [Lew22a]. We focus on the non-singular case, that is where the exponent  $s$  ranges within  $(-1, 0)$ , in which case the sign of the interaction is chosen negative so as to make  $v_s$  a repulsive potential. We remark that in this case, the potential does not decay at infinity. We provide numerical evidence which presumably rules in favor of the existence of a phase transition with respect to the temperature occurring at finite temperature. At high temperature, we found the pair correlation  $g(r)$  to converge monotonically to the average density  $\rho$  at large distance as in a fluid, whereas  $g(r)$  displays long-lasting oscillations in the low temperature regime, accounting for the existence of a long-range order. At low enough temperature, we found the system to display crystalline features. From a closer investigation, we were led to suspect the existence of two distinct critical temperatures, hereafter denoted  $\tilde{T}_s$  and  $T_s$ . The first one separates between a fluid and a quasisolid phase reminiscent of the *Berezinski–Kosterlitz–Thouless* (BKT) transition [KT73; Her07; Mud14], and another one below which the system is a true solid — see Figure 3.1.

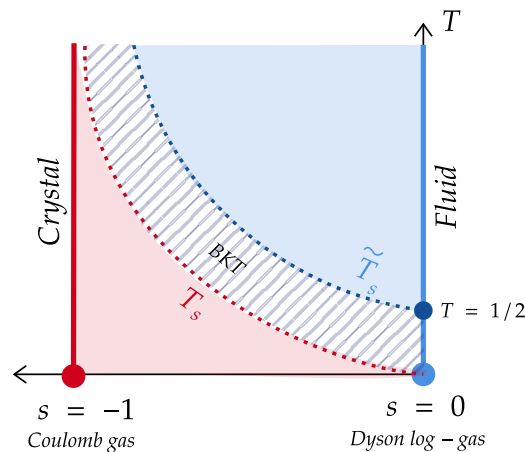


Figure 3.1: Schematic phase diagram of 1D Riesz gases with respect to the temperature  $T$  and the exponent  $s$  of the interaction, following the intuition of [Lew22a]. The 1D Riesz gas exhibits a phase transition at finite temperature  $\tilde{T}_s > 0$  separating between a fluid phase (blue area) and an ordered phase (red and hatched areas). At low enough temperature  $T \ll \tilde{T}_s$ , the system displays crystalline order (red area), whereas in the regime  $0 \ll T < \tilde{T}_s$  we suspect a quasi-ordered phase of a BKT type. Therefore, we believe in the existence of two sets of critical temperatures, namely  $\tilde{T}_s$  which separates between a fluid and a quasisolid phase, and  $T_s$  below which the Riesz gas is a solid — *i.e.* crystal. The limiting behaviour of the critical temperatures with respect to  $s$  are consistent with the phase diagrams of the Coulomb gas and the Dyson log-gas, corresponding to  $s = -1$  and  $s = 0$  respectively.

One-dimensional systems, despite their apparent oversimplificative physical traits, have been a continuously renewed source of exciting physics [BS12; LM67]. Such models are usually more accessible to analytical calculations while being able to describe to a certain extent many problems of actual physical relevance. As for the 1D Riesz gases, they are interesting as they can be seen as the most natural interpolation family between two important integrable models, both of which having received great shares of interest in physics and mathematics. Indeed, in the Coulomb case  $s = -1$ , one recovers as mentioned above the *1D Jellium*, which is also called *Coulomb gas* or *1D One-Component Plasma* (1dOCP). This is a beautiful and solvable statistical model which has been rather thoroughly investigated in the literature [AM80; AJJ10; CGZJ20; Cho75; Kun74]. In particular, the 1D Jellium is known to be crystallized at all temperatures [Kun74; AM80]. On the other hand, in the limit  $s \rightarrow 0^-$ , by considering the first-order term of the pair interaction  $v_s(r)$ , we recover the *Dyson log-gas* [Dys62a; Dys62b; Dys62c; DM63], that is particles on the line interacting *via* the logarithmic interaction  $-\ln|r|$ . This model is of particular importance and regularly occurs in different areas of physics and mathematics. In the context of *random matrix theory*, it is referred to as the  $\beta$ -ensemble or *sine- $\beta$  process* [For10; VV09]. For the special values  $\beta = 1, 2$  and  $4$ , one



recovers respectively the GOE (Gaussian Orthogonal Ensemble), GUE (Unitary, *mutatis mutandis*), and GSE (Symplectic, *idem*) ensembles [For10]. The log-gas is interesting from a statistical physics standpoint as an integrable toy model of particles interacting through a long-range and singular potential. We refer to the rather extensive [Lew22a, Sec. V.C] and the references therein, as well as the monograph [For10] for a very detailed account on the matter. As for its expected phase diagram, the Dyson log-gas is known to be crystallized at zero temperature [SS15], and it is believed that translation-invariance can never be broken at finite temperature [RW90]. A rigorous proof of this statement is given in [EHL21] in the case of stationary point processes [Der19], thus accounting for the case of the thermodynamic limit of the log-gas on the circle, that is with periodic boundary conditions. Our work is then motivated by a question asked in the recent review [Lew22a], where it is wondered whether or not there exists a smooth transition curve between those two limiting cases, namely the Coulomb gas  $s = -1$  and the Dyson log-gas  $s = 0$ . Our findings confirm this prediction.

*Remark 13* (BKT transition for  $s > 0$ ). In what follows, we will only investigate the case of negative exponents  $s \in (-1, 0)$ . For  $s > 0$ , it is expected that the translational symmetry will never be broken [Lew22a] — we also refer to § 5 of our introductory chapter, see 66. This is known rigorously for  $s > 2$  [Pap87]. Nevertheless, it might be that the BKT transition — which is not associated to a broken symmetry — that appears for the Dyson log-gas at  $T = 1/2$  (see § 3.2.3 below) and which, according to our results, also exists for  $s < 0$  (see Figure 3.1) does not cease to exist for  $s > 0$ , at least up to some threshold value of  $s$ . It would be interesting to investigate this question.

## §3.2. Riesz, Coulomb and Dyson gases

In this section, we define the periodic Riesz gases in one space-dimension. We discuss the special cases of the Coulomb gas and of the Dyson log-gas. As a sanity-check of our algorithm, which we will use later to study the Riesz gas with general exponent  $-1 < s < 0$ , we present numerics on the log-gas which are seen to be consistent with known — or at least suspected — theoretical results. These numerics might be of independent interest to some readers.

### §3.2.1. Definition of the periodic 1D Riesz gases

In the long-range case  $s < 1$ , the periodic 1D Riesz gas is defined as follows. We consider  $N$  particles constrained to the segment  $\ell_L = [0, L]$  and we impose periodic boundary conditions to suppress possible boundary effects in our numerical experiments. In the spirit of Jellium [Lew22a], we add a compensating uniform background of opposite charge with density  $\rho = N/L$  to ensure charge neutrality whence summability. In the periodic setting, this amounts to deleting the Fourier zero mode of the interaction potential, see below (3.1). A key parameter in the study of phase transitions is  $\Gamma^{-1} := \rho^{-s}T$ , where  $T$  is the effective temperature of the system. By scaling we will suppose without loss

of generality that  $\rho = 1$ , otherwise stated that  $N = L$ , so that our parameter of interest is the sole effective temperature  $T$ . The associated periodic Riesz potential  $\tilde{v}_{s,L}$  can be analytically expressed using special functions [Lew22a] (see Remark 14 below) contrary to higher dimensions where one needs to resort to some numerical computations often relying on *Ewald summation*. This potential is defined by its Fourier transform [Lew22a, Sec. IV.A.2] up to here unimportant multiplicative constant as

$$\widehat{\tilde{v}_{s,L}}(k) = \sum_{\substack{k \in 2\pi\mathbb{Z}/L \\ k \neq 0}} \frac{\delta_k}{|k|^{1-s}}. \quad (3.1)$$

The 1D Riesz gas is then formally defined as the system obtained in the thermodynamic limit, that is by considering the large  $N$  limit of the canonical ensemble  $Q_N$  defined as the Gibbs measure with density

$$Q_N(r_1, \dots, r_N) = \frac{1}{Z(s, \beta, N)} \exp\left(-\beta \sum_{1 \leq i < j \leq N} \tilde{v}_{s,N}(r_i - r_j)\right), \quad (3.2)$$

where  $Z(s, \beta, N)$  is the usual *partition function*, that is the normalizing constant such that  $Q_N$  is a probability measure on the  $N$ -torus. Here  $\beta$  is the inverse temperature, that is  $\beta = 1/T$ . We can define the *canonical free energy* of the 1D Riesz gas as the thermodynamic limit

$$f(s, \beta) := \lim_{N \rightarrow \infty} \frac{-\beta^{-1} \log Z(s, \beta, N)}{N}. \quad (3.3)$$

The existence of this limit at all temperature was proved in [Lew22a] extending an argument of [Har+15]. At zero temperature the energy per unit length is exactly known, and the system is crystallized [BHS19]. We also mention the works of Serfaty *et al.* [Ser15; Lew22a] where the cases  $s < 0$  are not treated *sic* but are covered by the theory to some extent [Ser]. To the best of our knowledge, no other theoretical results are rigorously known except for those mentioned above. In particular, the convergence of the correlation functions in the thermodynamic limit seems to be unknown for  $-1 < s < 0$  at the present time. We recall that the *k-point correlation function*  $\rho^{(k)}(r_1, \dots, r_k)$  is defined as

$$\rho^{(k)}(r_1, \dots, r_k) = \frac{N!}{(N-k)!} \int_{\mathbb{R}^{N-k}} Q_N(r_1, \dots, r_k, r'_{k+1}, \dots, r'_N) dr'_{k+1} \dots dr'_N \quad (3.4)$$

While the correlation functions are very important in the study of phase transitions, they are also very useful from a mathematical standpoint as they completely characterise the limiting object obtained from the canonical ensemble  $Q_N$  as one considers the thermodynamic limit  $N \rightarrow \infty$ . This limiting object is a (*Gibbs point process*) [Lew22a; Der19]. We emphasize that the question of its existence and *casu quo* of its uniqueness — related to the (non-)existence of phase transitions — while well-studied in the short-range case  $s > d$  in any dimension  $d$ , see [Rue99; Geo76; Geo11; DM67], is a complicated and subtle

problem which remains mainly open in the long-range case  $s < d$ , see [Lew22a, Sec. III]. In dimension  $d = 1$ , it was only very recently studied by Dereudre and Vasseur [DV23] and Boursier [Bou22] in the case  $0 < s < 1$ . In the logarithmic case  $s = 0$ , it is studied in [Der+21]. To the best of our knowledge, the case of negative exponents  $s < 0$  seems to have been eluded in the literature so far, at the exception of the Coulomb case  $s = -1$ , which has been extensively studied, see *e.g.* [Kun74; Cho75; AM80; Len04b; AJJ10; CGZJ20].

In what follows, we will focus our attention on the two-point correlation  $\rho^{(2)}(r, r')$  which we simply call the *pair correlation*. In the case of the periodic Riesz gases, the correlation between two particles only depends on their distance  $|r - r'|$  from one another, so that the pair correlation can be written as a function of a single variable, that is  $g(r) := \rho^{(2)}(0, r)$ . The function  $g(r)$  is an important quantity in statistical mechanics, especially in the study of phase transitions. It describes how the density of particles varies as a function of distance from a given reference particle. For example, in the case of a perfect crystal, the position of a single particle determines the positions of all the other particles — which are obtained from the position of the reference particle by translation with respect to the crystal period or, in full generality, by translation with respect to the primitive vectors describing the crystal lattice in general dimension  $d \geq 1$ . In this case the pair correlation  $g(r)$  is a periodic function with sharp maxima at the lattice site. At zero temperature it is simply a Dirac comb. On the other hand, in the case of a perfect fluid as an ideal gas, the particles are independent of each other. Therefore the pair correlation is a constant function, that is  $g \equiv 1$ . More generally, in the absence of long-range order, the density fluctuations between two particles should decrease rapidly at large distances, *i.e.*  $g(r)$  should converge rapidly to the average density  $\rho$ , whereas in the presence of long-range order,  $g(r)$  should display a slower decay or oscillations at large  $r$ .

Another important quantity to consider, and closely related to the pair correlation  $g(r)$ , is the (*static*) *structure factor*  $S(k)$  [HM13, Chap. 4]. In condensed matter physics and crystallography, the structure factor is a function that contains much relevant information about how a given material scatters an incident beam [DB08; GH04; Sir+89; Yar+73]. The structure factor is defined in the thermodynamic limit  $N \rightarrow \infty$  as the Fourier transform of the truncated pair correlation  $g(r) - 1$ , namely

$$S(k) := 1 + \frac{1}{2\pi} \int_{\mathbb{R}} e^{-irk} (g(r) - 1) dr. \quad (3.5)$$

In practice, the pair correlation  $g(r)$  is computed by determining experimentally the structure factor through scattering methods and expressing  $g(r)$  as the inverse Fourier transform of  $S(k)$ , that is

$$g(r) = 1 + \int_{\mathbb{R}} e^{irk} (S(k) - 1) dk. \quad (3.6)$$

In the finite length  $N < \infty$ , the definition (3.5) should be modified accordingly by considering the Fourier transform on the circle  $L^2(\mathbb{R}/N\mathbb{Z})$ . By appealing to the parity of  $g(r)$ , we get

$$S(k) := 1 + \frac{2}{N} \int_0^{N/2} \cos(2\pi kr) (g(r) - 1) dr, \quad k \in \mathbb{Z}/N. \quad (3.7)$$

If  $g(r)$  is oscillating, the structure factor should have a peak at  $k = 1$  or more generally at any multiple of the period of  $g(r)$ . In condensed matter physics and crystallography, such a peak is sometimes called a *Bragg peak*. On the other hand, if the pair correlation rapidly converges to the average density of our system, as in a fluid, the structure factor should be a smooth function of the wavenumber  $k$ .

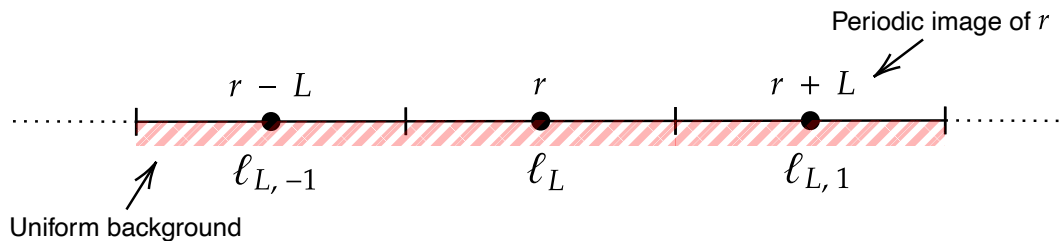


Figure 3.2: The periodic Riesz potential  $\tilde{v}_{s,L}$  is obtained by considering that each particles located at  $r \in \ell_L$  interacts with its periodic images in the  $\ell_{L,k}$  and the uniform background.

*Remark 14* (Periodic Riesz potentials). The periodic Riesz potential  $\tilde{v}_{s,L}$  on the line is defined, as for any periodization of a general potential, as the sum of the interactions between a particle located at  $r \in \ell_L$  and its periodic images in the segments  $r_k \in \ell_{L,k}$  that is  $r_k := r + kL$  for all  $k \in \mathbb{Z}$  (see Figure 3.2). Therefore, we have

$$\tilde{v}_{s,L}(r) = \sum_{k \in \mathbb{Z}} v_s(r + Lk). \quad (3.8)$$

We remark that in the short-range case  $s > 1$ , this infinite sum is convergent and is closely related to the *Hurwitz zeta function*  $\zeta(s, r)$ , as it can be expressed as

$$\tilde{v}_{s,L}(r) = \zeta(s, r) + \zeta(s, 1 - r) \quad \text{when } s > 1. \quad (3.9)$$

In the long-range case  $s < 1$  the resulting series is evidently divergent. To ensure summability — at least when  $s > -1$  — one may add in the spirit of Jellium a uniform background of opposite charge over each  $\ell_{L,k}$  in such a way as to ensure charge neutrality of the overall system. The periodic potential is then expressed for all  $s > -1$  as

$$\tilde{v}_{s,L}(r) = \lim_{q \rightarrow \infty} \left( \sum_{|k| \leq q} v_s(r + kL) - \rho \int_{\cup_{|k| \leq q} \ell_{L,k}} v_s(r - r') dr' \right). \quad (3.10)$$

It turns out rather beautifully that this normalization, which we emphasize to be very natural from the viewpoint of physics, exactly corresponds to the meromorphic extension to the half complex plane  $\{\Re(s) > -1\}$  of the periodic Riesz potential in the short-range case  $s > 1$  with a pole at  $s = 1$  [BBT85;

[Bor+88; BBS89; BBS14; Lew22a]. Therefore,  $\tilde{v}_{s,L}$  rewrites as in (3.9) if one agrees to use the meromorphic continuation of the Hurwitz zeta function to the punctured complex plane  $\mathbb{C} \setminus \{1\}$  on the right-hand side. We note that, although this entails that the periodic potential  $\tilde{v}_{s,L}$  can actually be continued over the entire complex plane — at the exception of  $s = 1$  — one should be aware that the above formula (3.10) is *a priori* only valid when  $\Re(s) > -1$ . Pushing  $s$  below this threshold usually requires another kind of normalization. We refer to [Lew22a, Section IV. A] for further details on this question and more generally on the analytic continuation of the periodic Riesz potential in arbitrary dimension  $d \geq 1$ .

### §3.2.2. The 1D Coulomb Gas

In this section, we review important results regarding the *1D Coulomb gas*, also known as the *1D Jellium* or *1D One-Component Plasma* (1dOCP). This remarkable model, which corresponds to the choice  $s = -1$ , was extensively studied by Kunz in [Kun74], where the thermodynamic limit of the free energy (3.3) and the correlations functions  $\rho^{(k)}(r_1, \dots, r_k)$  — see (3.4) — are computed through transfer matrix techniques [Bax16]. It is to be noted that the Coulomb case is very special, as the force between two distinct particles does not depend on their mutual distance, so that ordering the particles on the line somehow leads to a form of “conditional independence”. This fortuitous property was leveraged by Aizenman and Martin in [AM80] where similar results as of [Kun74] are proved using the electric field as the key variable and appealing to ergodic arguments to conclude. Directly extending these methods to other values of  $s$  seems complicated, if not impossible. Altogether, the authors of [Kun74; AM80] managed to prove that the correlation functions were proper periodic functions at all temperature, and thus that the Coulomb gas is crystallized at all temperature. We shall now briefly explain the strategy of both papers, and verify the results numerically.

The strategy of Kunz in [Kun74] essentially boils down to the celebrated *transfer-matrix method* in statistical physics [Bax16]. The strategy is used by Kunz both with free and periodic boundary conditions. It should be noted that the argument heavily relies on both the one-dimensional nature of the system and on the very peculiar form of the Coulomb potential in dimension  $d = 1$ , namely  $-|r|$ . Indeed, the Jellium energy — that is, when the particles interact with each other as well as with the uniform compensating background, see (3.11) below — is a quadratic function once restricted to the set of ordered configuration. Indeed, if we suppose that  $-N/2 \leq r_1 \leq \dots \leq r_N \leq N/2$ , the Jellium energy in the non-periodic setting rewrites as

$$\begin{aligned} - \sum_{1 \leq i < j \leq N} |r_i - r_j| + \sum_{i=1}^N \int_{-\frac{N}{2}}^{\frac{N}{2}} |r_i - r| dr - \frac{1}{2} \int_{-\frac{N}{2}}^{\frac{N}{2}} \int_{-\frac{N}{2}}^{\frac{N}{2}} |r - r'| dr dr' \\ = \sum_{i=1}^N \left( r_i - i + \frac{N+1}{2} \right)^2 + \frac{N}{2}. \end{aligned} \quad (3.11)$$

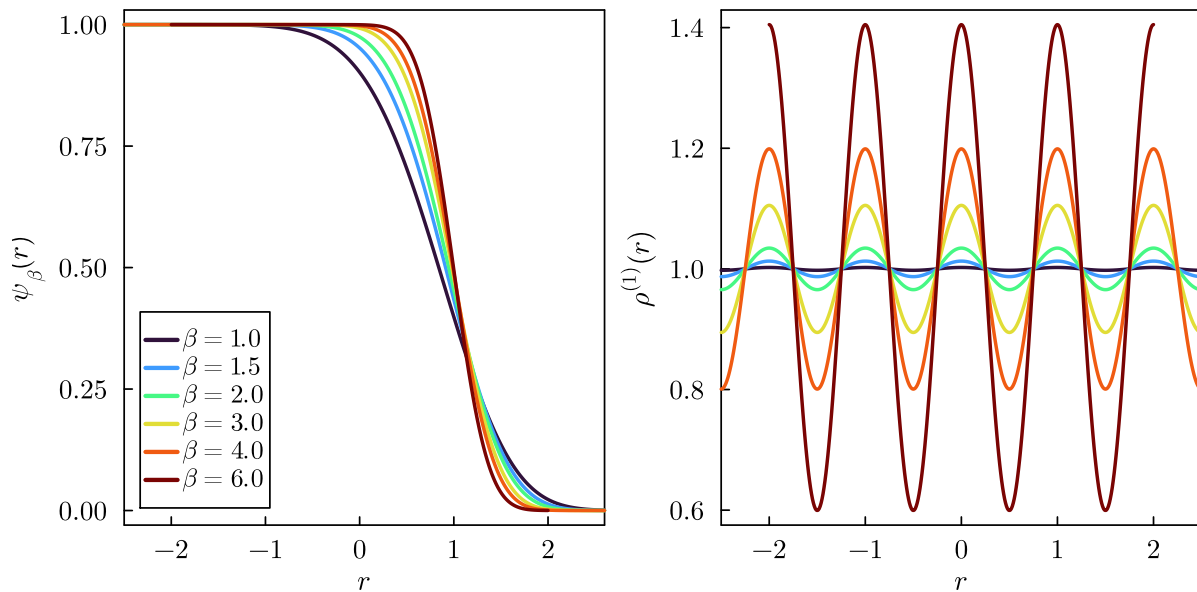


Figure 3.3: We compute the Perron–Frobenius eigenvector  $\psi_\beta$  of the operator  $K$  defined in (3.13) by a straightforward discretization for several temperatures. We then compute the density  $\rho^{(1)}(r)$  using Equation (3.14). We observe that the density is a proper periodic function of period  $\rho = 1$  as proved by Kunz. We also retrieve the properties that  $\psi_\beta$  converges to 1 as  $r \rightarrow -\infty$  and to 0 as  $r \rightarrow \infty$ , and that  $\psi_\beta$  converges to the Heaviside function centered at  $r = 1$  in the vanishing temperature limit, see [Kun74, Appendix & p. 315]. All figures in this work were made using Julia and the `Plots.jl` package.

In particular, the canonical Gibbs measure  $Q_N$  associated to the above energy is a Gaussian once restricted to the set of ordered configurations. Using this property, Kunz was able to rewrite the free energy  $f_N(\beta)$  in the finite length  $N$  and at inverse temperature  $\beta$  as

$$f_N(\beta) = \left\langle g_\beta, K^N g_\beta \right\rangle_{L^2(\mathbb{R}_+)}. \quad (3.12)$$

Here,  $K$  is a compact operator with positive kernel over the Hilbert space  $L^2(\mathbb{R}_+)$  which serves as an infinite-dimensional analogue to the so-called *transfer matrix* and  $g_\beta$  is an explicit function in  $L^2(\mathbb{R}_+)$ . We emphasize that  $K$  depends on the inverse temperature  $\beta$  but does *not* depend on the number of particles  $N$ . It is given by the operator

$$Kf(r) := \int_{r-1}^{\infty} e^{-\beta u^2} f(u) du, \quad (3.13)$$

which is an integrable operator with kernel  $K(r, r') = e^{-\beta r'^2} \mathbf{1}(r' \geq r - 1)$ . Appealing to the *Perron–Frobenius theorem* [Kun74, Lem. 1 in Appendix] it follows from positivity and compactness of  $K$  that it has a simple largest eigenvalue  $\lambda(\beta)$  associated to a unique positive normalized eigenfunction  $\psi_\beta \in L^2(\mathbb{R}_+)$ . By discretizing the operator  $K$  as defined in (3.13) above, we can



compute numerically the eigenvector  $\psi_\beta$ , see Figure 3.3. The thermodynamic limit of the free energy can then be readily expressed using those quantities [Kun74, Eq. (17)]. The correlation functions can be dealt with in a very similar manner. For instance, Kunz found that the one-point correlation function  $\rho^{(1)}(r)$  converges to the periodic function given by

$$\rho_\tau^{(1)}(r) = \sum_{k \in \mathbb{Z}} \psi_\beta(-r - k - \tau) \psi_\beta(r + k + \tau) \quad (3.14)$$

for some  $\tau \in \mathbb{R}$  [Kun74, Eq. (40-41)]. In fact, the scalar  $\tau$  depends on the sequence of the number of particles  $N$ 's considered in the thermodynamic limit, which is very clear manifestation of the breaking of symmetry. Furthermore, Kunz managed to prove that all the correlation functions were periodic. Nevertheless, to ensure that crystallization really happens, one still needs to prove that those functions are properly periodic, that is that they are not constant function. By appealing to analyticity [Kun74, at p. 314], he managed to prove this fact at low enough temperature  $\beta \gg 1$ . This was eventually generalized to all temperature  $\beta > 0$  by Aizenman and Martin [AM80]. In Figure 3.3, we show  $\rho_\tau^{(1)}(r)$  computed for several temperatures. Finally, Kunz studied the problem with periodic boundary conditions, in which case he found that the correlation functions  $\rho_{\text{per}}^{(k)}$  were all obtained by averaging their counterparts  $\rho_\tau^{(k)}$  in the non-periodic setting over their period, that is

$$\rho_{\text{per}}^{(k)}(r_1, \dots, r_k) = \int_0^1 \rho_\tau^{(k)}(r_1, \dots, r_k) d\tau. \quad (3.15)$$

It follows from (3.15) that crystallization cannot be detected on  $\rho^{(1)}(r)$  anymore in the periodic setting, and that one should look at the pair correlation  $g(r)$ , which will then be a proper periodic function of the distance  $r$ . This is a clear manifestation of the breaking of symmetry. In what will follow regarding the Riesz gas with general exponent  $-1 < s < 0$ , we will look at the pair correlation for the same reasons.

Aizenman and Martin [AM80], building on previous works of Lenard [Len04b; Len04a] and Edwards–Lenard [EL04], took a different road than that of Kunz to study the 1D Coulomb gas. Their key idea is to work with the *electric field*  $E(r)$  rather than the particles themselves. Indeed, there is a one-to-one correspondence between the set of configurations and the set of possible electric fields, as proved in [AM80, Lem. 4]. This can be seen from the fact that, given a configuration of particles  $X = (r_1, \dots, r_N)$ , the electric field  $E_X(r)$  generated by this configuration has a very simple structure, as it is a piecewise linear function of unit slope with a jump of unit size located at each particle  $r_j$ , see Figure 3.4. This allows to view the electric field as a random jump process whose semigroup can be readily expressed [AM80, Eq. (49) sqq.]. The most important thing to stress out here is the *Markovian* nature of this process, as can be intuited from Figure 3.4. This allows yet once more to appeal to Perron–Frobenius to cope with the thermodynamic limit, see [AM80, Eq. (4.11)]. Finally, using an ergodic theorem, Aizenman and Martin proved the periodicity of the correlations functions for all  $\beta > 0$ , thus extending the result

of [Kun74] mentioned earlier. They were able to show that the associated limiting point process obtained in the thermodynamic limit  $N \rightarrow \infty$  can be defined by the usual set of characterisations such as *Dobrushin–Lanford–Ruelle* (DLR), *Bogoliubov–Born–Green–Kirkwood–Yvon* (BBGKY) and *Kubo–Martin–Schwinger* (KMS) equations — we refer the reader to [Lew22a; Der19] on this matter.

We note that the electric field is a very convenient variable in the case of the Coulomb potential because the energy (3.11) can be expressed as a positive quadratic form in the variable  $E_X$  using the *carré du champ* operation [AM80, Eq. (2.7) and (2.9)]. For arbitrary exponent  $s$ , this very useful *carré du champ* is no longer available *as-is*. Nevertheless, it is still possible — although much more involved — to work with the electric field rather than the particles themselves. We refer to the long line of work initiated by Serfaty and collaborators — see [Ser15] for a self-contained reference, or the references in [Lew22a].

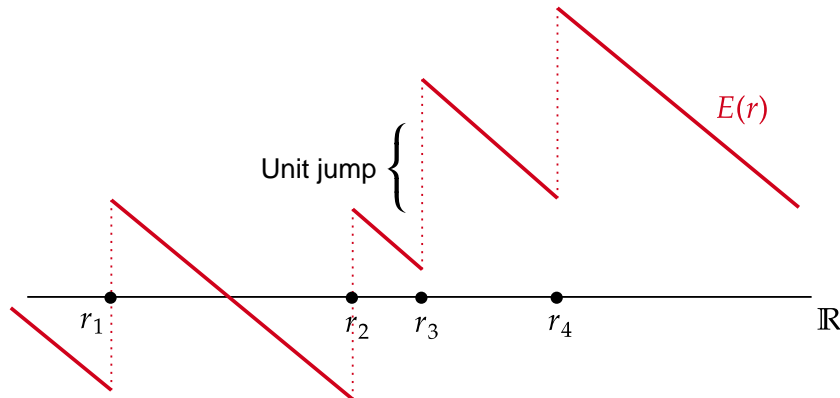


Figure 3.4: The electric field generated by a configuration of particles  $r_1, \dots, r_N$  has a very nice structure in the case of the Coulomb potential. It is a piecewise linear function with unit slope and jump of unit size at each particle  $r_j$ . The position of the particles can therefore be retrieved from the positions of the jumps.

### §3.2.3. The Dyson log-gas

The *Dyson log-gas* [Dys62a; Dys62b; Dys62c; DM63], which corresponds to the choice  $s = 0$  and for which we recover the logarithmic interaction  $v_0(r) = -\ln(r)$ , is also accessible to analytical computations for specific values of the inverse temperature  $\beta$ , using the tenacious analogy between log-gases and random matrix models [For10]. We recall that, in the context of random matrices, the Dyson log-gas is called the  $\beta$ -ensemble. Using a fairly general theory of one-dimensional quantum fluids of Haldane [Hal81], it was conjectured by Forrester



in [For84] that the leading term in the expansion of the pair correlation  $g(r)$  at large  $r$  would be given by

$$g(r) \underset{r \rightarrow \infty}{\sim} \begin{cases} 1 - \frac{1}{\pi^2 \beta r^2} & \text{for } \beta < 2, \\ 1 + \frac{\cos(2\pi r)}{2\pi^2 r^2} - \frac{1}{2\pi^2 r^2} & \text{for } \beta = 2, \\ 1 + c \frac{\cos 2\pi r}{r^{4/\beta}} - \frac{1}{2\pi^2 r^2} & \text{for } \beta > 2 \end{cases} \quad (3.16)$$

for some universal constant  $c > 0$ . The expansion is rigorous for  $\beta = 1, 2$  and 4 as can be shown using the analogy between Dyson log-gas and standard Gaussian ensembles [For10]. It is also veracious in the case of even or rational  $\beta$ 's as proved in [For93] and [For10, Chap. 13]. We see that the decay of  $g(r)$  in the large  $r$  limit exhibits a transition at  $\beta = 2$  from an universal monotonous power-law decay  $r^{-2}$  to an oscillating and non-universal decay whose power depends on the temperature. This is a celebrated example of a *Berezinski–Kosterlitz–Thouless* (BKT) transition [KT73], see also [Her07; Mud14]. In the vanishing temperature limit  $\beta \rightarrow \infty$ , as the oscillations become predominant,  $g(r)$  converges to a periodic function and the system is crystallized onto a (floating) *Wigner crystal* [SS15; Leb15].

It follows from the expansion (3.16) that the behaviour of the structure factor  $S(k)$  (3.5) in the small wavenumber limit  $k \rightarrow 0$  is to be given by that of the  $-1/r^2$  term to the leading order. Indeed, although the leading term of the (truncated) pair correlation is of order  $1/r^{4/\beta}$  as soon as  $\beta > 2$ , the cosine term shifts its contribution to the Fourier transform at  $k \sim 1$ . More generally, all the oscillating terms  $\cos(2\pi nr)/r^{4n/\beta}$  in the expansion of the pair correlation of the log-gas when  $\beta > 2$  — which we did not write in (3.16), see [For84] — only contribute to the structure factor at  $k \sim n$ . Altogether, the term  $-1/r^2$  is the only one which contributes to the behaviour of  $S(k)$  near  $k = 0$ , so that

$$S(k) \sim 2\beta^{-1}|k| \quad \text{as } k \rightarrow 0. \quad (3.17)$$

It also follows from the expansion (3.16) that  $S(k)$  should feature a singularity at  $k = 1$  as soon as  $\beta \geq 4$ . This singularity will be logarithmic at the threshold  $\beta = 4$  and should diverge as an inverse power-law when  $\beta > 4$ , that is (up to multiplicative constant)

$$S(k) \sim \frac{1}{|1 - k|^{1-4\beta^{-1}}} \quad \text{as } k \rightarrow 1^-. \quad (3.18)$$

In any case, we emphasize that this singularity is of an integrable type. This is in clear contrast with what one would expect in a crystal. Indeed, as explained earlier, in the case of a crystal the structure factor  $S(k)$  should have a sharp peak at  $k = 1$  corresponding to that of a Dirac mass, as expected from the periodic nature of the pair correlation  $g(r)$ .

Although our main goal in this paper is to investigate the long-range situation  $-1 < s < 0$ , we show that the previous claims on the behaviour of  $S(k)$  near  $k = 0$  (3.17) and near  $k = 1$  (3.18) are confirmed numerically. Although we

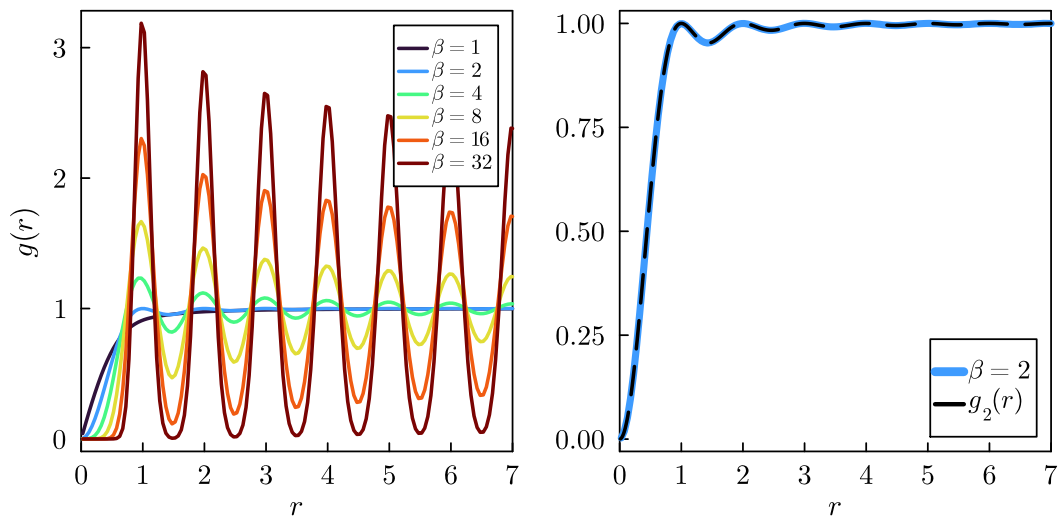


Figure 3.5: On the left, we display the pair correlation  $g(r)$  for the Dyson log-gas at several temperatures obtained by our algorithm. Below the critical temperature  $\beta = 2$ , the pair correlation converges monotonically to the average density — here set to  $\rho = 1$  — whereas above the critical temperature  $g(r)$  we observe oscillations which eventually vanish as  $g(r)$  converges to  $\rho$  in the large  $r$  limit. On the right, the approximation of  $g(r)$  at  $\beta = 2$  obtained numerically is seen to be consistent with the exact formula for the pair correlation [For10]. We used  $N = 100$  particles and built the pair correlation by binning.

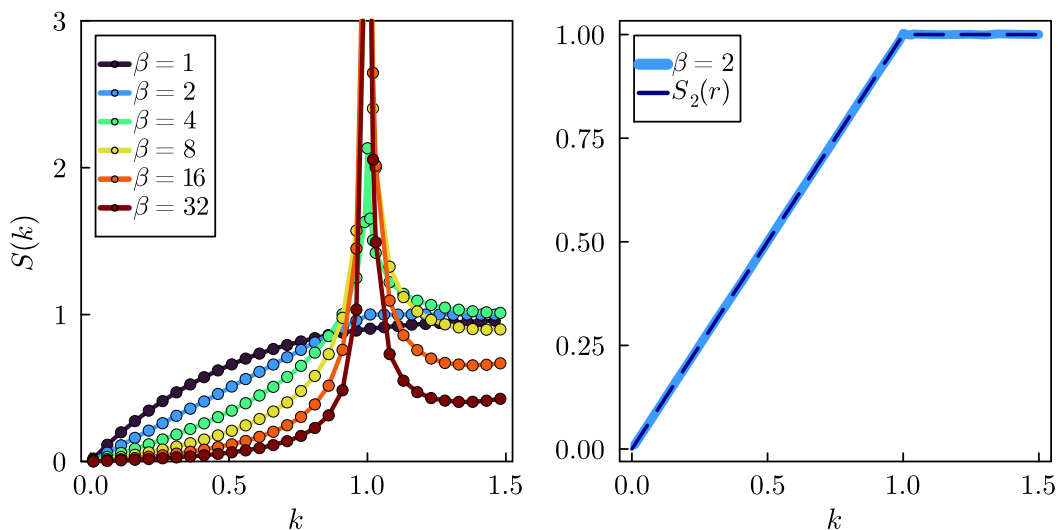


Figure 3.6: On the left, we display the structure factor  $S(k)$  for the Dyson log-gas at several temperatures, using the pair correlations previously computed and (3.5). Above the critical temperature  $\beta = 2$ , the structure factor displays a peak at  $k = 1$  as expected from the (3.17). On the right, the approximation of  $S(k)$  at  $\beta = 2$  obtained numerically is seen to be consistent with the exact formula for the pair correlation [For10]. We thinned the number of displayed wavenumbers for visual convenience.

do so as a sanity-check for our algorithm, which is presented in Appendix A<sup>2</sup>, these results may be of independent interest for some readers. In Figure 3.5, we display an approximation of the pair correlation  $g(r)$  obtained numerically for several inverse temperatures  $\beta$ . We observe that below the critical temperature  $\beta = 2$ , the correlation converges monotonically to the average density  $\rho = 1$ . From the critical temperature  $\beta = 2$  further on, the pair correlation  $g(r)$  displays damped oscillations whose amplitude strengthen as the temperature is further decreased, and eventually  $g(r)$  converges to the average density in the large  $r$  limit. Our approximation fits perfectly with the exact formula for  $g(r)$  at  $\beta = 2$  [For10]. In Figure 3.6, we display the associated structure factor  $S(k)$ , which is obtained by computing the (discrete) Fourier transform of the pair correlation  $g(r)$  as in (3.5). We then regress  $S(k)$  near  $k = 0$  to obtain the universal behaviour of the slope at which  $S(k)$  approaches  $k = 0$ , and we regress at  $k = 1$  to obtain the exponent at which  $S(k)$  diverges. The results, displayed in Figure 3.7, are in clear adequation with (3.17) and (3.18) — and therefore are numerical confirmations of the veracity of the expansion (3.16) conjectured by Forrester.

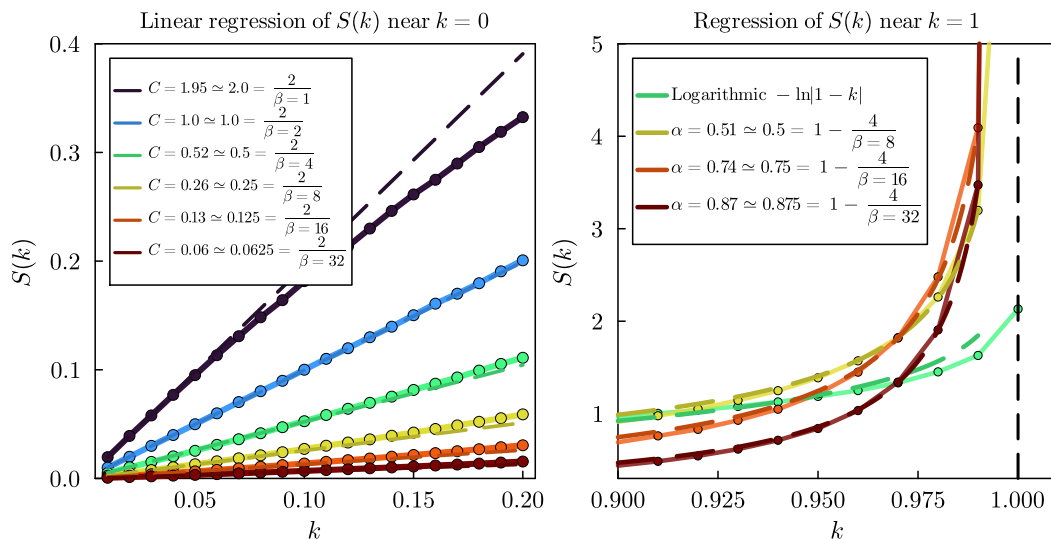


Figure 3.7: On the left, we regress the structure factor  $S(k)$  of the Dyson log-gas near  $k = 0$  on  $C|k|$ . The slope coefficient  $C$  is found to be close to  $2\beta^{-1}$ , which is consistent with (3.17) — and therefore consistent with the Forrester’s expansion (3.16). On the right, we regress the structure factor  $S(k)$  near  $k = 1$  as  $k \rightarrow 1^+$  on  $c|1 - k|^\alpha$  for some here unimportant constant  $c$ . We found  $\alpha$  to be closed to  $1 - 4\beta$  for  $\beta > 4$ , which is consistent with (3.17). When  $\beta = 4$ , the structure factor behaves logarithmically  $S(k) \sim \log|1 - k|$ , as expected.

<sup>2</sup>Our code is available at <https://github.com/rodrigue/PTRiesz>.

### §3.3. Evidence for the existence of a phase transition

In this section, we investigate numerically the 1D Riesz gases for general exponent  $-1 < s < 0$ . In § 3.3.1, we show evidence which strongly advocates for the existence of a phase transition with respect to the temperature  $T$  depending on  $s$ . In § 3.3.2, we further study the behaviour of the pair correlation  $g(r)$  and most importantly of the structure factor  $S(k)$  to make our claim that there coexists two separate phase transitions, namely a fluid–quasisolid transition of a BKT-type similar to that of the Dyson log-gas, and a freezing point below which the system is crystallized. A phase diagram is then determined numerically according to a set of criteria summarized in Table 3.1.

#### §3.3.1. Long-range order at low enough temperature and existence of a critical temperature

We compute the pair correlation  $g(r)$  at several temperatures for various exponents  $-1 < s < 0$ . As a first evidence for the (non-)existence of some long-range order, we wonder whether or not  $g(r)$  features persistent oscillations at low enough temperature, or equivalently if the structure factor  $S(k)$  has a sharp peak at  $k = 1$ . From our experiments, we see that for all  $s$  in the range  $-1 \leq s \leq 0$ , at high enough temperature  $g(r)$  rapidly and monotonically converges to the average density as  $r \rightarrow \infty$ , as one would expect in a fluid phase, whereas for small enough temperatures it exhibits long-lasting oscillations which are ever more amplified as the temperature is further lowered, consistent with the fact that the system is crystallized at zero temperature [BHS19]. This is clearly seen in Figure 3.8, where we display  $g(r)$  and  $S(k)$  for the 1D Riesz gas for  $s = -0.5$  at varying temperature. Furthermore, we found this conspicuous qualitative change of behaviour to occur within a range of temperatures depending on the exponent  $s$ . In Figure 3.9, we fix two temperatures and we vary the exponent  $s$ . We observe that the oscillations appear sooner, that is at higher temperature, as the exponent  $s$  gets closer to  $s = -1$ , and conversely that they appear later as  $s$  gets closer to  $s = 0$ . This is consistent with the fact that the Coulomb gas is crystallized at all temperature and with the fact that the Dyson log-gas is expected to be a fluid at all positive temperature.

From what precedes, we are brought to believe in the existence of a critical temperature  $\mathfrak{T}_s$  depending on the exponent  $-1 < s < 0$  which separates between a fluid phase in the high temperature regime  $T > \mathfrak{T}_s$  and an ordered-phase in the low temperature regime  $T < \mathfrak{T}_s$ . The critical temperature should interpolate between the Coulomb gas, that is  $\mathfrak{T}_s \rightarrow \infty$  as  $s \rightarrow -1$ , and the Dyson log-gas, that is  $\mathfrak{T}_s \rightarrow 0$  as  $s \rightarrow 0$ . Nevertheless, a clear determination of  $\mathfrak{T}_s$  is evidently complicated, pertaining to both the underlying limitations of numerics and the absence of an absolute criterion to either rule in favor or out of the appearance of a long-range order. Furthermore, it is unclear whether or not the oscillations which appear in the pair correlation  $g(r)$  eventually vanish in the large  $r$  limit,

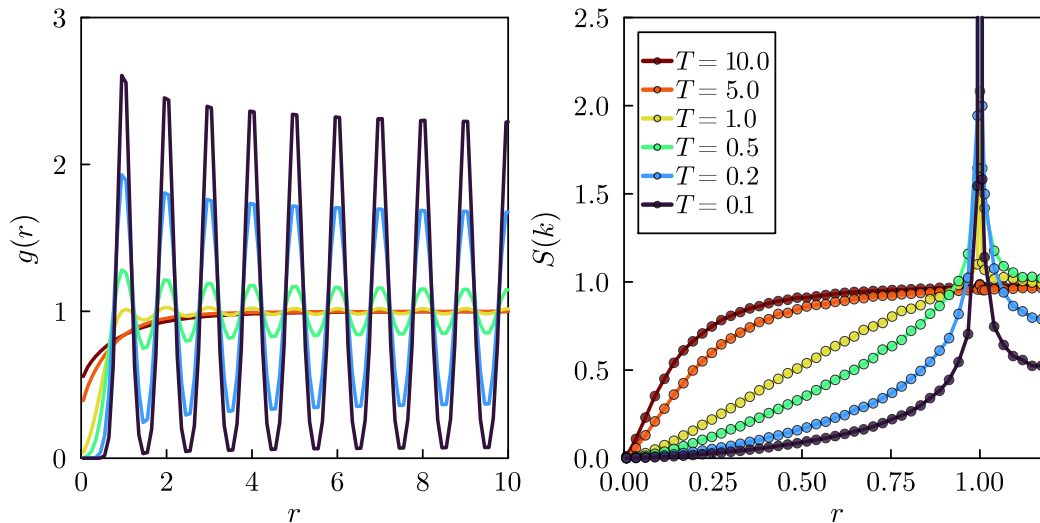


Figure 3.8: On the left (resp. right) we display the pair correlation  $g(r)$  (resp. the structure factor  $S(k)$ ) at various temperatures for the Riesz gas of exponent  $s = -0.5$ . At high enough temperature,  $g(r)$  converges rapidly and monotonically to the average density, whereas in the low temperature regime it features persistent oscillations whose amplitude increases as the temperature is furthered lowered. In this regime, we observe that the structure factor  $S(k)$  has a sharp peak at  $k = 1$  whose width (resp. height) decreases (resp. increases) as the temperature is lowered, hinting the presence of a Dirac mass accounting for the periodicity of  $g(r)$  in the large  $r$  limit. We used  $N = 150$  particles.

as in the Berezinski–Kosterlitz–Thouless paradigm. The rest of our paper is dedicated to obtain a better understanding of  $\mathfrak{T}_s$ .

### §3.3.2. Determination of the critical temperature and nature of the transition

It remains to determine the nature of the transition which was put in evidence in the previous section, as well as the behaviour of the transition curve with respect to  $s$ , which was loosely denoted  $\mathfrak{T}_s$  above. In this section, we give several criteria to determine whether the system is in a fluid, quasisolid or solid phase. In fact, we make the claim that there actually coexists two distinct set of critical temperatures, denoted hereafter  $\tilde{T}_s$  and  $T_s$ . The first one separates between a fluid and a quasisolid phase reminiscent of the BKT transition and similar to that of the Dyson log-gas discussed earlier, while the second one corresponds to the point at which the system is frozen onto a true solid. We use these criteria to give a — at least schematic — phase diagram of the Riesz gas with respect to the effective temperature  $T$  and the exponent  $-1 < s < 0$ .

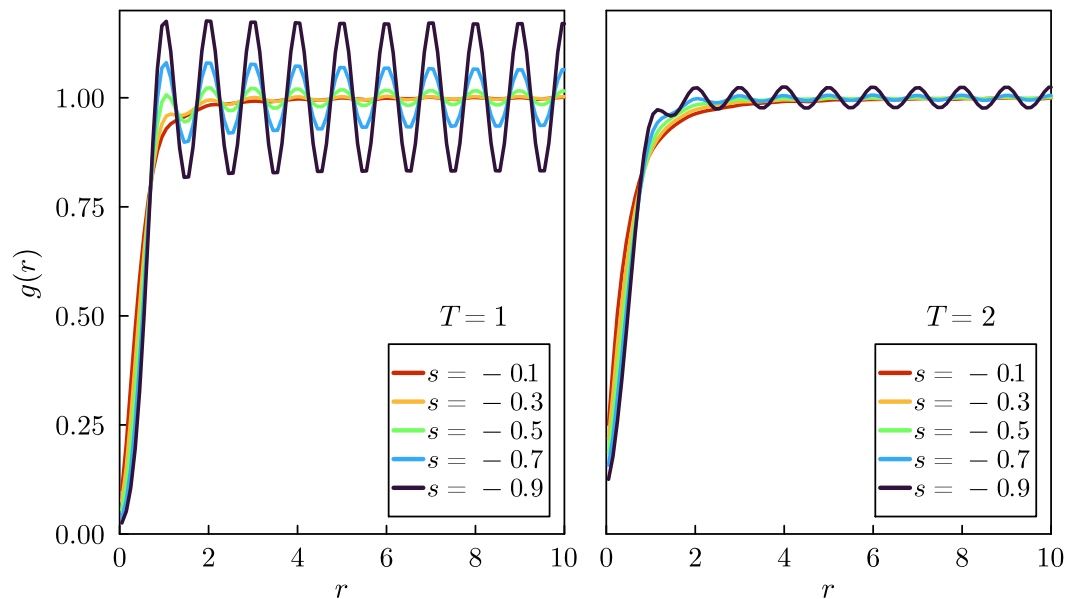


Figure 3.9: We fix two temperatures  $T = 1$  (left) and  $T = 2$  (resp. right) and we vary the exponent  $s$ . We observe that the oscillations of  $g(r)$  appear at lower temperatures as the exponent  $s$  gets closer to the Coulomb gas  $s = -1$ , which is consistent with the fact that the Coulomb gas is crystallized at all temperature. On the other hand, the oscillations appear at much lower temperature as  $s$  gets closer to the Dyson log-gas  $s = 0$ , which is consistent with the fact that the Dyson log-gas is expected to be a fluid at all positive temperature. From this, we are led to claim that the critical temperature  $\mathfrak{T}_s$  depends on the exponent  $s$  in such a way as to interpolate between the phase diagrams of the 1D Coulomb gas and the Dyson log-gas, as conjectured in [Lew22a].

### Behavior of $S(k)$ in the limit $k \rightarrow 0$

In the preceding section, we were attentive to whether or not the pair correlation  $g(r)$  converges to a periodic function in the large  $r$  limit, as this is a clear manifestation of the crystallization. This is evidently related to the appearance in the Fourier space of a peak at  $k = 1$ , or for that matters at any multiple of the period of  $g(r)$ . Nevertheless, it turns out that the breaking of symmetry can be seen on the behaviour of the structure factor in the limit  $k \rightarrow 0$ . We stress out that this is a non-trivial fact. Indeed, Aizemann, Goldstein and Lebowitz gave in [AGL01] a sufficient condition for translational symmetry to be broken in one-dimensional systems. This result, which is related to the notion of *hyperuniformity* [TS03; Tor18], essentially says that if the structure factor  $S(k)$  behaves like  $|k|^\eta$  in the small wavenumber vicinity  $k \rightarrow 0$  for some  $\eta > 1$ , then translational symmetry must be broken in the thermodynamic limit. We note that this is *not* in contradiction with the expected phase diagram of the Dyson log-gas, for which  $\eta = 1$  as shown previously.

On the other hand, it follows from the extension of a heuristical argument of Forrester [For10] — see also [AM85] — that, if the Riesz gas at exponent

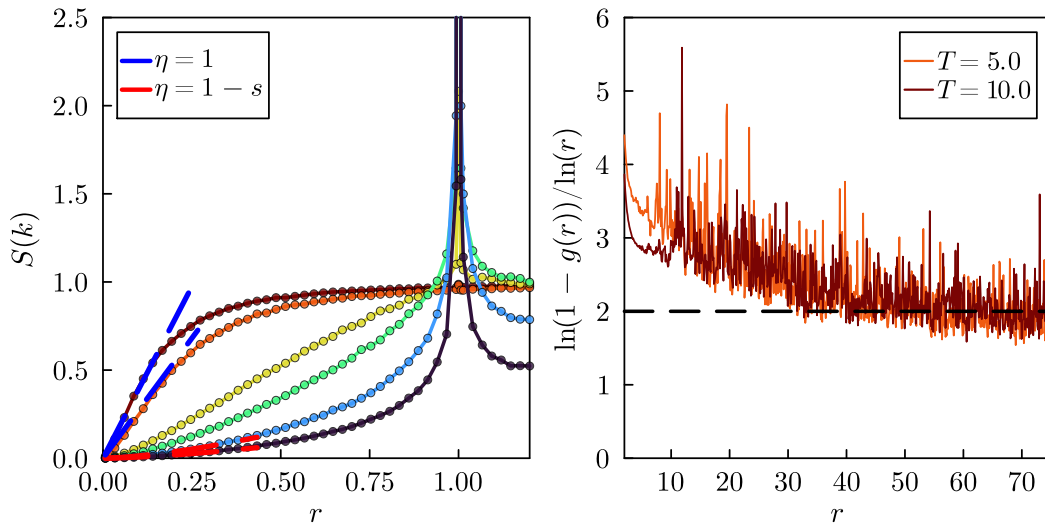


Figure 3.10: On the left, we regress the structure factor  $S(k)$  near  $k = 0$  on  $C|k|^\eta$ , for the same values as of Figure 3.8. We allow  $\eta \in \{1, 1 - s\}$ , where  $\eta = 1 - s$  is the natural exponent for the crystallisation regime, whereas  $\eta = 1$  corresponds to that of a liquid. In the high temperature regime, we see that  $S(k)$  behaves linearly as  $k \rightarrow 0$ . This is coherent with the behaviour of the pair correlation at large  $r$ , which decays as  $-1/r^2$ , as seen on the second figure on the right, where we display  $\ln(1 - g(r)) / \ln(r)$ , which is seen to approach the value 2 as  $r$  gets large. At low enough temperature the choice  $\eta = 1 - s$  is found to yield a smaller regression residue, which is a strong evidence that the Riesz gas is crystallized according to Aizemann *et al.* [AGL01]

$s$  is crystallized, then  $S(k)$  must behave like  $S(k) \simeq C|k|^{1-s}$  in the small wavenumber limit  $|k| \rightarrow 0$  for some constant  $C > 0$ . The exponent  $\eta = 1 - s$  is very natural, as it fits with the Dyson log-gas, for which  $\eta = 1$ , and the Coulomb gas for which  $\eta = 2$ . It also fits with the results of obtained by Boursier [Bou22] in the case where  $0 < s < 1$ . The argument of Forrester, which can be found in [For10, Chap. 11] in the case of the log-gas, can be extended as follows for any  $s$ . If we perturb our system at equilibrium by a fluctuating charge density  $\varepsilon e^{-ikr}$ , and if we denote by  $\rho_\varepsilon(r)$  the density of the perturbed system, then it must be that

$$\rho_\varepsilon(r) - \rho(r) \underset{k \rightarrow 0}{\sim} -\varepsilon e^{ikr} \quad (3.19)$$

where  $\rho(r)$  is the density of the original system. We emphasize that this equivalence is only formal and *a priori* not rigorous. It says that the system responds in an appropriate manner to the perturbation, that is in such a way as to cancel the perturbation and remain in equilibrium. In fact, this can be viewed as characteristic of a crystalline order. Indeed the crystal should be able to remain stable under perturbation of large enough wavelength  $\lambda \gg a$  — or small enough wavenumber  $k \ll 1/a$ , as in (3.19) — where  $a$  is the crystal constant.

Now, by letting  $\varepsilon \rightarrow 0$  in (3.19), and using the well-known relations which links the functional derivatives of the free energy and the correlation functions



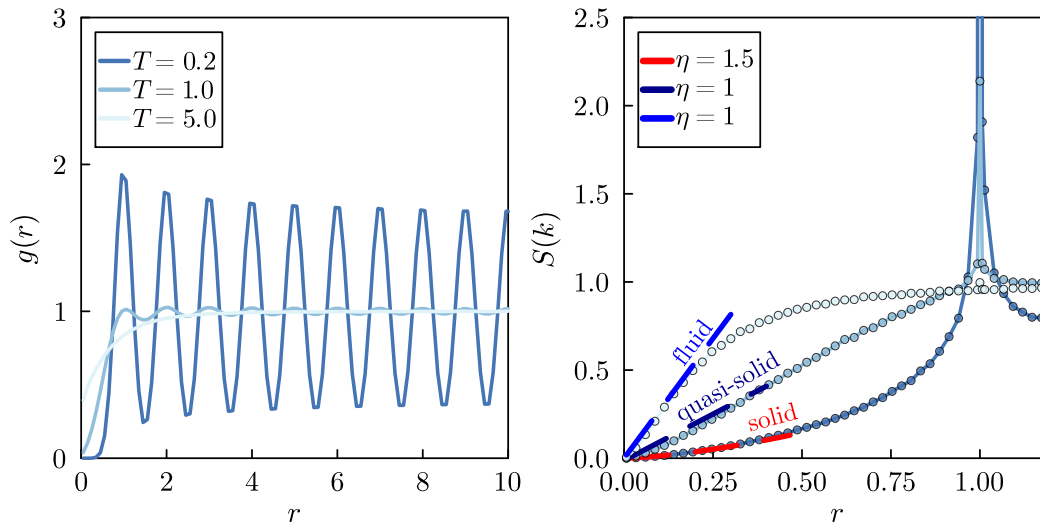


Figure 3.11: From a closer investigation on the structure factor (right), we are led to believe in the existence of three distinct regimes for  $s = -0.5$ . At high temperature, the structure factor behaves linearly near the origin and it has no peak at  $k = 1$ , hinting a fluid phase. As the temperature is decreased, a peak appears at  $k = 1$ , but the structure factor remains linear in the limit  $k \rightarrow 0$ . This is characteristic of a quasisolid. Finally, at low enough temperature,  $S(k)$  flattens out near the origin and behaves as  $|k|^{1-s}$ : it is a crystal.

[HM13], the left-hand side of (3.19) can be written as

$$\rho_\varepsilon(r) - \rho(r) \sim -\varepsilon\beta \int_{\mathbb{R}} W_\varepsilon(r') \rho^{(2)}(r, r') \quad (3.20)$$

where  $W(r) := -|r|^{-s} * e^{ikr}$  is the potential associated to the charge density which perturbs the system, and  $\rho^{(2)}(r, r')$  is the two-point correlation as defined earlier (3.4). By using the invariance by translation, we get

$$\rho_\varepsilon(r) - \rho(r) \sim -\varepsilon\beta \int_{\mathbb{R}} W_\varepsilon(r') \rho^{(2)}(r, r') \sim \varepsilon\beta \frac{1}{|k|^{1+s}} S(k). \quad (3.21)$$

Putting (3.19) and (3.21) yields that  $\eta = 1 - s$ . Furthermore, the coefficient  $C$  should be linear in the temperature, and in fact it should be given by  $C = 2\beta^{-1}$  similarly to the Dyson log-gas, as seen in (3.17). In Figure 3.10, we observe that at high enough temperature, the structure factor  $S(k)$  converges linearly to 0 as  $k \rightarrow \infty$ . This is seen to be consistent with the truncated pair correlation  $g(r) - 1$  decaying as  $-1/r^2$  in the large  $r$  limit. On the contrary, as the temperature is decreased, the structure factor  $S(k)$  flattens near the origin, and at low enough temperature it is seen to decrease sublinearly to 0 as  $k \rightarrow 0$ , in fact as  $|k|^{1-s}$  as expected from the above heuristic.

### Behaviour of $S(k)$ in the limit $k \rightarrow 1$

In the case of a crystal, the structure factor  $S(k)$  should have a sharp peak at  $k = 1$  corresponding to a Dirac mass. We recall that in the case of the Dyson



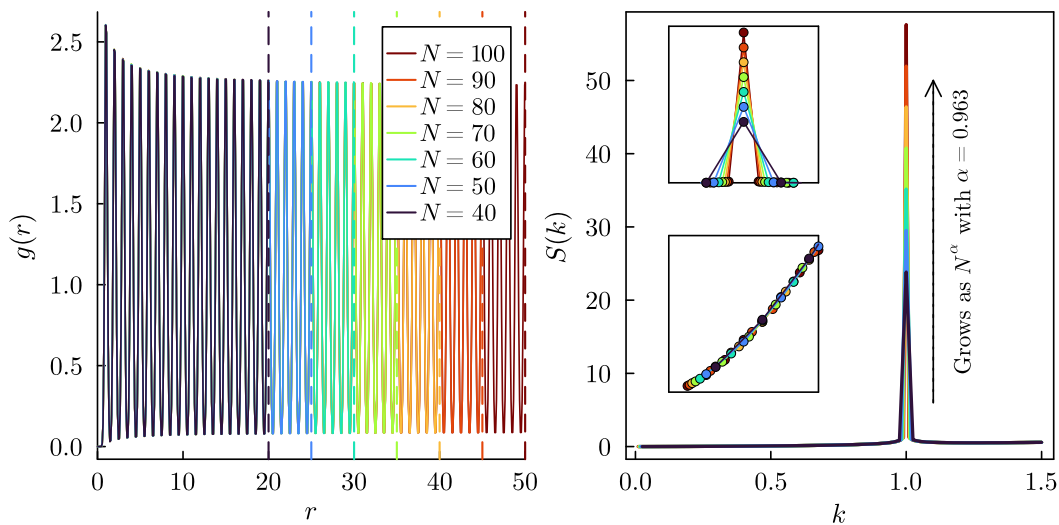


Figure 3.12: We set  $s = -0.5$  and  $T = 0.1$ , and we vary the number of particles  $N$ . We then regress the successive height of the peaks  $S(1)$  on  $N^\alpha$ . Here, we found  $\alpha = 0.963$ , which is therefore rather close to that of a Dirac mass.

log-gas, the structure factor also has a peak at  $k = 1$  for  $\beta > 4$ , but it is an integrable function and not a Dirac mass — and, in the case  $2 < \beta < 4$ , the function  $S(k)$  is non-monotonous near  $k = 1$ . In Figure 3.10, we see that the appearance of the peak at  $k = 1$  precedes that of the flattening of the structure factor near the origin, and that there exists a range of temperatures for which the peak exists but the structure factor seems to behave linearly near the origin. This is clearly seen for  $s = -0.5$  in Figure 3.11.

We are therefore brought to believe in the existence of two distinct phase transitions. At high enough temperature, the Riesz gas is a fluid. As the temperature is decreased down to a certain threshold, there is a BKT transition similar to that of the Dyson log-gas as discussed earlier, corresponding to the formation of a quasisolid. Eventually, as the temperature is furthered decreased, there is another threshold at which the system is frozen into a true crystal. This is clearly depicted in Figure 3.11.

To determine whether or not the peak at  $k = 1$  is of an integrable type or a Dirac mass, we may look at its behaviour as one varies the number of particles  $N$ . Indeed, in the case of a Dirac mass, the height of the peak  $S(1)$  should grow as  $N$ . On the other hand, if the structure factor diverges as  $|1 - k|^{-\alpha}$  as  $k \rightarrow 0$  for some  $\alpha < 1$ , as in a quasisolid, then the height of the peak should grow as  $N^\alpha$ . An example is given in Figure 3.12 and Figure 3.13, in which we fix  $s = -0.5$  and two different temperatures, namely  $T = 0.1$  and  $T = 0.6$ . We then vary the number of particles and determine how the height of the peak of the structure factor grows with  $N$ . When  $T = 0.6$ , we find that the peak grows as  $N^\alpha$  for  $\alpha = 0.655$ , which seems to indicate a quasisolid phase. When  $T = 0.1$ , we find  $\alpha = 0.963$ , which is closer to indicating a Dirac mass and therefore a solid phase.

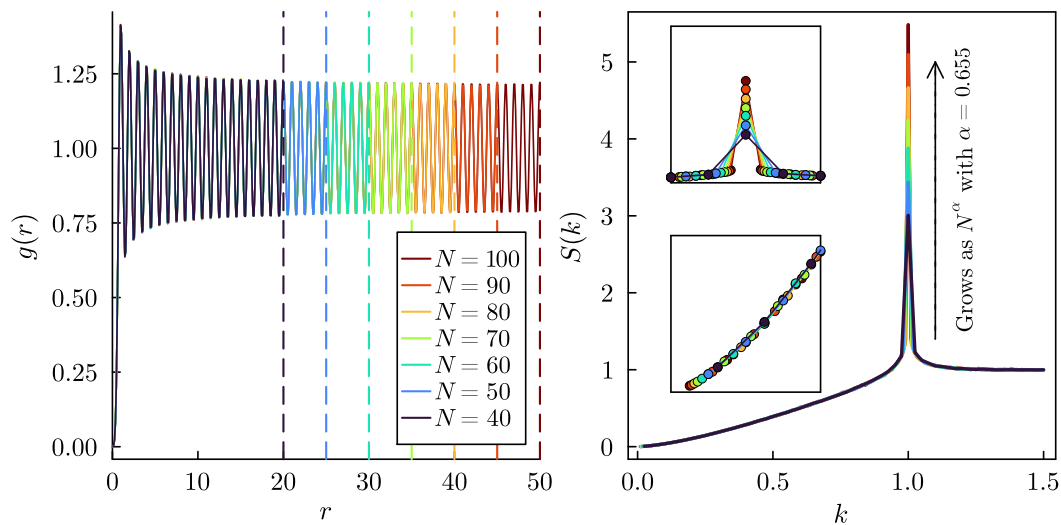


Figure 3.13: Same as in Figure 3.12, but for  $T = 0.6$ . For this temperature, we have  $\alpha = 0.655$ , which would say that at this temperature, the Riesz gas is a quasisolid. We emphasize that it is not evident that the oscillations of the pair correlation  $g(r)$  eventually vanish in the large  $r$  limit, as expected for a quasisolid. This is a well-known problem in the literature, which makes the BKT-type transitions very delicate to detect numerically.

### §3.3.3. Phase diagram with respect to the temperature

Using the different criteria as summarized in Table 3.1, we may draw a schematic phase diagram of the 1D Riesz gas with respect to the effective temperature  $T$  and the exponent  $-1 \leq s \leq 0$ . The diagram is depicted in Figure 3.14.

We should emphasize that a precise determination of the transition curves  $\tilde{T}_s$  and  $T_s$  is evidently complicated. The transition curve  $\tilde{T}_s$ , corresponding to what we believe to be a BKT transition separating between a fluid and a quasisolid phase, can be determined as the threshold temperature at which  $S(k)$  start having a peak at  $k = 1$  — thus becoming non-monotonous near  $k = 1$ . We see in Figure 3.14 that the behaviour of  $\tilde{T}_s$  is consistent with the Dyson log-gas for which the BKT transition occurs at  $T = 1/2$ .

As for the transition curve  $T_s$ , corresponding to the fluid-solid transition, its values are somewhat harder to determine. According to our criterion, it corresponds to the temperature at which  $S(k)$  flattens at the origin and behaves as  $|k|^{1-s}$  and at which  $S(k)$  as a Dirac mass at  $k = 1$ . Although a precise determination of this threshold is evidently complicated in the finite length  $N < \infty$ , we are confident that the phase diagram depicted in Figure 3.14 is qualitatively sound.

## §3.4. Conclusion

We provided numerical evidence for the existence of two distinct phase transitions with respect to the temperature in 1D Riesz gases. The first transition

Table 3.1: Summary of criteria

	$S(k)$ near $k = 0$	$S(k)$ near $k = 1$	$g(r)$ far away
Solid	<b>If</b> $S(k) \sim C k ^\eta$ for $\eta > 1$ and some constant $C > 0$ , then translational-symmetry is broken according to Aizenman <i>et al.</i> [AGL01]. Conversely, <b>if</b> the symmetry is broken then according to a heuristic of Forrester [For10], see also [AM85], it must be that $\eta = 1 - s$ .	$S(k)$ has a Dirac at $k = 1$ , accounting for the periodicity of $g(r)$ . Numerically, the peak should grow as $N$ or equivalently its width should decrease as $N^{-1}$	$g(r) \not\rightarrow \rho = 1$ as $r \rightarrow \infty$ , and periodic in the large $r$ limit. This is reminiscent of the Coulomb gas with periodic boundary conditions [Kun74].
Quasi-solid	$S(k)$ should be linear in the limit $k \rightarrow 0$ . This is reminiscent of the Dyson log-gas, see (3.17).	$S(k)$ should be non-monotonic near $k = 1$ . It can be singular but must remain integrable, that is $S(k) \sim \frac{1}{ k-1 ^\alpha}$ for $\alpha < 1$ . Numerically, the peak should grow as $N^\alpha$ . This is reminiscent of the Dyson log-gas, see (3.18).	$g(r) \rightarrow \rho = 1$ as $r \rightarrow \infty$ but has oscillations which slowly vanish in the large $r$ limit. The power of leading order term should depend on the temperature.
Fluid	$S(k)$ behaves linearly in the limit $k \rightarrow 0$ .	$S(k)$ is monotonic near $k = 1$	$g(r) \sim 1 - 1/r^2$ in the large $r$ limit.

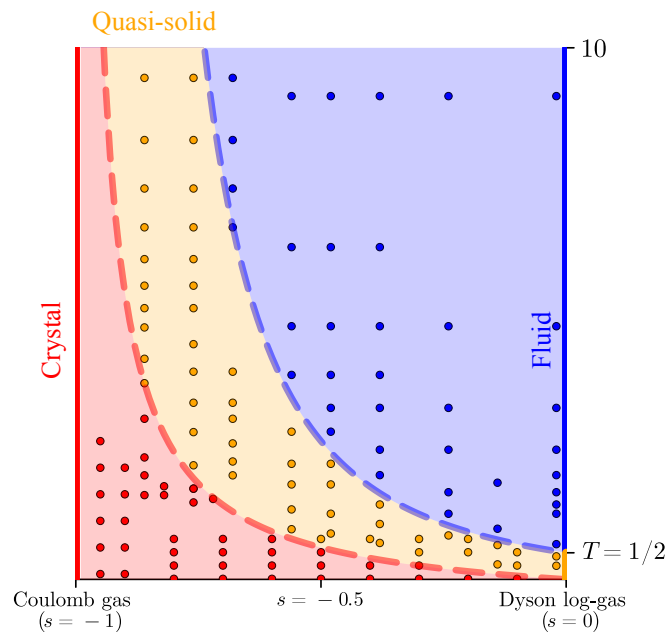


Figure 3.14: Phase diagram of the one-dimensional Riesz gas with respect to the effective temperature  $T$  and the exponent  $-1 \leq s \leq 0$ . The blue dots correspond to the couple  $(s, T)$  for which we found the system to be a fluid phase. The orange dots correspond to that of the quasi-solid phase, for which the structure factor  $S(k)$  has a (integrable) peak at  $k = 1$  and  $S(k)$  behaves linearly in the limit  $k \rightarrow 0$ . Finally, the red dots correspond to the solid phase, for which  $S(k)$  as a Dirac mass at  $k = 1$  and  $S(k)$  behaves as  $|k|^{1-s}$ . We then draw schematically the transition curve  $\tilde{T}_s$ , corresponding to the BKT transition, and the transition curve  $T_s$ , corresponding to the solid-phase. Those curves are seen to be consistent with the phase diagrams of the Coulomb gas and the Dyson log-gas, for which  $s = -1$  and  $s = 0$  respectively.

corresponds to a BKT transition similar to that of the Dyson log-gas. The second one corresponds to the critical temperature below which the system is crystallized. The transition curves interpolate between the known — or at least expected — phase diagrams of the 1D Coulomb gas and the Dyson log-gas. Although a precise quantitative determination on the critical temperatures is delicate, using a set of different criteria we were able to draw a schematic phase diagram of the 1D Riesz gases with respect to the temperature  $T$  and the exponent  $s$ .

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N°725528).

## §A. Implementation details

All the numerics were performed using the Julia programming language. Our implementation is available online<sup>3</sup>. All the plots can be reproduced. If one is interested in generating and plotting the pair correlation  $g(r)$  and the structure factor  $S(k)$  for the Riesz gas with homogeneity parameter  $s$  at inverse temperature  $\beta$ , obtained by generating  $n$  samples for  $N$  particles, it suffices to use to launch the following command in a terminal window

```
julia [-t] PTRiesz.jl -N N -b  $\beta$  -s s -n n.
```

If the option `-t` is provided, the computation will be parallelized using the provided (and available) number of threads on the machine. The option `-T` can also be used to provide the temperature instead of the inverse temperature. To compute  $g(r)$  and  $S(k)$  for several temperatures, say from  $\beta_1$  to  $\beta_2$  with a step of size given by  $\delta\beta$ , it suffices to launch the following command

```
julia [-t] PTRiesz.jl -N N -brange  $\beta_1$   $\delta\beta$   $\beta_2$  -s s -n n.
```

This will produce all the plots as well as an animated version. The options `-Nrange` and `-srange`, used in a similar manner, are also available to vary the number of particles and/or the homogeneity parameter  $s$ . The options `-xlist` are also available for  $x = b, T, N, s$  to provide the parameters one by one.

### §A.1. Some general words

We benefit heavily in our numerics of the one-dimensional nature of our system, which makes things much simpler relative to the higher dimensions  $d \geq 2$ . Indeed, in dimensions  $d = 2, 3$  one probably needs to resort to much finer and rather complicated implementation. Although well-known implementations to compute the quantities such as  $g(r)$  and its Fourier transform have been around for a long time, such as LAMMPS<sup>4</sup>, which is very well maintained *Molecular Dynamics* code implemented in C++, we are not aware of packages which are tailored for the dimension one. Moreover, to the best of our knowledge, these implementations are usually tailored for Lennard-Jones-type potentials, and are not made to work with system interacting through long-range potential as the Riesz potential in the case  $s < d$ . In particular, the pair correlation is usually computed in these codes by using some nearest-neighbour convention, which hugely impact positively the computation burden. Nevertheless, in the long-range case, using this convention would be absurd.

<sup>3</sup>See <https://github.com/rodrigue1/1DRieszGases.git>

<sup>4</sup>See <https://lammps.org/>

## §A.2. Sampling the Riesz gases

The canonical ensemble  $Q_N$  can be sampled using rather straightforward *Markov Chain Monte Carlo* (MCMC) methods [RC04; RC10]. We remember that a crucial element to ensure the good quality of MCMC sampling is the careful selection of the size  $\sigma$  of the proposal jump. To swerve away from this delicate issue, which is more often than not a critical component which can greatly deteriorate the sampling quality if overlooked, one can resort to *adaptive* methods which select  $\sigma$  on the fly to ensure that the *rate of acceptance* is close to a fixed value [Vih20]. A very nice implementation of these methods is available in the package `AdaptiveMCMC.jl`<sup>5</sup>. Nevertheless, those adaptive methods suffers a problem in the context of phase transitions. Indeed, as one seeks for the existence of a phase transition, one must vary the temperature within a certain range of values. This is in contrast with the main use of MCMC methods in the context of optimization, where one is usually interested in the low temperature regime  $\beta \gg 1$ , which features some challenges for the practitioner [Rob+18]. Indeed, as the temperature is lowered, the canonical ensemble concentrates around the minimizers of the energy. At some threshold temperature, it becomes almost infeasible to further explore the energy landscape properly for the chain will be trapped in local minima for very long periods of time. On the other hand, as the temperature is increased, the entropy eventually dominates over the energy, and the canonical ensemble will typically be closer — in a very informal sense — to a uniform distribution. This naturally causes the chain to often accept the proposal states, as the difference of energies is evermore flattened. An adaptive method, which aims at a specific acceptance rate, will therefore increase the proposal jump size  $\sigma$  to decrease the acceptance, although this is in contradiction with the physics. We found this to greatly deteriorate the quality of the sampling, especially near what we believe to be the critical temperature.

We therefore used a bare *Random Walk Metropolis* (RWM), where the size  $\sigma$  of the proposal jump is determined phenomenologically beforehand. We found the choice  $\sigma^* \simeq c\sqrt{\beta/N}$  for some fixed tuning constant  $c > 0$  to be appropriate both in the low and high temperature regimes for all  $s$  in the range  $(-1, 0)$ . In fact, this should not come as a surprise, since the canonical ensemble associated to the Coulomb gas  $s = -1$  is essentially — up to reordering of the particles — an isotropic multivariate Gaussian of deviation  $\sigma^*$  [Kun74]. We note that, in the singular case  $s \geq 0$  one could certainly benefit resorting to fancier MCMC methods, typically gradient-informed ones [CFS21; LRS10]. Our algorithm is then parallelized and launched on a cluster infrastructure to draw many samples.

We used some well-known rough diagnostic rule of thumbs to assess the mixing of the chain, *e.g.* monitoring of the energy and the acceptance rate, and sensitivity with respect to the initial configuration. In fact, we benefit from the fact that the minimizer of the energy at zero temperature is known for all  $s$  in a periodic setting [BHS19]. Therefore, we start our chain on this minimizer, so

<sup>5</sup>See <https://github.com/mvihola/AdaptiveMCMC.jl>

that no warm-up period is needed. We also appealed to some finer diagnostic tools such as monitoring the *effective size sample* and its related statistical quantities [Roy20].

### §A.3. Sampling the pair correlation and the structure factor

The pair correlation  $g(r)$  is obtained by binning. That is, for each sample generated by the chain, the mutual distances between the particles in the configurations are computed, and those distances are binned into an histogram which is eventually normalized. The number of bins should evidently depend of the number of samples used and the accuracy needed. As a general rule, we chose to consider ten bins *per* unit length. We actually found this to be usually consistent with the *Freedman–Diaconis rule* [FD81]. To the best of our knowledge, binning the pair correlation seems to be the usual method in computational physics, at the exception of the work [PR17]. Once the pair correlation has been properly binned into an histogram, we compute the structure factor  $S(k)$  by considering the (discrete) Fourier transform of this histogram, which is then only defined on the values  $\mathbb{Z}/N$ . Once again, this seems to be the usual procedure in the literature to compute  $S(k)$  [PR17; Haw+23].

### §A.4. Implementing the periodic Riesz potential

We explained earlier that the periodic Riesz potential  $\tilde{v}_{s,L}$  can be expressed for all  $s > -1$  through the Hurwitz zeta function  $\zeta(s, x)$ , seen as a generalisation of the Riemann zeta function  $\zeta(s)$  and extended to the entire complex plane  $\mathbb{C}$  by meromorphic continuation with a pole at  $s = 1$ . Nowadays, special functions are easily available in scientific programming languages. In `Julia`, the function `zeta(s::Complex, x::Real)` is implemented in the `SpecialFunctions.jl` package. Nevertheless, those functions are usually more expensive to compute than regular functions. For instance, a straightforward benchmarking yields that

```
@btime zeta(-0.5, 1)
» 395.109 ns (0 allocations : 0 bytes).
```

In comparison, the computation of the (co)sine function or other usual functions is done in around  $1\text{ ns}$ , which represents a huge difference. To alleviate the computational burden, we *tabulate* and interpolate the periodic potential  $\tilde{v}_{s,L}$  on the circle of length  $N$ . This represents a huge speed-up. For instance, for the exact potential, implemented as `periodic_riesz(s, N, r)`, where we consider that  $L = N$ , we find

```
@btime periodic_riesz(-0.5, 5, 1)
» 3.463 μs (0 allocations: 0 bytes).
```

The interpolated version of the potential can be built by closure `itp_periodic_riesz = build_periodic_riesz(s, N, grid_size)`. Here, the `grid_size` argu-

ment is the number of discretization points over which to interpolate. It must evidently be increased with  $N$ . The default value considers 10000 points *per* unit length. We find

```
itp_periodic_riesz = build_periodic_riesz(-0.5, 5)
@btime itp_periodic_riesz(1)
» 57.689 ns (1 allocation: 16 bytes)
```

which represents a huge speed-up.



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## RÉSUMÉ

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Nous étudions les gaz de Coulomb et Riesz au travers du prisme du transport optimal multimarge. Dans une première partie, nous montrons que le potentiel de Kantorovich s'écrit comme un potentiel électrostatique générée par une distribution de charge dont nous étudions les propriétés. Cela nous permet d'introduire une nouvelle discrétisation pour résoudre numériquement le transport optimal multimarge, basée sur sa formulation duale. Dans une deuxième partie, nous étudions le comportement asymptotique du potentiel de Kantorovich dans le cadre des coûts de transport répulsifs. Finalement, dans une troisième et dernière partie, nous présentons des résultats numériques qui prédisent l'existence de transitions de phases pour les gaz de Riesz à très longue portée en une dimension d'espace.

## MOTS CLÉS

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Transport optimal multimarge, théorie du potentiel, gaz de Coulomb et Riesz, mécanique statistique, transition de phases, analyse numérique

## ABSTRACT

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In this thesis, we study the Coulomb and Riesz gases from the perspective of the multimarginal optimal transport. In the first chapter, we show that the Kantorovich potential can be written as an electrostatic potential generated by a charge distribution. We study the properties of this charge, and use it to introduce a novel discretization to solve the multimarginal optimal transport problem, based on its dual formulation. In the second chapter, we study the asymptotic behaviour of the Kantorovich potential in the context of repulsive costs. Finally, in the third and last chapter of this thesis, we provide numerical evidence for the existence of phase transitions in the one-dimensional Riesz gases with long-range potential.

## KEYWORDS

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Multimarginal optimal transport, potential theory, Coulomb and Riesz gases, statistical mechanics, phase transitions, numerical analysis