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Existence and Uniqueness of Monge Minimizers for a Multi-Marginal Optimal Transport Problem with Intermolecular Interactions Cost

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Dedicated to Giuseppe Buttazzo on the occasion of his 70th birthday.

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We investigate a new multi-marginal optimal transport problem arising from a dissociation model in the Strong Interaction Limit of Density Functional Theory. In this short note, we introduce such dissociation model, the corresponding optimal transport problem as well as show preliminary results on the existence and uniqueness of Monge solutions assuming absolute continuity of at least two of the marginals. Finally, we show that such marginal regularity conditions are necessary for the existence of an unique Monge solution.

Keywords: Multi-marginal optimal transport, density functional theory, dissociation energy.

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1. Introduction

In this work, we introduce the following variational problem, motivated by Density Functional Theory:

$$\inf_{\gamma\in\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)}\int_{\mathbb{R}^{d(N_\alpha+N_\beta)}}\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}x_i\cdot y_j^* d\gamma(x_1,\dots,x_{N_\alpha},y_1,\dots,y_{N_\beta}),\qquad(1)$$

where $y_j^*=(-2y_j^1,y_j^2,\dots,y_j^d)$ for each $j\in\{1,\dots,N_\beta\}$, $N_\alpha,N_\beta\in\mathbb{N}$ are integers, ρ_α and ρ_β are probability measures, and $\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)$ denotes the set of transport plans $\gamma=\gamma_\alpha\otimes\gamma_\beta$ such that γ_α has N_α marginals equal to ρ_α and γ_β has N_β marginals equal to ρ_β.

The multi-marginal optimal transport problem (1) thus has an attractive harmonic cost in the direction $x_1\in\mathbb{R}$ and repulsive harmonic cost in all other space directions

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$(x_2, \dots, x_d) \in \mathbb{R}^{d-1}$. The problem with fully attractive or repulsive harmonic cost have been considered, e.g. in [11, 14, 15, 17].

The analysis of (1) naturally appears in the study of the dissociation energy in a Density Functional Theory framework (DFT) [13, 24]. The dissociation energy is the energy required to break apart a chemical bond and translate the constituent atoms or molecules to an infinitely large distance from each other.

Our mathematical analysis focuses on the asymptotics of the problem and minimizers, in the limit when the ground-state energy of a cluster of interacting molecules $\alpha\beta$ becomes infinitely separated into individual clusters α and β. In our model, the probabilities ρ_α and ρ_β in (1) are the single-particle densities of the isolated systems α and β. Such dissociation limit is of significant importance in the study of molecular properties and chemical reactions, such as energy transfer, bond strength, stability of compounds, atomic and molecular spectroscopy.

Multi-marginal Optimal Transport theory: In the multi-marginal optimal transport, we correlate a finite number of probability measures to minimize some notion of overall cost. Formally, for given Borel probability measures $\rho_1, \dots, \rho_N$ on open sets $X_i \subseteq \mathbb{R}^d$, $i = 1, \dots, N$ (respectively), and c a given cost function on the product space $X := \prod_{i=1}^N X_i$, one seeks to minimize the *total cost*

$$\int_X c(x_1, \dots, x_N)\, d\gamma, \tag{KP}$$

among all Borel probability measures γ on X whose marginals are the ρ_i. We say that ρ_i is the ith marginal of γ if, for any Borel set $A \subseteq X_i$, we have

$$\gamma(X_1 \times \dots \times X_{i-1} \times A \times X_{i+1} \times \dots \times X_N) = \rho_i(A).$$

This formulation is known as the Kantorovich Problem (KP) and it is, in fact, a relaxation of a restricted problem: the Monge Problem (MP).

In the Monge formulation, one seeks to minimize

$$\int_{X_1} c(x_1, T_2 x_1, \dots, T_N x_1)\, d\rho_1, \tag{MP}$$

among all $(N-1)$-tuples of maps $(T_2, \dots, T_N)$ such that $(T_i)_\sharp \rho_1 = \rho_i$ for all numbers $i = 2, \dots, N$, where $(T_i)_\sharp \rho_1$ denotes the *image measure* of ρ_1 through T_i, defined by $(T_i)_\sharp \rho_1(A) = \rho_1(T_i^{-1}(A))$, for any Borel set $A \subseteq X_i$.

When $N = 2$, (KP) and (MP) reduce respectively to the Kantorovich and Monge formulations of the classical optimal transport problem. This case is well understood; in particular, if the map $x_2 \mapsto D_{x_1} c(x_1, x_2)$ is injective for each fixed x_1 and ρ_1 is absolutely continuous with respect to the d-dimensional Lebesgue measure $\mathcal{L}^d$, there exists a unique solution to (KP) and it is induced by a measurable map [4, 28, 34, 36].

Under the same regularity condition on the first marginal ρ_1, Kim and Pass [22] extended this result to the multi-marginal case covering a wide class of cost functions [6, 14, 19, 23, 29, 30], namely the cost function satisfies the so-called *twist condition on c-splitting sets* [7, 12], i.e. the mapping

$$(x_2, \dots, x_N) \mapsto D_{x_1} c(x_1^0, x_2, \dots, x_N)$$

is injective on the subset of S where $D_{x_1}c(x_1^0, x_2, \dots, x_N)$ exists, for each fixed element $x_1^0 \in X_1$ and c-splitting set $S \subseteq \{x_1^0\} \times X_2 \times \dots X_N$. See Definition 3.2.

Although being fairly general, the *twist condition on c-splitting sets* does not hold for many costs (e.g., cyclic Euler cost [4]), or is very difficulty to be verified (e.g., Coulomb cost), see [31–33] for details and further examples. When $N_\alpha > 1$, the cost in (1) is not twisted on splitting sets and an alternative approach must be used. In that case, Pass&Vargaz-Jiménez [33] show that extra regularity conditions on some of the marginals (in addition to a regularity condition on ρ_1) are sufficient to guarantee uniqueness of Monge solution.

Main result and proof strategy: In this paper, we show that, under some regularity conditions on some of the marginals, the multi-marginal optimal transport problem (1) admits a unique Monge solution, see Theorem 3.6. The general case $N_\alpha > 1$ requires the use of a more general approach developed by Pass and Vargas-Jiménez [32], which generalizes the twist on c-splitting sets condition. Roughly speaking, Pass&Vargaz-Jiménez condition requires the mapping

$$(x_2, \dots, x_N) \mapsto D_{x_1}c(x_1, x_2, \dots, x_N)$$

to be injective on special subsets generated by c-splitting functions, see Definition 3.4 for details. In particular, this guarantees that every solution to (KP) is concentrated on a graph of a measurable map, and therefore, by simply using a standard argument uniqueness of Monge solution for (1) is obtained. Finally, Lemma 3.7 shows that such regularity conditions on the marginals are necessary for the uniqueness of Monge solutions.

Organization of the paper: In section 2, we define the dissociation energy (as defined in (10)) of many-electron quantum systems as well as the corresponding energy in the so-called Strong Interaction limit of Density Functional Theory. In section 2.1, we introduce the mathematical framework and compute the asymptotic development of the multi-marginal optimal transport problem with Coulomb cost in the dissociation limit. Finally, in section 3 we prove preliminary results on the existence and uniqueness of Monge solutions for the problem (1).

2. Dissociation energy of many-electrons quantum systems

We consider a quantum mechanical system of $N = N_\alpha + N_\beta$ non-relativistic electrons (of mass m_e and charge $-e$), representing clusters of molecules α and β respectively, interacting with each other, moving around classical nuclei with positions $\mathbf{R}_1, \dots, \mathbf{R}_M \in \mathbb{R}^d$ and charges $Z_1 e, \dots, Z_M e$ (Born-Oppenheimer approximation).

The electrons are described by a wave function $\Psi : (\mathbb{R}^d \times \mathbb{Z}_2)^N \to \mathbb{C}$ of N positions $z \in \mathbb{R}^d$ and spin coordinates $s_i \in \{\uparrow, \downarrow\} = \mathbb{Z}_2$ that are antisymmetric with respect to permutations of the electron coordinates,

$$\Psi(z_{\sigma(1)}, s_{\sigma(1)}, \dots, z_{\sigma(N)}, s_{\sigma(N)}) = \mathrm{sign}(\sigma)\Psi(z_1, s_1, \dots, z_N, s_N), \quad \sigma \in \mathfrak{S}_N, \tag{2}$$

where $\mathfrak{S}_N$ denotes the group of permutations of the indices $1, ..., N$. The set of square-integrable N-electron wave functions, $\{\Psi \in L^2((\mathbb{R}^d \times \mathbb{Z}_2)^N; \mathbb{C}) : (2) \text{ holds } \}$, will be denoted $\bigwedge_{i=1}^N L^2(\mathbb{R}^d \times \mathbb{Z}_2; \mathbb{C})$.

The total energy $E^{\alpha\beta}[\Psi, v]$ of the fermonic state of the coupled system $\alpha\beta$ with external potential $v : \mathbb{R}^d \to \mathbb{R}$ is given, in atomic units, by

$$E^{\alpha\beta}[\Psi, v] = T[\Psi] + \mathrm{V}_{ee}[\Psi] + V_{\mathrm{ne}}[\Psi, v], \tag{3}$$

where $T[\Psi]$ is the *kinetic energy*

$$T[\Psi] = \frac{1}{2} \sum_{s_1 \in \mathbb{Z}_2} \int_{\mathbb{R}^3} \cdots \sum_{s_{N_\alpha} \in \mathbb{Z}_2} \int_{\mathbb{R}^3} \sum_{i=1}^{N} |\nabla \Psi(z_1, s_1 \ldots, z_N, s_N)|^2 dz_1 \ldots dz_N,$$

$\mathrm{V}_{ee}[\Psi]$ is the *Coulomb electronic-electronic interaction energy*

$$\mathrm{V}_{ee}[\Psi] = \sum_{s_1 \in \mathbb{Z}_2} \int_{\mathbb{R}^d} \cdots \sum_{s_N \in \mathbb{Z}_2} \int_{\mathbb{R}^d} \sum_{1 \le i < j \le N} \frac{1}{|z_i - z_j|} |\Psi(z_1, s_1 \ldots, z_N, s_N)|^2 dz_1 \ldots dz_N,$$

and $V_{\mathrm{ne}}[\Psi, v]$ is the *electron-nuclei interaction energy*

$$V_{\mathrm{ne}}[\Psi, v] = \sum_{s_1 \in \mathbb{Z}_2} \int_{\mathbb{R}^d} \cdots \sum_{s_N \in \mathbb{Z}_2} \int_{\mathbb{R}^d} \sum_{i=1}^{N} v(z_i) |\Psi(z_1, s_1 \ldots, z_N, s_N)|^2 dz_1 \ldots dz_N.$$

Typically, v is the Coulomb potential generated by M nuclei which are at positions R_ν and have charges Z_ν,

$$v(z) = -\sum_{\nu=1}^{M} \frac{Z_\nu}{|z - R_\nu|}. \tag{4}$$

The *ground state energy* is defined by

$$E_0[v] = \min \left\{ E^{\alpha\beta}[\Psi, v] \,:\, \Psi \in L^2((\mathbb{R}^d \times \mathbb{Z}_2)^N; \mathbb{C}), \|\Psi\|_2 = 1 \text{ and (2) holds} \right\}. \tag{5}$$

Similarly, we define the total energies E^α, E^β of the isolated clusters of molecules α and β

$$E^\alpha[\Psi^\alpha, v^\alpha] = T^\alpha[\Psi^\alpha] + \mathrm{V}_{ee}[\Psi^\alpha] + V_{\mathrm{ne}}[\Psi^\alpha, v^\alpha], \tag{6}$$

and

$$E^\beta[\Psi^\beta, v^\beta] = T^\beta[\Psi^\beta] + \mathrm{V}_{ee}[\Psi^\beta] + V_{\mathrm{ne}}[\Psi^\beta, v^\beta], \tag{7}$$

as well as their corresponding ground-state energies E_0^α and E_0^β

$$E_0^\alpha[v^\alpha] = \min \left\{ E^\alpha[\Psi^\alpha, v^\alpha] : \Psi \in L^2((\mathbb{R}^d \times \mathbb{Z}_2)^{N_\alpha}; \mathbb{C}), \|\Psi^\alpha\|_2 = 1 \text{ and (2) holds} \right\}, \tag{8}$$

and

$$E_0^\beta[v^\beta] = \min \left\{ E^\beta[\Psi^\beta, v^\beta] : \Psi \in L^2((\mathbb{R}^d \times \mathbb{Z}_2)^{N_\beta}; \mathbb{C}), \|\Psi^\beta\|_2 = 1 \text{ and (2) holds} \right\}. \tag{9}$$

We are interested in the dissociation energy of the system $\alpha\beta$, i.e. the energy difference between the fully coupled system $\alpha\beta$ and the sum of the energies of the isolated systems α and β:

$$E_{\mathrm{diss}} = E_0 - (E_0^\alpha + E_0^\beta). \tag{10}$$

In the following, we will focus on the so-called *Strong-Interaction limit of Density Functional Theory*, which was introduced in [24].

The strong interaction limit of Density Functional Theory (DFT)

By integrating the N-point probability distribution over the spins, we obtain the N-point position density,

$$\pi_N^\Psi(z_1,\dots,z_N) := \sum_{s_1,\dots,s_N\in\mathbb{Z}_2} |\Psi(z_1,s_1,\dots,z_N,s_N)|^2, \quad \Psi\in\mathcal{W}^N. \tag{11}$$

The single particle density $\rho_\Psi(z_j)$ is then obtained by integrating out all but one electron position $z_j\in\mathbb{R}^d$,

$$\rho_\Psi(z_j) := N\int_{\mathbb{R}^{d(N-1)}} \pi_N^\Psi(z_1,z_2,\dots,z_j,\dots,z_N)\prod_{i\neq j} dz_i, \quad \forall\, j\in\{1,...,N\}. \tag{12}$$

We denote by $\Psi\mapsto\rho$ the relation between Ψ and ρ given by equations (11), (12). This means that the wave function Ψ has single-electron density ρ.

Following the work of Hohenberg and Kohn [20], Levy [25] and Lieb [27] showed that the electronic ground state problem (5) can be recast as a minimization over single-electron densities ρ instead of many-electron wavefunctions Ψ:

$$E_0[v_{\rm ne}^{\alpha\beta}] = \inf_{\rho\in\mathcal{D}^N}\left\{F_{\rm LL}^{\alpha\beta}[\rho] + N\int_{\mathbb{R}^d} v_{\rm ne}^{\alpha\beta}(r)\rho(r)dr\right\}, \tag{13}$$

with
$$F_{\rm LL}^{\alpha\beta}[\rho] = \min\left\{T[\Psi] + \mathrm{V}_{ee}[\Psi] : \Psi\in\mathcal{W}^N, \Psi\mapsto\rho\right\}, \tag{14}$$

where $F_{\rm LL}[\rho]$ is the Levy-Lieb functional. The space $\mathcal{D}^N$ (characterized in [27]) is defined as the set of densities ρ such that $\Psi\mapsto\rho$ for some $\Psi\in\mathcal{W}^N$,

$$\mathcal{W}^N = \left\{\Psi\in\bigwedge_{i=1}^N H^1(\mathbb{R}^d\times\mathbb{Z}_2;\mathbb{C}) : \sum_{s_1,\dots,s_N\in\mathbb{Z}_2}\int_{\mathbb{R}^{dN}} |\nabla\Psi|^2 dz_1\dots dz_N < +\infty,\ ||\Psi|| = 1\right\}.$$

Analogously, when we replace ρ, N by ρ_α, N_α or by ρ_β, N_β, we obtain the ground state energies of the cluster of molecules α and β, which will be denoted, resp., by

$$E_0^\alpha[v_{\rm ne}^\alpha] = \inf_{\rho_\alpha\in\mathcal{D}^{N_\alpha}}\left\{F_{\rm LL}^\alpha[\rho_\alpha] + N_\alpha\int_{\mathbb{R}^d} v_{\rm ne}^\alpha(r)\rho_\alpha(r)dr\right\}, \tag{15}$$

and
$$E_0^\beta[v_{\rm ne}^\beta] = \inf_{\rho_\beta\in\mathcal{D}^{N_\beta}}\left\{F_{\rm LL}^\beta[\rho_\beta] + N_\beta\int_{\mathbb{R}^d} v_{\rm ne}^\beta(r)\rho_\beta(r)dr\right\}. \tag{16}$$

Strong interaction limit of Density Functional Theory: The Strong-Interaction limit functional is the limit when $\hbar\to 0^+$ of the Hohenberg-Kohn-Levy-Lieb functional (14) [8, 9, 35] (see also [13, 37] for a complete overview), and has the following form:

$$\mathrm{V}_{ee}^{\mathrm{SCE}}[\rho] := \min_{\gamma\in\Pi(\rho)} \int_{\mathbb{R}^{dN}} \mathrm{V}_{ee}(z_1, ..., z_N)\, d\gamma(z_1, ..., z_N), \tag{17}$$

where
$$\mathrm{V}_{ee}(z_1, \dots, z_N) := \sum_{1\le i<j\le N} \frac{1}{|z_i - z_j|}$$

and
$$\Pi(\rho) := \left\{\gamma \in \mathcal{P}_{sym}(\mathbb{R}^{dN}) :\ \gamma \mapsto \rho\right\},$$

in which $\mathcal{P}_{sym}(\mathbb{R}^{dN})$ denotes the set of probability measures that are invariant under permutation of the N coordinates in $\mathbb{R}^{dN} = (\mathbb{R}^d)^N$ and, analogously to (12), the notation $\gamma \mapsto \rho$ means that γ has all marginals equal to ρ, i.e. $(e_i)_\sharp\gamma = \rho$ for all $i \in \{1, \dots, N\}$ with $e_i : \mathbb{R}^{dN} \to \mathbb{R}^d$ the projection operator. The equation (17) corresponds to an optimal transport problem with finitely many marginals and Coulomb cost [1, 5, 8–11, 16, 18, 26].

2.1. A dissociation model in the strong interaction limit of DFT

Let $\eta > 0$ be a positive number and $\rho_\alpha, \rho_\beta \in \mathcal{P}(\mathbb{R}^3)$ be two probability measures in $\mathbb{R}^3$. Define the probability measure ρ_η by

$$\rho_\eta(z) := \frac{N_\alpha}{N_\alpha + N_\beta}\, \rho_\alpha(z - r_\alpha) + \frac{N_\beta}{N_\alpha + N_\beta}\, \rho_\beta(z - r_\beta), \tag{18}$$

where
$$r_\alpha = r_\alpha^{(\eta)} = \frac{1}{2\eta}\, \mathbf{e}_1 \quad \text{and} \quad r_\beta = r_\beta^{(\eta)} = -\frac{1}{2\eta}\, \mathbf{e}_1.$$

The probability density ρ_η is the single particle density of the composite system $\alpha\beta$ of $N = N_\alpha + N_\beta$ electrons formed by the isolated systems α and β having single-particle densities given by, respectively, ρ_α and ρ_β. This model captures the change of the single-particle density ρ_η of the system $\alpha\beta$ on varying the distance $R = \eta^{-1}$ between the two clusters α and β.

We are interested in the asymptotic development in $\eta \to 0^+$ of the SCE functional (17) $\mathrm{V}_{ee}^{\mathrm{SCE}}[\rho_\eta]$.

Center-of-molecule coordinates: As indicated by (18), as $|r_\alpha^{(\eta)} - r_\beta^{(\eta)}| \to \infty$ the positions of the two molecules α, β change, but the particle densities ρ_α and ρ_β do not change. It is then natural to work with new coordinates x, y, defined via

$$x := z - r_\alpha^{(\eta)}, \qquad y := z - r_\beta^{(\eta)}. \tag{19}$$

Then (18) can be written as

$$\rho_\eta(z) := \frac{N_\alpha}{N_\alpha + N_\beta}\rho_\alpha(x) + \frac{N_\beta}{N_\alpha + N_\beta}\rho_\beta(y). \tag{20}$$

We are now in the position to study the dissociation or electron-electron interaction of the entire cluster. As molecule α has N_α electrons and molecule β has N_β electrons, it is natural to use center-of-molecule coordinates in which N_α of the positions $z_1, \dots, z_N$ are re-centered at $r_\alpha^{(\eta)}$ and the remaining N_β are re-centered at $r_\beta^{(\eta)}$. With these choices, we rewrite the Coulomb electronic-electronic potential V_{ee} as

$$\mathrm{V}_{ee}^{\eta}(\vec{x},\vec{y}) = \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta} \frac{\eta}{\sqrt{1-2\eta(x_i^1-y_j^1)+\eta^2|x_i-y_j|^2}} + \mathrm{V}_{ee}^{\alpha}(\vec{x}) + \mathrm{V}_{ee}^{\beta}(\vec{y}), \tag{21}$$

where $\mathrm{V}_{ee}^{\alpha}(\vec{x}) = \sum_{1\le i<j\le N_\alpha} \frac{1}{|x_i-x_j|}$, and $\mathrm{V}_{ee}^{\beta}(\vec{y}) = \sum_{1\le i<j\le N_\beta} \frac{1}{|y_i-y_j|}$.

Plans that respect the molecule structure. In order to model the dissociation via a simplified energy we introduce the simplified minimization in which N_α electrons are assigned to molecule α and N_β electrons are assigned to molecule β. Thus rather than admissible plan sets $\Pi(\rho_\eta)$ (from (18)) we are led to consider the following spaces:

$$\Pi^{\eta}_{N_\alpha,N_\beta}(\rho_\alpha,\rho_\beta) := \left\{ \gamma\in\mathcal{P}(\mathbb{R}^{dN}) : \begin{array}{l} \exists\,\gamma_\alpha\in\Pi_{N_\alpha}(\rho_\alpha),\gamma_\beta\in\Pi_{N_\beta}(\rho_\beta) \text{ such} \\ \text{that } \gamma = \left((\tau_{r_\alpha^{(\eta)}})_\sharp\gamma_\alpha \otimes (\tau_{r_\beta^{(\eta)}})_\sharp\gamma_\beta\right)_{sym} \end{array} \right\}.$$

Here for $x\in\mathbb{R}^d$ we denote by $\tau_x:\mathbb{R}^{Kd}\to\mathbb{R}^{Kd}$ the translation operation

$$(x_1,\dots,x_K)\mapsto(x_1+x,\dots,x_K+x)$$

and for a measure $\widetilde{\gamma}$ over $\mathbb{R}^{Nd}$ we set $(\widetilde{\gamma})_{sym} := \frac{1}{N!}\sum_{\sigma\in\mathfrak{S}_n}\sigma_\sharp\widetilde{\gamma}$, where a permutation σ acts by $(x_1,\dots,x_N)\mapsto(x_{\sigma(1)},\dots,x_{\sigma(N)})$.

In other words, for defining $\Pi^{\eta}_{N_\alpha,N_\beta}(\rho_\alpha,\rho_\beta)$ we take $\gamma_\alpha,\gamma_\beta$ with respectively N_α marginals equal to ρ_α and N_β marginals equal to ρ_β and then we apply the translations by $r_\alpha^{(\eta)}, r_\beta^{(\eta)}$ to them. Finally, we symmetrize the so-obtained plan, an operation that insures direct comparability to the set of competitors for $\Pi(\rho_\eta)$ from (17). Indeed, we have the following:

Lemma 2.1. *Let $\eta>0$, $r_\alpha^{(\eta)}, r_\beta^{(\eta)}\in\mathbb{R}^d$, $\rho_\alpha,\rho_\beta\in\mathcal{P}(\mathbb{R}^d)$ and ρ_η as defined in* (20). *Then, $\Pi^{\eta}_{N_\alpha,N_\beta}(\rho_\alpha,\rho_\beta)\subset\Pi(\rho_\eta)$.*

Proof. Take $\gamma\in\Pi^{\eta}(\rho_\alpha,\rho_\beta)$, $\gamma_\alpha\in\Pi_{N_\alpha}(\alpha)$ and choose $\gamma_\beta\in\Pi_{N_\beta}(\beta)$ be such that $\gamma = \left((\tau_{r_\alpha^{(\eta)}})_\sharp(\gamma_\alpha)\otimes(\tau_{r_\beta^{(\eta)}})_\sharp(\gamma_\beta)\right)_{sym}$. Clearly, $\gamma\in\mathcal{P}(\mathbb{R}^d)$ and for every $i\in\{1,\dots,N\}$, we have

$$\begin{aligned}
(e_i)_\sharp\gamma &= (e_i)_\sharp\left((\tau_{r_\alpha^{(\eta)}})_\sharp(\gamma_\alpha)\otimes(\tau_{r_\beta^{(\eta)}})_\sharp(\gamma_\beta)\right)_{sym} \\
&= (e_i)_\sharp\left(\frac{1}{N!}\sum_{\sigma\in\mathfrak{S}_N}\sigma_\sharp\left((\tau_{r_\alpha^{(\eta)}})_\sharp(\gamma_\alpha)\otimes(\tau_{r_\beta^{(\eta)}})_\sharp(\gamma_\beta)\right)\right) \\
&= (e_i)_\sharp\left(\frac{1}{N!}\sum_{\sigma\in\mathfrak{S}_N}\gamma_\alpha(z_{\sigma(1)}-r_\alpha^{(\eta)},\dots,z_{\sigma(N_\alpha)}-r_\alpha^{(\eta)})\gamma_\beta(z_{\sigma(N_\alpha+1))}-r_\beta^{(\eta)},\dots,z_{\sigma(N_\alpha+N_\beta)}-r_\beta^{(\eta)})\right), \\
&= \frac{N_\alpha}{N_\alpha+N_\beta}\,\rho_\alpha(z_i-r_\alpha^{(\eta)}) + \frac{N_\beta}{N_\alpha+N_\beta}\,\rho_\beta(z_i-r_\beta^{(\eta)}) = \rho_\eta(z_i),
\end{aligned}$$

where we denoted by $\mathfrak{S}_N$ the set of permutations of N elements. □

Note that $\Pi(\rho_\eta)$ is strictly larger than $\Pi^{\eta}_{N_\alpha,N_\beta}(\rho_\alpha,\rho_\beta)$. Nevertheless, for $\eta\to0$ one may expect that optimizers of $\mathrm{V}_{ee}^{\mathrm{SCE}}[\rho_\eta]$ will tend to a minimizer of

the restriction of the functional from $\mathrm{V}_{ee}^{\mathrm{SCE}}$ to $\Pi^{\eta}_{N_\alpha,N_\beta}(\rho_\alpha,\rho_\beta)$. This is because we do not allow electron tunneling over large distances of $1/\eta$, and thus the requirement of N_α, N_β electrons in the molecules becomes sharp in the limit. Note that in [2,3] the tunneling question is left open. The question of proving the absence of tunneling rigorously is a technical one, and is not the focus of this paper. Instead, here, we use plans that respect the molecular structure and consider their asymptotic.

We note that the energy V_{ee} is symmetric under permutation of the N coordinates of $\mathbb{R}^{dN} = (\mathbb{R}^d)^N$, thus we have

$$\int_{\mathbb{R}^{dN}} \mathrm{V}_{ee}(\vec{z}) d(\gamma)_{sym}(z) = \int_{\mathbb{R}^{dN}} \mathrm{V}_{ee}(\vec{z}) d\gamma(\vec{z}),$$

therefore the problem we study can be reformulated as follows:

$$\begin{aligned}
&\min\left\{\int_{\mathbb{R}^{dN}} \mathrm{V}_{ee}(\vec{z})d\gamma(\vec{z}) : \gamma\in\Pi^{\eta}_{N_\alpha,N_\beta}(\rho_\alpha,\rho_\beta)\right\}\\
&=\min\left\{\int_{\mathbb{R}^{dN}} \mathrm{V}_{ee}(\vec{z})d\left((\tau_{r_\alpha^{(\eta)}})_\sharp\gamma_\alpha\otimes(\tau_{r_\beta^{(\eta)}})_\sharp\gamma_\beta\right)(\vec{z}) : \gamma_\alpha\in\Pi_{N_\alpha}(\rho_\alpha),\gamma_\beta\in\Pi_{N_\beta}(\rho_\beta)\right\}\\
&=\min\left\{\int_{\mathbb{R}^{dN_\alpha}}\int_{\mathbb{R}^{dN_\beta}} \mathrm{V}^{\eta}_{ee}(\vec{x},\vec{y})d\gamma_\alpha(\vec{x})d\gamma_\beta(\vec{y}) : \gamma_\alpha\in\Pi_{N_\alpha}(\rho_\alpha),\gamma_\beta\in\Pi_{N_\beta}(\rho_\beta)\right\}\\
&=\min_{\gamma\in\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)}\int_{\mathbb{R}^{dN}} \mathrm{V}^{\eta}_{ee}(\vec{x},\vec{y})d\gamma(\vec{x},\vec{y}).
\end{aligned}\tag{22}$$

To justify the above, in the first step we used the symmetry of V_{ee} and in the second step we used the rewriting (21) of V_{ee} in center-of-molecule coordinates and the structure of the plans which respect the coordinate splitting from $(\vec{z})$ to $(\vec{x},\vec{y})$.

Taylor expansion of the Coulomb interaction energy when $\eta\to 0^+$. We now compute the asymptotic expansion of the Coulomb electronic-electronic interaction in (21) when the distance $R=\eta^{-1}$ of the cluster of molecules $\alpha\beta$ goes to $+\infty$.

Proposition 2.2. *Let $\eta>0$ be a positive number, N_A, N_B be positive integers, $\rho_\alpha,\rho_\beta\in\mathcal{P}(\mathbb{R}^d)$ and $V^\eta(\vec{x},\vec{y})$ as in (21). Then, for $\gamma\in\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)$ we have*

$$\int_{\mathbb{R}^{dN}} V^\eta(\vec{x},\vec{y})d\gamma = \int_{\mathbb{R}^{dN}} V^\alpha_{ee}(\vec{x})d\gamma_\alpha + \int_{\mathbb{R}^{dN}} V^\beta_{ee}(\vec{x})d\gamma_\beta + U^\eta_{\mathrm{int}}[\rho_\alpha,\rho_\beta] + \tag{23}$$

$$+\frac{1}{2}\eta^3\int_{\mathbb{R}^{dN}}\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}\left(3(x_i^1-y_j^1)^2-|x_i-y_j|^2\right)d\gamma+O(\eta^4), \tag{24}$$

where $\gamma_\alpha\in\Pi_{N_\alpha}(\rho_\alpha),\gamma_\beta\in\Pi_{N_\beta}(\rho_\beta)$ and the internal energy $U^\eta_{\mathrm{int}}[\rho_\alpha,\rho_\beta]$ is defined by

$$U^\eta_{\mathrm{int}}[\rho_\alpha,\rho_\beta] := N_\alpha N_\beta\left(\eta+\left(\int_{\mathbb{R}^d}x^1 d\rho_\alpha(x)-\int_{\mathbb{R}^d}y^1 d\rho_\beta(y)\right)\eta^2\right). \tag{25}$$

Proof. We first compute the Taylor expansion of $V^\eta(\vec{x},\vec{y})$ at $\eta = 0$:

$$\begin{aligned} V^\eta(\vec{x},\vec{y}) &= V^0(\vec{x},\vec{y}) + \frac{dV}{d\eta}^0(\vec{x},\vec{y})\eta + \frac{1}{2}\frac{d^2V}{d\eta^2}^0(\vec{x},\vec{y})\eta^2 + O(\eta^3) \\ &= \mathrm{V}_{ee}^\alpha(\vec{x}) + \mathrm{V}_{ee}^\beta(\vec{y}) + N_\alpha N_\beta \eta + \eta^2 \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}(x_i^1 - y_j^1) + \\ &\quad + \frac{1}{2}\eta^3 \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}\left(3(x_i^1 - y_j^1)^2 - |x_i - y_j|^2\right) + O(\eta^3) \end{aligned}$$

Let $\gamma \in \Pi_{N_\alpha}(\rho_\alpha) \otimes \Pi_{N_\beta}(\rho_\beta)$. Then there exists $\gamma_\alpha \in \Pi_{N_\alpha}(\rho_\alpha)$ and $\gamma_\beta \in \Pi_{N_\beta}(\rho_\beta)$ such that $\gamma = \left((\tau_{r_\alpha^{(\eta)}})_\sharp \gamma_\alpha \otimes (\tau_{r_\beta^{(\eta)}})_\sharp \gamma_\beta\right)_{sym}$. Notice that, upon integrating the above expression against γ, the terms in the development of order 1 and η^2 are given by

$$\int_{\mathbb{R}^{dN}} V^0(\vec{x},\vec{y})d\gamma = \int_{\mathbb{R}^{dN}} V_{ee}^\alpha(\vec{x})d\gamma_\alpha(\vec{x}) + \int_{\mathbb{R}^{dN}} V_{ee}^\beta(\vec{x})d\gamma_\beta(\vec{y}),$$

and
$$\int_{\mathbb{R}^{dN}} \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}(x_i^1 - y_j^1)d\gamma = N_\alpha N_\beta \left(\int_{\mathbb{R}^d} x^1 d\rho_\alpha(x) - \int_{\mathbb{R}^d} y^1 d\rho_\beta(y)\right).$$

Therefore, we directly obtain (23) and (25). □

3. Multi-marginal Optimal Transport problem

In this section, we will present partial results on the existence and uniqueness of Monge solutions for the multi-marginal problem with cost function arising in the term of order η^3 in the expansion of $\eta \to 0^+$ in (23), namely

$$\min_{\gamma\in\Pi(\rho_\alpha)\otimes\Pi(\rho_\beta)} \frac{1}{2}\int_{\mathbb{R}^{dN}} \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}\left(3(x_i^1 - y_j^1)^2 - |x_i - y_j|^2\right) d\gamma(x_1,...,x_{N_\alpha},y_1,...,y_{N_\beta}). \quad (26)$$

The first theorem shows that if the marginal distributions ρ_α and ρ_β have finite second moments, then the problem (26) is equivalent to the multi-marginal optimal transport problem introduced in (1).

Proposition 3.1. *Let $N_\alpha, N_\beta \in \mathbb{N}$ be integers, $N = N_\alpha + N_\beta$, $X_i \subseteq \mathbb{R}^d$, $i = 1,\dots,N$ be open sets and $X := \prod_{i=1}^N X_i$. If ρ_α and ρ_β are probability measures in $\mathbb{R}^d$ having finite moments, then, for any transport plan $\gamma \in \Pi_{N_\alpha}(\rho_\alpha) \otimes \Pi_{N_\beta}(\rho_\beta)$, we have*

$$\begin{aligned} &\frac{1}{2}\int_X \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}\left(3(x_i^1 - y_j^1)^2 - |x_i - y_j|^2\right) d\gamma \\ &\qquad = \int_X \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta} x_i \cdot y_j^*\, d\gamma(x_1,\dots,x_{N_\alpha},y_1,\dots,y_{N_\beta}) + C. \end{aligned}$$

where $y_j^* = (-2y_j^1, y_j^2, \dots, y_j^d)$ for each $j \in \{1,\dots,N_\beta\}$ and $C \in \mathbb{R}$ is a constant.

In particular, we have

$$\operatorname*{argmin}_{\gamma\in\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)}\int_X\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}\left(3(x_i^1-y_j^1)^2-|x_i-y_j|^2\right)d\gamma$$
$$=\operatorname*{argmin}_{\gamma\in\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)}\int_X\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}x_i\cdot y_j^*d\gamma.$$

Proof. We just need to expand the squares in the cost function in (26) and notice that for all $\gamma\in\Pi_{N_\alpha}(\rho_\alpha)\otimes\Pi_{N_\beta}(\rho_\beta)$ we have

$$\frac{1}{2}\int_X\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}\left(3(x_i^1-y_j^1)^2-|x_i-y_j|^2\right)d\gamma=\int_X-\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}3x_i^1y_j^1+\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}x_i\cdot y_jd\gamma+C$$
$$=\int_X\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}x_i\cdot y_j^*d\gamma+C,$$

where $y_j^*=(-2y_j^1,y_j^2,\dots,y_j^d)$ for each $j\in\{1,...,N_\beta\}$ and the constant C is given by

$$C=\int_{\mathbb{R}^{dN}}\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}3(x_i^1)^2+\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}3(y_j^1)^2-\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}x_i^2-\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}y_j^2d\gamma$$
$$=N_\alpha N_\beta\int_{\mathbb{R}^d}3(x_i^1)^2d\rho_\alpha+N_\alpha N_\beta\int_{\mathbb{R}^d}3(y_j^1)^2d\rho_\beta-N_\alpha N_\beta\int_{\mathbb{R}^d}(x_i)^2d\rho_\alpha-N_\alpha N_\beta\int_{\mathbb{R}^d}(y_j)^2d\rho_\beta.$$

This completes the proof. □

Assuming finite second moments for the measures ρ_α and ρ_β, the corresponding Kantorovich dual problem of (26) is the variational problem below. Notice that the existence of the maximizer is guaranteed in G. H. Kellerer [21].

$$\max_{u\in L^1(\rho_\alpha\otimes\rho_\beta)}\Bigg\{N_\alpha\int_{\mathbb{R}^{dN_\alpha}}u(x)d\rho_\alpha(x)+N_\beta\int_{\mathbb{R}^{dN_\beta}}u(y)d\rho_\beta(y)$$
$$\text{such that}\quad\sum_{i=1}^{N_\alpha}u(x_i)+\sum_{j=1}^{N_\beta}u(y_j)\le\sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta}x_i\cdot y_j^*\Bigg\}.$$

3.1. Existence and uniqueness of Monge solutions

Let us now recall some main concepts from [22, 32].

Definition 3.2. Let $X_i\subseteq\mathbb{R}^d$, $i=1,\dots,N$ be open sets, $X:=\prod_{i=1}^N X_i$ and $c:X\to\mathbb{R}$ be a cost function. A set $S\subseteq X$ is called a *c-splitting set* if there are Borel functions $u_i:X_i\mapsto\mathbb{R}$ such that

$$\sum_{i=1}^N u_i(x_i)\le c(x_1,\dots,x_N)\tag{27}$$

for every $(x_1,\dots,x_N)\in X$, and whenever $(x_1,\dots,x_N)\in S$ equality holds. The functions $u_1(x_1),\dots,u_N(x_N)$ are called *c-splitting functions* for S. □

Assume $\{k_i\}_{i=1}^r \subseteq \{2,\dots,N\}$, with $k_1 < k_2 < \ldots < k_r$. For a given N-tuple of Borel functions $(u_1,\dots,u_N)$ satisfying inequality (27), and $x_1^0 \in X_1$, we define

$$M_{x_1^0 k_1 \dots k_r}(u_1,\dots,u_N) := \Bigg\{ (x_2,\dots,x_N) \in \prod_{i=2}^N X_i : Du_{k_i}(x_{k_i}) \text{ exists for each } i = 1,...,r \text{ and } u_1(x_1^0) + \sum_{i=2}^N u_i(x_i) = c(x_1^0, x_2, \dots, x_N) \Bigg\}$$

Remark 3.3. Note that for a given set of the form $M_{x_1^0 k_1 \dots k_r}(u_1,\dots,u_N)$, if $Du_1(x_1^0)$ exists then $D_{x_1} c(x_1^0, x_2, \dots, x_N)$ exists for every $(x_2,\dots,x_N) \in M_{x_1^0 k_1 \dots k_r}(u_1,\dots,u_N)$ and

$$Du_1(x_1^0) = D_{x_1} c(x_1^0, x_2, \dots, x_N)$$

(for details see Lemma 2.2 in [32]). By definition of the set $M_{x_1^0 k_1 \dots k_r}(u_1,\dots,u_N)$ we also have $Du_{k_i}(x_{k_i}) = D_{x_{k_i}} c(x_1^0, x_2, \dots, x_N)$ for every $i = 1,\dots,r$. □

Definition 3.4. Let $X_i \subseteq \mathbb{R}^d$, $i = 1,\dots,N$ be open sets, $X := \prod_{i=1}^N X_i$ and let $c : X \to \mathbb{R}$ be a semi-concave cost function. Let $\{k_i\}_{i=1}^r \subseteq \{2,\dots,N\}$, with $k_1 < k_2 < \ldots < k_r$. We say c is *twisted on c-splitting sets* with respect to the variables $x_1, x_{k_1}, \dots, x_{k_r}$ if for every N-tuple of Borel functions $(u_1,\dots,u_N)$ satisfying inequality (27) and for every $x_1^0 \in X_1$ with $M_{x_1^0 k_1 \dots k_r} \neq \emptyset$, we get that the map

$$(x_2,\dots,x_N) \mapsto D_{x_1} c(x_1^0, x_2, \dots, x_N)$$

is injective on the subset of $M_{x_1^0 k_1 \dots k_r}$ where $D_{x_1} c(x_1^0, x_2, \dots, x_N)$ exists. □

Note that the special case of c being twisted on c-splitting sets with respect to the variable x_1 is equivalent to the twisted on c-splitting sets condition.

Remark 3.5. The main result in [32] states that if c is twisted on c-splitting sets with respect to the variables $x_1, x_{k_1}, \dots, x_{k_r}$, with $\{k_i\}_{i=1}^r \subseteq \{2,\dots,N\}$, where $k_1 < k_2 < \ldots < k_r$, then the solution γ in (KP) is concentrated on a graph of a measurable map $u : \mathbb{R}^d \to \mathbb{R}^{rd}$ and it is unique, as long as $\rho_1, \rho_{k_1}, \dots, \rho_{k_r}$ are absolutely continuous with respect to the d-dimensional Lebesgue measure $\mathcal{L}^d$.

In this work, to get uniqueness we need the standard regularity condition on ρ_1. Under this assumption we can focus on the set formed of all $x_1^0 \in X_1$ such that $Du_1(x_1^0)$ exists for some N-tuple $(u_1,\dots,u_N)$ of Borel functions satisfying inequality (27), and so if $M_{x_1^0 k_1 \dots k_r} \neq \emptyset$, the map

$$(x_2,\dots,x_N) \mapsto D_{x_1} c(x_1^0, x_2, \dots, x_N)$$

is injective on the subset of $M_{x_1^0 k_1 \dots k_r}$ where $D_{x_1} c(x_1^0, x_2, \dots, x_N)$ exists if and only if $M_{x_1^0 k_1 \dots k_r}$ is a singleton. See details in [32]. □

Our main result establishes that the cost in (1) provides unique Monge solutions under two regularity conditions. Note that in the case $N_\alpha = 1$ the cost function in 1 reduces to the simple structure $x_1 \cdot \sum_{j=1}^{N_\beta} y_j^*$, which clearly gives uniqueness if the first marginal ρ_1 is absolutely continuous with respect to the d-dimensional Lebesgue measure $\mathcal{L}^d$. In what follows we prove uniqueness for the case $N_\alpha > 1$.

Theorem 3.6. *Let ρ_i be Borel probability measures on open bounded sets $X_i \subseteq \mathbb{R}^d$, $i = 1, \dots, N$, with ρ_1 absolutely continuous with respect to the d-dimensional Lebesgue measure $\mathcal{L}^d$. Assume $N_\alpha > 1$ and there exists $p \in \{1, \dots, N_\beta\}$ such that $\rho_{N_\alpha+p}$ is absolutely continuous with respect to the Lebesgue measure $\mathcal{L}^d$. Then the multi-marginal optimal transport problem* (1)

$$\inf_{\gamma\in\Pi(\rho_\alpha)\otimes\Pi(\rho_\beta)} \int_{\mathbb{R}^{dN}} c(\vec{x},\vec{y})d\gamma := \inf_{\gamma\in\Pi(\rho_\alpha)\otimes\Pi(\rho_\beta)} \int_{\mathbb{R}^{d(N_\alpha+N_\beta)}} \sum_{i=1}^{N_\alpha}\sum_{j=1}^{N_\beta} x_i\cdot y_j^* d\gamma(x_1,...,x_{N_\alpha},y_1,...,y_{N_\beta}),$$

admits a unique Monge solution. As before we write $y_j^* = (-2y_j^1, y_j^2, \dots, y_j^d)$ for each $j \in \{1, \dots, N_\beta\}$.

Proof. Let $(u_1, \dots, u_N)$ be an N-tuple of Borel functions satisfying inequality (27) and fix $x_1^0 \in X_1$ such that $Du_1(x_1^0)$ exists and $M_{x_1^0(N_\alpha+p)}(u_1, \dots, u_N) \neq \emptyset$. We want to prove that $M_{x_1^0(N_\alpha+p)}(u_1, \dots, u_N)$ is a singleton.

Let $(x_{2k}, \dots, x_{N_\alpha k}, y_{1k}, \dots, y_{N_\beta k}) \in M_{x_1^0(N_\alpha+p)}$, $k = 1, 2$. Then c is differentiable with respect to x_1 at $(x_1^0, x_{2k}, \dots, x_{N_\alpha k}, y_{1k}, \dots, y_{N_\beta k})$ (see Remark 3.3) and it satisfies

$$Du_1(x_1^0) = D_{x_1} c(x_1^0, x_{2k}, \dots, x_{N_\alpha k}, y_{1k}, \dots, y_{N_\beta k}) = \sum_{j=1}^{N_\beta} y_{jk}^*, \quad k = 1, 2. \tag{28}$$

We prove that $x_{i1} = x_{i2}$ and $y_{j1} = y_{j2}$ for each $i \in \{2, ..., N_\alpha\}$ and $j \in \{1, ..., N_\beta\}$. From equation (28) we get

$$\sum_{j=1}^{N_\beta} y_{j1}^* = \sum_{j=1}^{N_\beta} y_{j2}^*. \tag{29}$$

Fix $s \in A := \{1, \dots, N_\beta\}$ and $k \in \{1, 2\}$, and for convenience of notation set $x_1^0 = x_{11} = x_{12}$. Note that

$$\begin{aligned}
\{y_{jk}\}_{j\in A\setminus\{s\}} \in \mathsf{argmin}\Big\{ \{y_j\}_{j\in A\setminus\{s\}} \mapsto\ & c(x_1^0, x_{2k}, ..., x_{N_\alpha k}, y_1, ..., y_{s-1}, y_{sk}, y_{s+1}, ..., y_{N_\beta}) \\
& - \sum_{i=1}^{N_\alpha} u_i(x_{ik}) - \sum_{j=1,\ j\neq s}^{N_\beta} u_{N_\alpha+j}(y_j) - u_{N_\alpha+s}(y_{sk}) \Big\} \\
= \mathsf{argmin}\Big\{ \{y_j\}_{j\in A\setminus\{s\}} \mapsto\ & c(x_1^0, x_{2k}, ..., x_{N_\alpha k}, y_1, ..., y_{s-1}, y_{sk}, y_{s+1}, ..., y_{N_\beta}) \\
& - \sum_{j=1,\ j\neq s}^{N_\beta} u_{N_\alpha+j}(y_j) \Big\} \\
= \mathsf{argmin}\Big\{ \{y_j\}_{j\in A\setminus\{s\}} \mapsto\ & \sum_{i=1}^{N_\alpha}\sum_{j=1, j\neq s}^{N_\beta} x_{ik}\cdot y_j^* - \sum_{j=1, j\neq s}^{N_\beta} u_{N_\alpha+j}(y_j) \Big\}
\end{aligned}$$

Then

$$\sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i2}\cdot y_{j2}^* - \sum_{\substack{j=1\\ j\neq s}}^{N_\beta} u_{N_\alpha+j}(y_{j2}) \leq \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i2}\cdot y_{j1}^* - \sum_{\substack{j=1\\ j\neq s}}^{N_\beta} u_{N_\alpha+j}(y_{j1}), \tag{30}$$

and $$\sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i1}\cdot y^*_{j1} - \sum_{\substack{j=1\\ j\neq s}}^{N_\beta} u_{N_\alpha+j}(y_{j1}) \le \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i1}\cdot y^*_{j2} - \sum_{\substack{j=1\\ j\neq s}}^{N_\beta} u_{N_\alpha+j}(y_{j2}).$$

Adding the above inequalities and eliminating similar terms we get

$$\sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i2}\cdot y^*_{j2} + \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i1}\cdot y^*_{j1} \le \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i2}\cdot y^*_{j1} + \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i1}\cdot y^*_{j2},$$

or equivalently, $$\sum_{i=1}^{N_\alpha}(x_{i2}-x_{i1})\cdot\sum_{\substack{j=1\\ j\neq s}}^{N_\beta}\left(y^*_{j2}-y^*_{j1}\right)\le 0. \tag{31}$$

On the other hand we have

$$y_{sk}\in \operatorname{argmin}\Big\{y_s\mapsto c(x^0_1,x_{2k},...,x_{N_\alpha k},y_{1k},...,y_{(s-1)k},y_s,y_{(s+1)k},...,y_{N_\beta k})-u_{N_\alpha+s}(y_s)\Big\},$$

hence, we also get $$\sum_{i=1}^{N_\alpha}(x_{i2}-x_{i1})\cdot(y^*_{s2}-y^*_{s1})\le 0. \tag{32}$$

Adding inequalities (31) and (32) and using (29) we conclude that in particular, equality holds in (30). Then

$$\begin{aligned}\sum_{i=1}^{N_\alpha} u_i(x_{i2})+u_{N_\alpha+s}(y_{s2})-\sum_{i=1}^{N_\alpha} x_{i2}\cdot y^*_{s2} &= \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i2}\cdot y^*_{j2} - \sum_{\substack{j=1\\ j\neq s}}^{N_\beta} u_{N_\alpha+j}(y_{j2})\\ &= \sum_{i=1}^{N_\alpha}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} x_{i2}\cdot y^*_{j1} - \sum_{\substack{j=1\\ j\neq s}}^{N_\beta} u_{N_\alpha+j}(y_{j1}).\end{aligned}$$

$(x_{22},\dots,x_{N_\alpha 2},y_{11},\dots,y_{(s-1)1},y_{s2},y_{(s+1)1},\dots,y_{N_\beta 1})\in M_{x^0_1(N_\alpha+p)}(u_1,\dots,u_N)$. Then by Remark 3.3 we get

$$\begin{aligned}\sum_{\substack{j=1\\ j\neq s}}^{N_\beta} y^*_{j1}+y^*_{s2} &= D_{x_1}c(x^0_1,x_{22},\dots,x_{N_\alpha 2},y_{11},\dots,y_{(s-1)1},y_{s2},y_{(s+1)1},\dots,y_{N_\beta 1})\\ &= Du_1(x^0_1)=D_{x_1}c(x^0_1,x_{21},\dots,x_{N_\alpha 1},y_{11},\dots,y_{N_\beta 1})=\sum_{j=1}^{N_\beta} y^*_{j1},\end{aligned}$$

which implies $y^*_{s2}=y^*_{s1}$; that is,

$$y_{s2}=y_{s1}\quad\text{for every } s\in\{1,\dots,N_\beta\}. \tag{33}$$

In particular we have $y_{p1}=y_{p2}$ and by Remark 3.3

$$Du_{N_\alpha+p}(y_{p1})=Du_{N_\alpha+p}(y_{p2})=D_{y_p}c(x^0_1,x_{2k},\dots,x_{N_\alpha k},y_{1k},\dots,y_{N_\beta k})=\sum_{i=1}^{N_\alpha} x^*_{ik},$$

where $x^*_{ik}=(-2x^1_{ik},x^2_{ik},\dots,x^d_{ik})$ for each i, $k=1,2$.

Using the above equation, we proceed to make a straightforward adaptation of the arguments used from Equation (28) to Equation (33) to prove that $x_{s1} = x_{s2}$ for every $s \in \{2, \dots, N_\alpha\}$. This completes the proof of the theorem. □

Note that cost function in (1) does not include interaction among the variables $x_1, \dots, x_{N_\alpha}$, $N_\alpha \geq 2$. However, all of them interact with y_j for any $j \in \{1, \dots \beta\}$. So, it is natural to connect the variables $x_1, \dots, x_{N_\alpha}$ via one of the y_j, using an extra regularity condition on $\rho_{N_\alpha + j}$. The following simple Lemma shows that this condition is necessary.

Lemma 3.7. *Consider Dirac measures $\rho_{N_\alpha+j} = \delta_{\hat{y}_j}$, $j = 1, \dots, N_\beta$, and let ρ_i be any non Dirac measure, for some $i \in \{2, \dots, N_\alpha\}$. Then there exists a solution of non-Monge form to the Kantorovich problem.*

Proof. We have $\gamma = \overline{\gamma} \otimes \delta_{\hat{y}_1} \otimes \dots \otimes \delta_{\hat{y}_{N_\beta}}$ for any transport plan γ to the Kantorovich problem, where $\overline{\gamma}$ is a probability measure on $\prod_{i=1}^{N_\alpha} X_i$ with marginals $\rho_1, \dots, \rho_{N_\alpha}$.

$$\text{Then} \qquad \int_{\prod_{i=1}^{N} X_i} c d\gamma = \int_{\prod_{i=1}^{N_\alpha} X_i} c(x_1, \dots, x_{N_\alpha}, \hat{y}_1, \dots, \hat{y}_{N_\beta}) d\overline{\gamma}$$

$$= \sum_{i=1}^{N_\alpha} \sum_{j=1}^{N_\beta} \int_{\prod_{i=1}^{N_\alpha} X_i} x_i \cdot (\hat{y_j})^* d\overline{\gamma} = \sum_{i=1}^{N_\alpha} \sum_{j=1}^{N_\beta} \int_{X_i} x_i \cdot (\hat{y_j})^* d\rho_i.$$

We then deduce that any transport plan γ is a solution. In particular, $\rho_1 \otimes \dots \otimes \rho_N$ is solution of non-Monge form, as there exists $i \in \{2, \dots, \beta\}$ with ρ_i non Dirac measure. □

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