Fermion N-representability for prescribed density and paramagnetic current density

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The N-representability problem is the problem of determining whether there exists N-particle states with some prescribed property. Here we report an affirmative solution to the fermion N-representability problem when both the density and the paramagnetic current density are prescribed. This problem arises in current-density functional theory and is a generalization of the well-studied corresponding problem (only the density prescribed) in density functional theory. Given any density and paramagnetic current density satisfying a minimal regularity condition (essentially that a von Weizäcker–like canonical kinetic energy density is locally integrable), we prove that there exists a corresponding N-particle state. We prove this by constructing an explicit one-particle reduced density matrix in the form of a position-space kernel, i.e., a function of two continuous-position variables. In order to make minimal assumptions, we also address mathematical subtleties regarding the diagonal of, and how to rigorously extract paramagnetic current densities from, one-particle reduced density matrices in kernel form.

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I. INTRODUCTION

The question of N-representability has been studied extensively in quantum chemistry and related fields [1]. In particular, it plays an important role in density-functional theory (DFT). Given prescribed values for quantities in a fermionic system, e.g., its electron density or its reduced density matrix, one may ask whether it can be obtained from a Slater determinant, from a pure N-particle state, or from a mixed N-particle state. Regarding the density, it is well known that any density with a finite von Weizsäcker kinetic energy may be reproduced using a Slater determinant that also has finite kinetic energy [2–6]. For a one-particle reduced density matrix (1RDM), Slater-determinant representability is equivalent to idempotency; in general, however, pure-state N-representability of 1RDMs is a difficult and largely unsolved problem [7–9]. On the other hand, any 1RDM with spin-orbital occupation numbers (eigenvalues) in the range $[0,1]$ and trace N may be obtained from a mixed N-particle state.

In this paper, we report the solution to the mixed-state N-representability problem when both the density and the paramagnetic current density are prescribed. More precisely, we answer the following question: given a density $\rho(\mathbf{r})$ and a paramagnetic current density $\mathbf{j}_\mathrm{p}(\mathbf{r})$, does there exist a mixed state Γ with the prescribed density and current density, written $\Gamma \mapsto (\rho,\mathbf{j}_\mathrm{p})$? We answer this question affirmatively by constructing an explicit 1RDM, from which the existence of the N-particle state follows.

We note that standard constructions demonstrating the Slater-determinant N-representability when only the density is prescribed rely on the use of equidensity orbitals. Also, early work by Ghosh and Dhara [10] sketched a construction of such equidensity orbitals that reproduces both densities and currents. However, such solutions have limited scope, since the vorticity vanishes when orbitals give rise to the same density.

Lieb and Schrader have shown a Slater determinant representability result for $(\rho,\mathbf{j}_\mathrm{p})$ and $N \geqslant 4$ [11]. Clearly, N-representability via a Slater determinant implies representability via a mixed state. However, Lieb and Schrader's result requires $N \geqslant 4$, and they also give a counterexample for $N = 2$, where no Slater determinant can exist (with continuously differentiable and single valued orbital phase functions). Our result, while showing a weaker sense of N-representability, has no condition on N. Moreover, both the present work and Ref. [11] have mild regularity and decay assumptions on $(\rho,\mathbf{j}_\mathrm{p})$ that ensure representability, but these are different in the two approaches. The techniques of proof are also otherwise significantly different: Lieb and Schrader rely on the so-called smooth Hobby–Rice theorem, while our approach is by direct construction of a 1RDM. The present work and the work of Lieb and Schrader are complementary, offering two different points of view and solutions to a long-standing problem.

The remainder of this paper contains five sections. In Sec. II, we give some background information and establish notation. Following a discussion of the relationship between a reduced density matrix and its associated density and paramagnetic current density in Sec. III, we construct in Sec. IV a reduced density matrix for a prescribed density and paramagnetic current density. Section V contains some concluding remarks. Finally, two appendixes are also provided. Appendix A contains a brief overview of some mathematical concepts and results on Hilbert-Schmidt operators needed for the main results of Sec. III. Appendix B contains proofs of theorems in Sec. III.

II. BACKGROUND

The N-representability problem with prescribed density ρ and paramagnetic current density $\mathbf{j}_\mathrm{p}$ arises in current-density functional theory (CDFT) [12]. In CDFT, a magnetic vector potential $\mathbf{A}$, in addition the scalar potential v, enters the (spin-free) N-electron Hamiltonian. In atomic units,

$$H[v,\mathbf{A}] = \frac{1}{2}\sum_{k=1}^{N}[-i\nabla_k + \mathbf{A}(\mathbf{r}_k)]^2 + \sum_{k=1}^{N} v(\mathbf{r}_k) + \sum_{k<l}\frac{1}{r_{kl}}. \quad (1)$$

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Here $\mathbf{r}_k$ is the position of electron k, the operator $\boldsymbol{\nabla}_k$ differentiates with respect to $\mathbf{r}_k$, and r_{kl} is the distance between electrons k and l. The corresponding ground-state energy is given by the Rayleigh-Ritz variation principle,

$$E[v,\mathbf{A}] = \inf_{\Gamma} \mathrm{Tr}(\Gamma H[v,\mathbf{A}]), \tag{2}$$

where the minimization is over all mixed states Γ with a finite canonical kinetic energy

$$T[\Gamma] := \tfrac{1}{2}\,\mathrm{Tr}(\boldsymbol{\nabla}\Gamma\boldsymbol{\nabla}^\dagger). \tag{3}$$

Introducing the constrained-search universal functional:

$$F[\rho,\mathbf{j}_\mathrm{p}] = \inf_{\Gamma\mapsto\rho,\mathbf{j}_\mathrm{p}} \mathrm{Tr}\left(\Gamma H[0,\mathbf{0}]\right), \tag{4}$$

we may rewrite the Rayleigh-Ritz variation principle in Eq. (2) in the form of a Hohenberg-Kohn variation principle,

$$E[v,\mathbf{A}] = \inf_{\rho,\mathbf{j}_\mathrm{p}} \left\{ F[\rho,\mathbf{j}_\mathrm{p}] + \int \left[\rho\left(v + \frac{1}{2}A^2\right) + \mathbf{j}_\mathrm{p}\cdot\mathbf{A}\right] d\mathbf{r} \right\}. \tag{5}$$

The mixed-state N-representability problem is directly related to how large the search domain in Eq. (5) needs to be: if no $\Gamma \mapsto (\rho,\mathbf{j}_\mathrm{p})$ exists, then $F[\rho,\mathbf{j}_\mathrm{p}] = +\infty$ by definition.

In Kohn-Sham theory, the idea is to express the densities in Eq. (5) in terms of a single Slater determinant of noninteracting particles and to approximate the kinetic-energy contributions to $F[\rho,\mathbf{j}_\mathrm{p}]$ by the noninteracting kinetic energy,

$$T_s[\rho,\mathbf{j}_\mathrm{p}] = \inf_{\{\phi_k\}_{k=1}^N \mapsto \rho,\mathbf{j}_\mathrm{p}} \frac{1}{2}\sum_{k=1}^{N} \langle \boldsymbol{\nabla}\phi_k, \boldsymbol{\nabla}\phi_k\rangle, \tag{6}$$

where the infimum is over an orthonormal set of orbitals ϕ_k or, equivalently, the corresponding Slater determinants or idempotent 1RDMs. At this point, the Slater-determinant N-representability problem arises.

In general, densities ρ and $\mathbf{j}_\mathrm{p}$ arising from a single orbital have a vanishing paramagnetic vorticity,

$$\boldsymbol{\nu} = \boldsymbol{\nabla} \times \frac{\mathbf{j}_\mathrm{p}}{\rho} = 0, \tag{7}$$

except for possible Dirac δ-singularities at points $\mathbf{r}$ where $\rho(\mathbf{r}) = 0$. Consequently, a closed-shell two-particle Kohn-Sham system can only reproduce paramagnetic densities with vanishing vorticity. In general, therefore, an extended Kohn-Sham approach with fractional occupation numbers is required (see Refs. [13–16] for work in this direction),

$$\bar{T}_s[\rho,\mathbf{j}_\mathrm{p}] = \inf_{\{n_k\phi_k\}\mapsto\rho,\mathbf{j}_\mathrm{p}} \frac{1}{2}\sum_{k=1}^{\infty} n_k \langle \boldsymbol{\nabla}\phi_k, \boldsymbol{\nabla}\phi_k\rangle, \tag{8}$$

where orthonormality, $0 \leqslant n_k \leqslant 1$, and $\sum_k n_k = N$ are additional constraints on the infimum. Alternatively, since n_k and ϕ_k are eigenvalues and eigenvectors of 1RDMs, the minimization may equivalently be performed over 1RDMs. Here, the mixed-state N-representability problem appears.

For a mixed state $\Gamma \mapsto (\rho,\mathbf{j}_\mathrm{p})$, it is known that

$$T_\mathrm{W}[\rho] + T_\mathrm{p}[\rho,\mathbf{j}_\mathrm{p}] \leqslant T[\Gamma], \tag{9}$$

where the von Weizsäcker kinetic-energy functionals are given by

$$T_\mathrm{W}[\rho] := \frac{1}{8}\int \rho(\mathbf{r})^{-1}|\boldsymbol{\nabla}\rho(\mathbf{r})|^2 dr, \tag{10}$$

$$T_\mathrm{p}[\rho,\mathbf{j}_\mathrm{p}] := \frac{1}{2}\int \rho(\mathbf{r})^{-1}|\mathbf{j}_\mathrm{p}(\mathbf{r})|^2 dr. \tag{11}$$

A necessary condition for a finite-kinetic-energy representability is therefore that $T_\mathrm{W}[\rho] + T_\mathrm{p}[\rho,\mathbf{j}_\mathrm{p}] < +\infty$. For the case $\mathbf{j}_\mathrm{p} = 0$, this is also a sufficient condition. It is of interest to know whether this sufficiency generalizes to $\mathbf{j}_\mathrm{p} \neq 0$. In this paper, we prove sufficiency under mild additional conditions on the current density.

III. DIAGONALS OF DENSITY OPERATORS

We do not explicitly consider spin and therefore take as our point of departure an N-electron density matrix that depends only on spatial coordinates, with the spin coordinates integrated out:

$$\Gamma(\mathbf{r}_{1:N},\mathbf{s}_{1:N}) = \sum_i p_i \Psi_i(\mathbf{r}_{1:N})\Psi_i^*(\mathbf{s}_{1:N}). \tag{12}$$

Such a density matrix is an element of a Lebesgue space,

$$\Gamma \in L^2(\mathbb{R}^{3N}\times\mathbb{R}^{3N}), \tag{13}$$

and is symmetric with respect to permutations π of the coordinate labels, $(\mathbf{r}_k,\mathbf{s}_k) \overset{\pi}{\mapsto} (\mathbf{r}_{\pi(k)},\mathbf{s}_{\pi(k)})$. The right-hand side of Eq. (12) is a convex combination of properly normalized pure states, $\Psi_i \in L^2(\mathbb{R}^{3N})$, with coefficients $p_i \geqslant 0$ such that $\sum_i p_i = 1$. Moreover, each (spin-free) pure state is either totally symmetric or antisymmetric. In what follows, it does not matter whether each pure state is required to be antisymmetric, symmetric, or either antisymmetric or symmetric with respect to index permutations π of the spatial coordinates. We occasionally simplify the presentation by taking Γ to be a pure state.

A. Density matrices

The 1RDM belonging to a pointwise-defined Γ on the form (12) is given by the convex combination

$$D_\Gamma(\mathbf{r},\mathbf{s}) := N\sum_i p_i \int_{\mathbb{R}^{3N-3}} \Psi_i(\mathbf{r},\mathbf{r}_{2:N})\Psi_i^*(\mathbf{s},\mathbf{r}_{2:N}), d\mathbf{r}_{2:N} \tag{14}$$

and belongs to $L^2(\mathbb{R}^3\times\mathbb{R}^3)$. For pure states $\Gamma = |\Psi\rangle\langle\Psi|$, we may alternatively write D_Ψ. Due to permutation symmetry, the 1RDM is independent of which $N-1$ coordinates have been integrated out.

Given that $D_\Gamma \in L^2(\mathbb{R}^3\times\mathbb{R}^3)$, D_Γ is by definition the *kernel of a Hilbert-Schmidt integral operator*. Our discussion makes extensive use of this basic fact about the reduced density matrix. In particular, Γ and D_Γ are both *trace-class operators*, that is, Hilbert-Schmidt operators for which the matrix trace has a meaningful generalization. Let $\{\phi_k\} \subset L^2(X)$ be an orthonormal basis. By definition, A is a trace-class operator if

and only if the trace

$$\mathrm{Tr}\, A := \sum_k \langle \phi_k, A\phi_k \rangle \tag{15}$$

has a finite value, independent of the orthonormal basis.

For two Hilbert-Schmidt operators B and C, the kernel of the operator product $A = B * C$ is easily seen to be

$$(B * C)(x,y) := \int_X B(x,z)C(z,y)\, dz, \tag{16}$$

which is also Hilbert-Schmidt. Importantly, it can be shown that A is (the kernel of) a trace-class operator if and only if $A = B * C$ with B and C Hilbert-Schmidt operators. (Indeed, this is often taken as an alternative definition of trace-class operators.) The trace is then given by the integral of the diagonal [17],

$$\mathrm{Tr}\, A = \int_X (A * B)(x,x)\, dx. \tag{17}$$

If A is diagonable (e.g., symmetric positive semidefinite), then $\mathrm{Tr}\, A$ is the sum of the eigenvalues, like in the finite-dimensional case. For further information on these operator classes, see, for example, Ref. [18].

We denote by $\mathcal{D}_N$ the set of mixed N-electron states Γ and by $\mathcal{D}_{N,1}$ the set of 1RDMs that belong to some mixed N-electron state. The set $\mathcal{D}_{N,1}$ has the following well-known characterization:

Theorem 1. $\mathcal{D}_{N,1}$ consists of those $D \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ with the following properties:

(1) D is the kernel of a trace-class operator on $L^2(\mathbb{R}^3)$.

(2) D is Hermitian: $D(\mathbf{r},\mathbf{s}) = D^*(\mathbf{s},\mathbf{r})$ for almost all $(\mathbf{r},\mathbf{s})$.

(3) D is positive semidefinite: $0 \leqslant \int \phi^*(\mathbf{r})D(\mathbf{r},\mathbf{s})\phi(\mathbf{s})d\mathbf{s}$ for all $\phi \in L^2(\mathbb{R}^3)$.

(4) D has no eigenvalues greater than 2: $2 \geqslant \int \phi^*(\mathbf{r})D(\mathbf{r},\mathbf{s})\phi(\mathbf{s})d\mathbf{s}$ for all $\phi \in L^2(\mathbb{R}^3)$.

(5) D has eigenvalues that add up to N: $\mathrm{Tr}\, D = N$.

Proof. See Ref. [19], Sec. 2.6. ■

The last three conditions mean that $D(\mathbf{r},\mathbf{s})$ has eigenvalues in the interval $[0,2]$—eigenvalues interpretable as fermion occupation numbers—and that the sum of the eigenvalues $\mathrm{Tr}\, D$ is equal to N, the number of particles.

Since $D \in \mathcal{D}_{N,1}$ is Hermitian and positive, it is easy to show that there always exists a factorization of the form $D = G^\dagger * G$, meaning that we may write the density matrix in the form

$$D(\mathbf{r},\mathbf{s}) = (G^\dagger * G)(\mathbf{r},\mathbf{s}) = \int_{\mathbb{R}^3} G^*(\mathbf{u},\mathbf{s})G(\mathbf{u},\mathbf{r})d\mathbf{u}, \tag{18}$$

which plays an important role in the following.

B. Density

We now define the *density* ρ_Ψ associated with the wave function Ψ as

$$\rho_\Psi(\mathbf{r}) := D_\Psi(\mathbf{r},\mathbf{r}) = N \int_{\mathbb{R}^{3N-3}} |\Psi(\mathbf{r},\mathbf{r}_{2:N})|^2 d\mathbf{r}_{2:N}. \tag{19}$$

For almost all $\mathbf{r}$, it holds that $\Psi(\mathbf{r},\cdot) \in L^2(\mathbb{R}^{3N-3})$. Using the Cauchy-Schwarz inequality, we see from Eq. (19) that $\rho_\Psi(\mathbf{r}) = D_\Psi(\mathbf{r},\mathbf{r})$ is well defined for almost all $\mathbf{r}$. For a mixed state $\Gamma \in \mathcal{D}_N$, the density ρ_Γ is defined in the same manner but from D_Γ.

The following point is subtle but important here. We write $\Gamma \mapsto D$ whenever $\|D_\Gamma - D\|_{L^2(\mathbb{R}^3\times\mathbb{R}^3)} = 0$. This statement does not imply that that $D_\Gamma = D$ everywhere, only that D_Γ and D are equal as elements of $L^2(\mathbb{R}^3 \times \mathbb{R}^3)$. Consequently, D_Γ and D may differ at a set of measure zero, *including the totality of the diagonal*. Therefore, we need to examine carefully the validity or meaning of the statement "$\rho_\Gamma(\mathbf{r}) = D(\mathbf{r},\mathbf{r})$" for a state and density matrix related by $\Gamma \mapsto D$.

Suppose next that we are able to assign a diagonal $\mathrm{diag}\, D$ to D in some unambiguous way and let $\Gamma,\Gamma' \in \mathcal{D}_N$ be two (possibly distinct) states such that $\Gamma \mapsto D$ and $\Gamma' \mapsto D$, meaning that $D = D_\Gamma = D_{\Gamma'}$ almost everywhere in $\mathbb{R}^3 \times \mathbb{R}^3$. Is it then true that $\rho_\Gamma = \rho_{\Gamma'} = \mathrm{diag}\, D$ almost everywhere in $\mathbb{R}^3$? Intuitively, this should be so.

The following theorem, which is proved in Appendix B, resolves the issue:

Theorem 2. Let $D \in \mathcal{D}_{N,1}$, and suppose that $G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ is such that

$$D(\mathbf{r},\mathbf{s}) = (G^\dagger * G)(\mathbf{r},\mathbf{s})$$

almost everywhere in $\mathbb{R}^3 \times \mathbb{R}^3$. Then, for every $\Gamma \in \mathcal{D}_N$ such that $\Gamma \mapsto D$, it holds that

$$\rho_\Gamma(\mathbf{r}) = (G^\dagger * G)(\mathbf{r},\mathbf{r})$$

almost everywhere in $\mathbb{R}^3$.

Since the factorization of $D = G^\dagger * G$ does exist following the discussion in Sec. III A, it is indeed meaningful to talk about "the density ρ of D" without reference to a specific $\Gamma \mapsto D$:

$$\rho_D(\mathbf{r}) = \mathrm{diag}\, D(\mathbf{r}) := (G^\dagger * G)(\mathbf{r},\mathbf{r}) \quad \text{a.e.} \tag{20}$$

In particular, it follows that

$$\mathrm{Tr}\, D = \int \rho_D(\mathbf{r})\, d\mathbf{r}. \tag{21}$$

We emphasize that we only define the diagonal when a factorization is present and that this diagonal is independent of the factorization.

C. Momentum density

Before considering the momentum density, we note that all derivatives that occur in the subsequent treatment are *distributional or weak derivatives*. A function $f \in L^p(X)$, with $X \subset \mathbb{R}^n$ open, is said to have a weak derivative $g = \partial_\alpha f \in L^1_{\mathrm{loc}}(X)$ if, for all smooth, compactly supported "test functions" $u \in \mathcal{C}_c^\infty(X)$,

$$\int_X g(x)u(x)\, dx = -\int_X f(x)\partial_\alpha u(x)\, dx. \tag{22}$$

Thus, the weak derivative acts just like the standard derivative $\partial f/\partial x_\alpha$ when we apply integration by parts, coinciding with the classical derivative whenever this exists. Higher-order weak derivatives are defined in a similar manner. A standard monograph for weak derivatives is Ref. [20].

By analogy with the density in Eq. (19), we now define the *momentum density* $\mathbf{c}_\Psi$ of a state Ψ as

$$\begin{aligned}\mathbf{c}_\Psi(\mathbf{r}) &:= N \int_{\mathbb{R}^{3N-3}} [-i\nabla_{\mathbf{r}}\Psi(\mathbf{r},\mathbf{r}_{2:N})]\Psi^*(\mathbf{r},\mathbf{r}_{2:N})d\mathbf{r}_{2:N} \\ &= -i\nabla_{\mathbf{r}} D_\Psi(\mathbf{r},\mathbf{s})|_{\mathbf{r}=\mathbf{s}},\end{aligned} \tag{23}$$

whose real part is the paramagnetic current density

$$\mathbf{j}_{\mathrm{p}\Psi}(\mathbf{r}) = \mathrm{Re}\,\mathbf{c}_{\Psi}(\mathbf{r}) \tag{24}$$

with an analogous definition for a mixed state Γ. We note, however, that this definition may not make sense without additional assumptions on the wave function Ψ, beyond those needed for the definition of the density. We also observe that the second equality in Eq. (23) needs to be justified further since $\nabla_{\mathbf{r}} D_{\Psi}(\mathbf{r},\mathbf{s})|_{\mathbf{r}=\mathbf{s}}$ is only defined pointwise almost everywhere and since integration may not commute with differentiation.

To assign unambiguously a momentum density $\mathbf{c}_D(\mathbf{r})$ to $D \in \mathcal{D}_{N,1}$, we first introduce the notion of a *locally finite kinetic energy*.

Definition 1: Locally finite kinetic energy. We say that $D \in \mathcal{D}_{N,1}$ has a locally finite kinetic energy if the weak derivative $\boldsymbol{\nabla}_1 \cdot \boldsymbol{\nabla}_2 D$ is the kernel of a trace class operator over $L^2(K)$ for every compact $K \subset \mathbb{R}^3$. Likewise, we say that $\Psi \in L^2(\mathbb{R}^{3N})$ has a locally finite kinetic energy if $\boldsymbol{\nabla}_1 \Psi \in L^2(K \times \mathbb{R}^{3N-3})$ for every compact $K \subset \mathbb{R}^3$; that is, $\boldsymbol{\nabla}_1 \Psi \in L^2(\mathbb{R}^3_{\mathrm{loc}} \times \mathbb{R}^{3N-3})$. (See Appendix A 2.) A mixed state $\Gamma \in \mathcal{D}_N$ has a locally finite kinetic energy if $\sum_i p_i \|\boldsymbol{\nabla}_1 \Psi_i\|^2_{L^2(K\times\mathbb{R}^{3N-3})}$ is finite for every compact $K \subset \mathbb{R}^3$.

Note that the pure-state definition of locally finite kinetic energy follows from that of the mixed state. The various definitions of a locally finite kinetic energy are connected, as summarized in the following theorem, proved in Appendix B.

Theorem 3. For $D \in \mathcal{D}_{N,1}$, the following statements are equivalent:

(1) D has a locally finite kinetic energy.

(2) There exists a factorization $D = G^\dagger * G$ (almost everywhere) with G a Hilbert-Schmidt operator and $\nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3_{\mathrm{loc}})$.

(3) Any $\Gamma \in \mathcal{D}_N$ with $\Gamma \mapsto D$ has a locally finite kinetic energy.

If D has a locally finite kinetic energy, then the associated *kinetic-energy density* is defined as

$$\begin{aligned}\tau_D(\mathbf{r}) &:= \frac{1}{2}\|\nabla_2 G(\cdot,\mathbf{r})\|^2_{L^2(\mathbb{R}^3)} \\ &= \frac{1}{2}\int_{\mathbb{R}^3} [\nabla_2 G(\mathbf{u},\mathbf{r})]^* \cdot [\nabla_2 G(\mathbf{u},\mathbf{r})]\, d\mathbf{u} \\ &= \frac{1}{2}\,\mathrm{diag}(\nabla_1 \cdot \nabla_2 D)(\mathbf{r}),\end{aligned} \tag{25}$$

which is finite almost everywhere. From the proof of Theorem 3 in Appendix B, it follows that τ_D is in fact the kinetic energy density of any $\Gamma \mapsto D$. We also see that $\tau_D \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$ and that the total kinetic energy is finite if and only if $\tau_D \in L^1(\mathbb{R}^3)$.

Finally, the following theorem (proved in Appendix B) states that, if D has a locally finite kinetic energy, then the momentum density of Eq. (23) is also well defined.

Theorem 4. Let $D \in \mathcal{D}_{N,1}$ have a locally finite kinetic energy and let $G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ be such that $D = G^\dagger * G$ and $\nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3_{\mathrm{loc}})$. For each $\Gamma \in \mathcal{D}_N$ with $\Gamma \mapsto D$, it then holds that $\mathbf{c}_\Gamma \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$ and that

$$\begin{aligned}\mathbf{c}_\Gamma(\mathbf{r}) &= ([-i\nabla_2 G]^\dagger * G)(\mathbf{r},\mathbf{r}) \quad \text{a.e.} \\ &= \mathrm{diag}(-i\nabla_1 D)(\mathbf{r}).\end{aligned}$$

D. Summary

For easy reference, we collect the main conclusions of this section in a separate theorem.

Theorem 5. Let $D = G^\dagger * G$ with $G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3) \in \mathcal{D}_{N,1}$, $\nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3_{\mathrm{loc}})$. For every $\Gamma \in \mathcal{D}_N$ such that $\Gamma \mapsto D$, it then holds that the density $\rho_\Gamma = \rho \in L^1(\mathbb{R}^3)$, the momentum density $\mathbf{c}_\Gamma = \mathbf{c} \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$, and the kinetic energy density $\tau_\Gamma = \tau \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$ are given almost everywhere by the expressions

$$\begin{aligned}\rho(\mathbf{r}) &= \int_{\mathbb{R}^3} G^*(\mathbf{u},\mathbf{r}) G(\mathbf{u},\mathbf{s})\, d\mathbf{u}, \\ \mathbf{c}(\mathbf{r}) &= \int_{\mathbb{R}^3} [-i\nabla_2 G^*(\mathbf{u},\mathbf{r})]\, G(\mathbf{u},\mathbf{s})\, d\mathbf{u}, \\ \tau(\mathbf{r}) &= \frac{1}{2}\int_{\mathbb{R}^3} [\nabla_2 G^*(\mathbf{u},\mathbf{r})] \cdot [\nabla_2 G(\mathbf{u},\mathbf{s})]\, d\mathbf{u}.\end{aligned}$$

IV. A REDUCED DENSITY MATRIX FOR A PRESCRIBED DENSITY AND PARAMAGNETIC CURRENT DENSITY

Let a density ρ be given. We assume that the density is non-negative and that it belongs to the intersection of two Lebesgue spaces,

$$\rho(\mathbf{r}) \geqslant 0 \quad \text{and} \quad \rho \in L^1(\mathbb{R}^3) \cap L^q(\mathbb{R}^3), \tag{26}$$

for some $q > 1$. The latter condition amounts to

$$N := \|\rho\|_1 = \int |\rho(\mathbf{r})|\, d\mathbf{r} < +\infty, \tag{27}$$

$$\|\rho\|_q^q = \int |\rho(\mathbf{r})|^q\, d\mathbf{r} < +\infty, \tag{28}$$

where N is the number of particles in the density ρ. (For simplicity, we restrict ourselves to states with integral N but note the 1RDM constructions given below are valid also for fractional N.) Furthermore, let an arbitrary measurable vector-valued function $\boldsymbol{\kappa} : \mathbb{R}^3 \to \mathbb{R}^3$ be given and let it prescribe a paramagnetic current density by the relation

$$\mathbf{j}_{\mathrm{p}}(\mathbf{r}) = \tfrac{1}{2}\rho(\mathbf{r})\boldsymbol{\kappa}(\mathbf{r}). \tag{29}$$

We now consider the question: does there, for every pair of ρ and $\mathbf{j}_{\mathrm{p}}$ satisfying these minimal requirements, exist a $D \in \mathcal{D}_{N,1}$ that reproduces ρ and $\mathbf{j}_{\mathrm{p}}$? In short, we seek a reduced density matrix D such that

(a) $\rho(\mathbf{r}) = \rho_D(\mathbf{r}) = (\mathrm{diag}\, D)(\mathbf{r})$,

(b) $\mathbf{j}_{\mathrm{p}}(\mathbf{r}) = \mathrm{Re}\,\mathbf{c}_D(\mathbf{r}) = -\frac{i}{2}(\mathrm{diag}\,\nabla_1 D)(\mathbf{r}) + \text{c.c.}$,

assuming that D has a locally finite kinetic energy for requirement (b) to be well defined. We can indeed find such a density matrix $D \in \mathcal{D}_{N,1}$ but we will later see that the condition of a locally finite kinetic energy of D implies mild additional conditions on ρ and $\boldsymbol{\kappa}$.

A. Factorized elements P_λ and Q_λ

Our strategy is to construct explicitly factorized elements $P_\lambda = G^\dagger_\lambda * G_\lambda$ and $Q_\mu = H^\dagger_\mu * H_\mu$ in $\mathcal{D}_{N,1}$ with a locally finite kinetic energy. Here, $\lambda, \mu > 0$ are real parameters that allow some freedom, noting that a convex combination $D_{\lambda\mu} = (P_\lambda + Q_\mu)/2$ remains in $\mathcal{D}_{N,1}$, also with a locally finite kinetic energy. The flexibility of having several independent

factorized reduced density matrices P_λ and Q_μ allows the convex combination to reproduce the desired current.

The two terms are defined by the factorized expressions

$$P_\lambda(\mathbf{r},\mathbf{s}) = \sqrt{\rho(\mathbf{r})\rho(\mathbf{s})}\int_{\mathbb{R}^3} g^*(\mathbf{u},\mathbf{r})g(\mathbf{u},\mathbf{s})\,d\mathbf{u}, \tag{30}$$

$$Q_\mu(\mathbf{r},\mathbf{s}) = \sqrt{\rho(\mathbf{r})\rho(\mathbf{s})}\int_{\mathbb{R}^3} h^*(\mathbf{u},\mathbf{r})h(\mathbf{u},\mathbf{s})\,d\mathbf{u}, \tag{31}$$

where

$$g(\mathbf{u},\mathbf{v}) = \frac{\sqrt{8}\lambda^{3/4}}{\pi^{3/4}} e^{-i\mathbf{v}\cdot\boldsymbol{\kappa}(\mathbf{v})} e^{-2\lambda(\mathbf{u}-\mathbf{v})^2}, \tag{32}$$

$$h(\mathbf{u},\mathbf{v}) = \frac{\sqrt{8}\mu^{3/4}}{\pi^{3/4}} e^{i\mathbf{u}\cdot\boldsymbol{\kappa}(\mathbf{v})} e^{-2\mu(\mathbf{u}-\mathbf{v})^2}. \tag{33}$$

Clearly, these operators may be written in the form

$$P_\lambda = G_\lambda^\dagger * G_\lambda, \quad G_\lambda(\mathbf{r},\mathbf{s}) = g(\mathbf{r},\mathbf{s})\sqrt{\rho(\mathbf{s})}, \tag{34}$$

$$Q_\mu = H_\mu^\dagger * H_\mu, \quad H_\mu(\mathbf{r},\mathbf{s}) = h(\mathbf{r},\mathbf{s})\sqrt{\rho(\mathbf{s})}. \tag{35}$$

It is straightforward to verify that $G_\lambda, H_\lambda \in L^2(\mathbb{R}^3\times\mathbb{R}^3)$.

The integration over $\mathbf{u}$ may be performed analytically, yielding the alternative expressions

$$P_\lambda(\mathbf{r},\mathbf{s}) = \sqrt{\rho(\mathbf{r})\rho(\mathbf{s})} e^{-\lambda|\mathbf{r}-\mathbf{s}|^2} e^{i[\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{r})-\mathbf{s}\cdot\boldsymbol{\kappa}(\mathbf{s})]}, \tag{36}$$

$$\begin{aligned} Q_\mu(\mathbf{r},\mathbf{s}) = {}& \sqrt{\rho(\mathbf{r})\rho(\mathbf{s})} e^{-\mu|\mathbf{r}-\mathbf{s}|^2} \\ & \times e^{-\frac{i}{2}(\mathbf{r}+\mathbf{s})\cdot[\boldsymbol{\kappa}(\mathbf{r})-\boldsymbol{\kappa}(\mathbf{s})]-|\boldsymbol{\kappa}(\mathbf{r})-\boldsymbol{\kappa}(\mathbf{s})|^2/16\mu}. \end{aligned} \tag{37}$$

These operators were found by making the initial ansatz $\phi(\mathbf{r}) = \sqrt{\rho(\mathbf{r})}e^{i\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{r})}$ for an unnormalized natural orbital. The corresponding paramagnetic current is then almost correct but contains an extra term that is most easily canceled if the density matrix contains exponential factors of the form $e^{i\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{s})}$. Since the elements of $\mathcal{D}_{N,1}$ and their properties are conveniently described if an explicit factorization is available (see Theorem 5), Gaussian kernels are suitable since since they allow mixed phase factors of the type $e^{i\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{s})}$ to survive the integration.

B. The density of P_λ and Q_λ

We now need to verify that P_λ and Q_μ are elements of $\mathcal{D}_{N,1}$ by checking points (1)–(5) of Theorem 1.

Theorem 6. Let $\rho \in L^1(\mathbb{R}^3)\cap L^q(\mathbb{R}^3)$ for some $q>1$, $\rho \geqslant 0$ a.e., $\|\rho\|_1 = N$, and let $\lambda,\mu\in\mathbb{R}$ be such that

$$\lambda,\mu \geqslant \frac{2p}{\pi}\left(\frac{1}{4}N\|\rho\|_q\right)^{2p/3},$$

where $1/p+1/q=1$. Then P_λ and Q_μ in Eqs. (30)–(33) are elements of $\mathcal{D}_{N,1}$, with

$$\rho_{P_\lambda}(\mathbf{r}) = \rho_{Q_\mu}(\mathbf{r}) = \rho(\mathbf{r})$$

almost everywhere. The same is true for any convex combination $\theta P_\lambda + (1-\theta)Q_\mu \in \mathcal{D}_{N,1}$ with $\theta\in[0,1]$.

Proof. Both operators are Hermitian and positive semidefinite. From the expressions in Eqs. (36) and (37), $(\operatorname{diag} P_\lambda)(\mathbf{r}) = (\operatorname{diag} Q_\lambda)(\mathbf{r}) = \rho(\mathbf{r})$ almost everywhere. It follows that $\operatorname{Tr} P_\lambda = \operatorname{Tr} Q_\lambda = \int\rho(\mathbf{r})d\mathbf{r} = N$.

It remains to compute a bound on the largest eigenvalues, demonstrating point (4) of Theorem 1 for the corresponding parameter values λ and μ. For an arbitrary normalized orbital,

$$\begin{aligned} n^2 &\leqslant \left|\int \phi^*(\mathbf{r})P_\lambda(\mathbf{r},\mathbf{s})\phi(\mathbf{s})\,d\mathbf{r}d\mathbf{s}\right|^2 \\ &\leqslant \left(\int |\phi(\mathbf{r})|\sqrt{\rho(\mathbf{r})\rho(\mathbf{s})}e^{-\lambda|\mathbf{r}-\mathbf{s}|^2}|\phi(\mathbf{s})|\,d\mathbf{r}\,d\mathbf{s}\right)^2 \\ &= \left(\int |\phi(\mathbf{r})|\sqrt{\rho(\mathbf{r})}\left(\int\sqrt{\rho(\mathbf{s})}e^{-\lambda|\mathbf{r}-\mathbf{s}|^2}|\phi(\mathbf{s})|d\mathbf{s}\right)d\mathbf{r}\right)^2. \end{aligned} \tag{38}$$

Given that $\phi,\sqrt{\rho}\in L^2(\mathbb{R}^3)$, the Cauchy-Schwarz inequality may be applied twice to give

$$\begin{aligned} n^2 &\leqslant \left(\int|\phi(\mathbf{r}')|^2d\mathbf{r}'\right) \\ &\quad\times\left(\int\rho(\mathbf{r})\left(\int\sqrt{\rho(\mathbf{s})}e^{-\lambda|\mathbf{r}-\mathbf{s}|^2}|\phi(\mathbf{s})|d\mathbf{s}\right)^2 d\mathbf{r}\right) \\ &\leqslant \int\rho(\mathbf{r})\left(\int|\phi(\mathbf{s}')|^2d\mathbf{s}'\int\rho(\mathbf{s})e^{-2\lambda|\mathbf{r}-\mathbf{s}|^2}d\mathbf{s}\right)d\mathbf{r} \\ &\leqslant \int\rho(\mathbf{r})\left(\sup_{\mathbf{c}}\int\rho(\mathbf{s})e^{-2\lambda|\mathbf{c}-\mathbf{s}|^2}d\mathbf{s}\right)d\mathbf{r} \\ &= N\sup_{\mathbf{c}}\int\rho(\mathbf{s})e^{-2\lambda|\mathbf{c}-\mathbf{s}|^2}d\mathbf{s}. \end{aligned} \tag{39}$$

Finally, exploiting the fact that $\rho\in L^q(\mathbb{R}^3)$, the integral over $\mathbf{s}$ may be bounded by invoking the Hölder inequality,

$$n^2 \leqslant N\|\rho\|_q\sup_{\mathbf{c}}\|e^{-2\lambda|\mathbf{c}-\mathbf{s}|^2}\|_p = N\|\rho\|_q\left(\frac{\pi}{2p\lambda}\right)^{3/2p}, \tag{40}$$

where $1/p+1/q=1$. This bound is independent of the current density. Hence, P_λ has no eigenvalues greater than 2 if

$$\lambda \geqslant \frac{2p}{\pi}\left(\frac{1}{4}N\|\rho\|_q\right)^{2p/3}. \tag{41}$$

These steps hold also for Q_μ, showing that it has no eigenvalues greater than 2 when $\mu\geqslant\frac{2p}{\pi}(\frac{1}{4}N\|\rho\|_q)^{2p/3}$.

Finally, consider a convex combination $D_\theta = \theta P_\lambda + (1-\theta)Q_\mu$, which belongs to $\mathcal{D}_{N,1}$ since this set is convex. Moreover, $\operatorname{diag} A$ is linear in A since $\operatorname{diag}(A+B)(\mathbf{r}) = \operatorname{diag}(A)(\mathbf{r}) + \operatorname{diag}(B)(\mathbf{r})$ almost everywhere. Therefore, $\operatorname{diag} D_\theta = \theta\operatorname{diag} P_\lambda + (1-\theta)\operatorname{diag} Q_\lambda = \rho$ almost everywhere. ■

C. The canonical kinetic energy of P_λ and Q_λ

We now turn to the question of whether the current $\mathbf{j}_\mathrm{p}$ can be reproduced by D. Indeed, a formal calculation shows that

$$-\frac{i}{2}\frac{\partial}{\partial r_\alpha}P_\lambda(\mathbf{r},\mathbf{s})|_{\mathbf{s}=\mathbf{r}} + \text{c.c.} = \rho(\mathbf{r})\left(\kappa_\alpha(\mathbf{r}) + \mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha}\right) = 2j_{\mathrm{p};\alpha}(\mathbf{r}) + \rho(\mathbf{r})\,\mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha} \tag{42}$$

and

$$-\frac{i}{2}\frac{\partial}{\partial r_\alpha}Q_\mu(\mathbf{r},\mathbf{s})|_{\mathbf{s}=\mathbf{r}} + \text{c.c.} = -\rho(\mathbf{r})\,\mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha}. \tag{43}$$

Thus, we expect $\mathbf{j}_{\mathrm{p}\,D_{\lambda\mu}} = \frac{1}{2}\mathbf{j}_{\mathrm{p}\,P_\lambda}(\mathbf{r}) + \frac{1}{2}\mathbf{j}_{\mathrm{p}\,Q_\mu}(\mathbf{r}) = \mathbf{j}_\mathrm{p}(\mathbf{r})$ to hold almost everywhere. To prove this result, it suffices to find conditions on ρ and κ such that P_λ and Q_μ have a locally finite kinetic energy.

The kinetic energy density of P_λ is

$$\tau_P(\mathbf{r}) = \frac{1}{2}\|\nabla_2 G_\lambda(\cdot,\mathbf{r})\|^2_{L^2(\mathbb{R}^3)} = \frac{1}{2}\nabla_\mathbf{r}\cdot\nabla_\mathbf{s}P_\lambda(\mathbf{r},\mathbf{s})|_{\mathbf{s}=\mathbf{r}} = \frac{|\nabla\rho(\mathbf{r})|^2}{8\rho(\mathbf{r})} + \frac{1}{2}|\nabla(\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{r}))|^2\rho(\mathbf{r}) + \lambda\rho(\mathbf{r}). \tag{44}$$

Here, we have used the fact that the integral in $\|\nabla_2 G(\cdot,\mathbf{r})\|^2_{L^2(\mathbb{R}^3)}$ can be performed analytically, so that the evaluation at $\mathbf{s}=\mathbf{r}$ after the second equality is in fact well defined. Similarly,

$$\tau_Q(\mathbf{r}) = \frac{1}{2}\nabla_\mathbf{r}\cdot\nabla_\mathbf{s}Q_\mu(\mathbf{r},\mathbf{s})|_{\mathbf{s}=\mathbf{r}} = \frac{|\nabla\rho(\mathbf{r})|^2}{8\rho(\mathbf{r})} + \frac{1}{2}\left[\sum_{\alpha=1}^3\left(\sum_{\beta=1}^3 r_\beta\frac{\partial\kappa_\beta(\mathbf{r})}{\partial r_\alpha}\right)^2 + \frac{1}{8\mu}\sum_{\beta=1}^3|\nabla\kappa_\beta(\mathbf{r})|^2\right]\rho(\mathbf{r}) + \mu\rho(\mathbf{r}).$$

The total kinetic energy density becomes

$$\tau_D(\mathbf{r}) = \frac{|\nabla\rho(\mathbf{r})|^2}{8\rho(\mathbf{r})} + \frac{1}{4}\sum_{\alpha=1}^3\left[\left(\frac{\partial}{\partial r_\alpha}\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{r})\right)^2 + \left(\mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha}\right)^2 + \frac{1}{8\mu}\sum_{\beta=1}^3\left(\frac{\partial\kappa_\beta(\mathbf{r})}{\partial r_\alpha}\right)^2\right]\rho(\mathbf{r}) + \frac{1}{2}(\lambda+\mu)\rho(\mathbf{r}), \tag{45}$$

where we have used the fact that the kinetic energy density is linear in the density matrix. Using the special form $2|ab| \leqslant a^2 + b^2$ of Young's inequality, with $a = \kappa_\alpha(\mathbf{r})$ and $b = \mathbf{r}\cdot\partial\boldsymbol{\kappa}(\mathbf{r})/\partial r_\alpha$, we find that

$$\left|\frac{\partial}{\partial r_\alpha}\mathbf{r}\cdot\boldsymbol{\kappa}(\mathbf{r})\right|^2 = \left|\kappa_\alpha(\mathbf{r}) + \mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha}\right|^2 \leqslant 2|\kappa_\alpha(\mathbf{r})|^2 + 2\left|\mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha}\right|^2. \tag{46}$$

Hence, the canonical kinetic energy density is bounded by

$$\begin{aligned}\tau_D(\mathbf{r}) &\leqslant \frac{|\nabla\rho(\mathbf{r})|^2}{8\rho(\mathbf{r})} + \frac{1}{2}\left[|\boldsymbol{\kappa}(\mathbf{r})|^2 + \frac{3}{2}\left(\mathbf{r}\cdot\frac{\partial\boldsymbol{\kappa}(\mathbf{r})}{\partial r_\alpha}\right)^2 + \frac{1}{16\mu}\sum_{\beta=1}^3\left(\frac{\partial\kappa_\beta(\mathbf{r})}{\partial r_\alpha}\right)^2\right]\rho(\mathbf{r}) + \frac{1}{2}(\lambda+\mu)\rho(\mathbf{r}) \\ &\leqslant \frac{|\nabla\rho(\mathbf{r})|^2}{8\rho(\mathbf{r})} + \frac{1}{2}\left[|\boldsymbol{\kappa}(\mathbf{r})|^2 + \left(\frac{3}{2}r^2 + \frac{1}{16\mu}\right)\sum_{\alpha,\beta=1}^3\left(\frac{\partial\kappa_\beta(\mathbf{r})}{\partial r_\alpha}\right)^2\right]\rho(\mathbf{r}) + \frac{1}{2}(\lambda+\mu)\rho(\mathbf{r}),\end{aligned} \tag{47}$$

where the second inequality was obtained by using $|r_\beta| \leqslant |\mathbf{r}|$. A finite canonical kinetic energy of P_λ, Q_μ, and $D_{\lambda\mu}$ is ensured if

$$T_\mathrm{W}[\rho] = \int\frac{|\nabla\rho|^2}{8\rho}\,d\mathbf{r} = \frac{1}{2}\int|\nabla\sqrt{\rho}|^2\,d\mathbf{r} < \infty, \tag{48}$$

$$T_\mathrm{p}[\rho,\mathbf{j}_\mathrm{p}] = \int\frac{|\mathbf{j}_\mathrm{p}|^2}{2\rho}\,d\mathbf{r} = \frac{1}{8}\int\rho\kappa^2\,d\mathbf{r} < \infty, \tag{49}$$

$$T_{\alpha\beta}[\rho,\mathbf{j}_\mathrm{p}] = \int(1+r^2)\rho\left(\frac{\partial\kappa_\beta(\mathbf{r})}{\partial r_\alpha}\right)^2 d\mathbf{r} < \infty. \tag{50}$$

We remark that, by the definition of the vorticity, $\boldsymbol{\nu} = \nabla\times\rho^{-1}\mathbf{j}_\mathrm{p} = \frac{1}{2}\nabla\times\boldsymbol{\kappa}$, a consequence of the last condition is that

$$\int(1+r^2)\rho\nu^2 d\mathbf{r} < \infty. \tag{51}$$

We have thus proved the following result.

Theorem 7. Let ρ and $\boldsymbol{\kappa}$ be given such that $\rho \geqslant 0$, $\sqrt{\rho} \in H^1(\mathbb{R}^3)$, $\rho\kappa_\alpha^2 \in L^1(\mathbb{R}^3)$, $(1+r^2)\rho(\partial\kappa_\beta/\partial r_\alpha)^2 \in L^1(\mathbb{R}^3)$ for all Cartesian components $\alpha,\beta\in\{1,2,3\}$. Then there exist real constants $\lambda,\mu \geqslant 0$ and a 1RDM D with density ρ and current $\mathbf{j}_\mathrm{p} = \frac{1}{2}\rho\boldsymbol{\kappa}$ such that the canonical kinetic energy is bounded by

$$\begin{aligned}\frac{1}{2}\,\mathrm{Tr}(\nabla_1\cdot\nabla_2 D) \leqslant T_W[\rho] + 4T_\mathrm{p}\left[\rho,\frac{1}{2}\rho\kappa\right] + \frac{1}{2}(\lambda+\mu)N \\ + \int\rho(\mathbf{r})\left(\frac{3}{4}r^2 + \frac{1}{32\mu}\right)\sum_{\alpha,\beta=1}^3\left(\frac{\partial\kappa_\beta(\mathbf{r})}{\partial r_\alpha}\right)^2 d\mathbf{r}.\end{aligned}$$

Note that, by a Sobolev inequality, $\sqrt{\rho}\in H^1(\mathbb{R}^3)$ implies that $\rho\in L^q(\mathbb{R}^3)$ for all $q\in[1,3]$.

D. Lifting the global integrability condition

Theorem 7 may be strengthened by replacing the global integrability conditions on the total kinetic energy by local integrability conditions, replacing integrals $\mathbb{R}^3$ by integrals over arbitrary compact sets $K \subset \mathbb{R}^3$. A larger class of ρ and $\boldsymbol{\kappa}$ is then seen to be reproducible, albeit with merely a locally finite kinetic energy.

Theorem 8. Let ρ and $\boldsymbol{\kappa}$ be given such that $\rho \geqslant 0$, $\rho \in L^1(\mathbb{R}^3) \cap L^q(\mathbb{R}^3)$, $q > 1$, $\rho^{-1}|\nabla\rho|^2 \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$, $\rho\kappa_\alpha^2 \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$, $(1+r^2)\rho(\partial\kappa_\beta/\partial r_\alpha)^2 \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$ for all Cartesian components $\alpha,\beta \in \{1,2,3\}$. Then there exists a 1RDM D with density ρ and current $\mathbf{j}_\mathrm{p} = \frac{1}{2}\rho\boldsymbol{\kappa}$.

V. DISCUSSION

We have provided an explicit construction of a 1RDM that reproduces a prescribed density and paramagnetic current density. This type of N-representability problem arises in Kohn-Sham CDFT, as it is known that not all current densities can be represented by a single Kohn-Sham orbital. Lieb and Schrader have recently proved [11], under some additional assumptions, that there also exist current densities that cannot be represented by two Kohn-Sham orbitals. The question is open for three orbitals. For four or more orbitals, Lieb and Schrader provide an explicit Slater determinant that reproduces any density and paramagnetic current that satisfy mild regularity conditions. Our results are complementary in that we establish that an *extended* Kohn-Sham approach, where fractional occupation numbers are allowed even if there is an integral total number of electrons, is flexible enough to represent any density and paramagnetic current density, under minimal regularity assumptions (finite T_W, T_p, and $T_{\alpha\beta}$).

The generalization from finite total canonical kinetic energy to finite local canonical kinetic energy is of some value in light of gauge freedom. The kinetic energy $T_\mathrm{p}[\rho,\mathbf{j}_\mathrm{p}]$ is not gauge invariant; on the contrary, it can made to become infinite by applying a gauge transformation $\mathbf{j}_\mathrm{p} \mapsto \mathbf{j}_\mathrm{p} + \rho\nabla\chi$ with a rapidly growing gauge function χ. Our results establish that such gauge transformations do not affect N-representability, as long as χ exhibits some minimal regularity.

The explicit constructions of density matrices can be used to provide orbital-free upper bounds on the canonical kinetic energy $T_s[\rho,\mathbf{j}_\mathrm{p}]$ for an extended Kohn-Sham formalism. Combining the above results with the standard lower bound $T_\mathrm{W} + T_\mathrm{p}$ on the kinetic energy, we get the following orbital-free bounds on the extended Kohn-Sham kinetic energy:

$$\begin{aligned} T_\mathrm{W} + T_\mathrm{p} &\leqslant \bar{T}[\rho,\mathbf{j}_\mathrm{p}] \\ &\leqslant T_\mathrm{W} + 4T_\mathrm{p} + \frac{1}{2}(\lambda+\mu)N \\ &\quad + \int \rho(\mathbf{r}) \left(\frac{3}{4}r^2 + \frac{1}{32\mu}\right) \left|\nabla_\alpha\kappa_\beta(\mathbf{r})\right|^2 d\mathbf{r}. \end{aligned} \tag{52}$$

Noting that several authors, following Vignale and Rasolt [21], have discussed CDFT formulations in terms of spin-resolved densities $(\rho_\uparrow,\rho_\downarrow,\mathbf{j}_{\mathrm{p};\uparrow},\mathbf{j}_{\mathrm{p};\downarrow})$, we also remark that our 1RDM construction is easily modified for spin-resolved 1RDMs $D^{\uparrow\uparrow}$ and $D^{\downarrow\downarrow}$. The eigenvectors then correspond to natural spin orbitals with eigenvalues bounded by one rather than by two as in the case of natural spatial orbitals. The modifications to the above presentation are trivial—condition 4 in Theorem 1 becomes that no occupation is larger than 1, and the factors $\frac{1}{4}$ consequently disappear from the equations in Theorem 6 and Eq. (41).

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APPENDIX A: THE REGULAR REPRESENTATION OF TRACE-CLASS OPERATORS

The density matrix $D(\mathbf{r},\mathbf{s})$ is an element of $L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ and also the kernel of a trace-class operator over $L^2(\mathbb{R}^3)$. As such, it is not pointwise defined everywhere. At the same time, we wish to make sense of "the diagonal $D(\mathbf{r},\mathbf{r})$" in order to define the density in an unambiguous manner.

Brislawn [17] has presented a thorough study of trace-class operators and their kernels. The basic tools are found in this reference, but we restate some results for a self-contained treatment. We begin by clarifying some points concerning Lebesgue spaces that are often glossed over but are important here.

1. Lebesgue spaces

Let $X \subset \mathbb{R}^n$ be an open set. The $L^p(X)$ norm of a measurable function $f : X \to \mathbb{C}$ is defined by

$$\|f\|_p := \left(\int_X |f(x)|^p \, dx\right)^{1/p}. \tag{A1}$$

The vector space $\mathcal{L}^p(X)$ consists of all functions f such that $\|f\|_p < +\infty$. The space $\mathcal{L}^p(X)$ is not a normed space, since $\|f\|_p = 0$ if and only if $f(x) = 0$ for almost all $x \in X$ (rather than for all $x \in X$). On the other hand, the set $L^p(X)$ consisting of all *equivalence classes* $[f] = \{g \in \mathcal{L}^p(X) : \|f-g\|_p = 0\}$ is a normed space. It is customary to speak of a function f as an element of $L^p(X)$ even though, strictly speaking, it is a *representative* of $[f] \in L^p(X)$.

This distinction between f and $[f]$ is not merely academic: two pointwise defined wave functions Ψ and Φ describe the same physical state if and only if $\|\Psi - \Phi\|_2 = 0$. Thus, $[\Psi] \in L^2(\mathbb{R}^{3N})$ is the wave function. Similarly, a reduced density matrix $D \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ is not defined pointwise: its formal diagonal $D(\mathbf{r},\mathbf{r})$ may therefore be redefined without changing the physics. If $[\Psi] = [\Phi]$, then $D_\Psi = D_\Phi$ almost everywhere, but if D is *given* there is no *a priori* way to know how the pointwise values $D(\mathbf{r},\mathbf{s})$ are affected by modifying the wave function on a set of zero measure.

2. Locally integrable functions

A function $f \in L^p_{\text{loc}}(\mathbb{R}^n)$ if and only if $f \in L^p(K)$ for every compact measurable $K \subset \mathbb{R}^n$. We furthermore have $L^q_{\text{loc}} \subset L^p_{\text{loc}}$ for $q \geqslant p$, and

$$L^p(\mathbb{R}^n) \subset L^p_{\text{loc}}(\mathbb{R}^n) \subset L^1_{\text{loc}}(\mathbb{R}^n). \tag{A2}$$

Clearly, L^1_{loc} is a large class of functions, and functions in L^1_{loc} are said to be "locally integrable." We also need a slightly more general notion of local integrability, as follows:

Definition 2. Let $X \subset \mathbb{R}^n$, $Y \subset \mathbb{R}^m$ be open sets. The set $L^p(X_{\text{loc}} \times Y)$ is the set of (equivalence classes of) all measurable functions $u : X \times Y \to \mathbb{C}$ such that for all compact measurable $K \subset X$, $u \in L^p(K \times Y)$. A similar definition is made for arbitrary products and positions of the subscript "loc." In particular, $L^p_{\text{loc}}(X) = L^p(X_{\text{loc}})$.

3. The regular representation

The goal of this section is to establish a unique representative $\tilde{f}$ of $[f] \in L^1_{\text{loc}}$, called the regular representative of f. This representative will aid in defining the diagonal of $D \in \mathcal{D}_{N,1}$. The first step is to introduce the local averaging operator A_ϵ.

Definition 3: Local averaging operator A_ϵ. Let $f \in L^1_{\text{loc}}(\mathbb{R}^n)$ and $\epsilon > 0$. For a box $C_\epsilon = [-\epsilon,\epsilon]^n$ of Lebesgue measure $|C_\epsilon| = (2\epsilon)^n$, the (linear) local averaging operator $A_\epsilon : L^1_{\text{loc}} \to L^1_{\text{loc}}$ is defined by

$$A_\epsilon f(x) := \frac{1}{|C_\epsilon|} \int_{C_\epsilon} f(x+y)\, dy. \tag{A3}$$

Since C_ϵ is compact, $A_\epsilon f(x)$ is everywhere finite and is independent of the particular representative f of $[f]$ that appears in the integrand. It can be shown that $A_\epsilon f(x)$ is continuous both in x and in $\epsilon > 0$ [22]. We are here interested in the limit $\epsilon \to 0$ and therefore invoke the Lebesgue differentiation theorem.

Theorem 9: Lebesgue differentiation theorem. Let $f \in L^1_{\text{loc}}(\mathbb{R}^n)$. Then for almost all $x \in \mathbb{R}^n$,

$$\lim_{\epsilon \to 0} A_\epsilon f(x) = f(x). \tag{A4}$$

Proof. See Ref. [22]. ■

Since $A_\epsilon f(x)$ is independent of the particular $f \in [f]$, this limit determines a unique representative.

Definition 4: Regular representative. The regular representative $\tilde{f}$ of $f \in L^1_{\text{loc}}(\mathbb{R}^n)$ is defined by

$$\tilde{f}(x) := \lim_{\epsilon \to 0} A_\epsilon f(x) \tag{A5}$$

whenever the limit in Eq. (A4) exists.

Since $\tilde{f}(x) = f(x)$ almost everywhere, $\tilde{f}$ and f represent the same element $[f] \in L^1_{\text{loc}}$. Moreover, it is easy to see that $\tilde{f}$ is independent of the starting representative f and that the set of zero measure (where $\tilde{f}$ is undefined) is uniquely given by $[f] \in L^1_{\text{loc}}$. Intuitively, $\tilde{f}$ is more regular than f, "smoothing out" unnecessary discontinuities, and so on.

Related to the regular representative is the Hardy-Littlewood maximal function and associated inequality.

Definition 5: Hardy-Littlewood maximal function. For $f \in L^1_{\text{loc}}(\mathbb{R}^n)$ and $C_\epsilon = [-\epsilon,\epsilon]^n$ of Lebesgue measure $|C_\epsilon| = (2\epsilon)^n$, the Hardy-Littlewood maximal function Mf is defined by

$$Mf(x) := \sup_{\epsilon > 0} \frac{1}{|C_\epsilon|} \int_{C_\epsilon} |f(x+y)|\, dy. \tag{A6}$$

The following theorem is also called the maximal theorem.

Theorem 10: Hardy-Littlewood maximal inequality. If $f \in L^p(\mathbb{R}^n)$, then $Mf(x)$ is finite almost everywhere. Moreover, there exists a constant C_p (independent of f and n) such that

$$\|Mf\|_p \leqslant C_p \|f\|_p.$$

Proof. See Ref. [22]. ■

Since $|A_\epsilon f(x)| \leqslant Mf(x)$ for all x, we obtain as a corollary that A_ϵ is a bounded linear operator from L^p to L^p. Using this fact, it is straightforward to show that A_ϵ not only smoothes f, but also smoothes the mode of convergence:

Lemma 1. Suppose $f_n \to f$ in $L^p(X)$. For all $\epsilon > 0$, $A_\epsilon f_n \to A_\epsilon f$ uniformly [i.e., in $L^\infty(X)$].

Proof. We show that, for every $\epsilon > 0$, there exists a constant $K(\epsilon)$ such that, for all $f \in L^p$,

$$\|A_\epsilon f\|_\infty \leqslant K(\epsilon)\|f\|_p.$$

We have

$$|A_\epsilon f(x)| \leqslant \frac{1}{|C_\epsilon|} \|f\|_{L^1(x+C_\epsilon)}.$$

Since C_ϵ is bounded in $\mathbb{R}^n$,

$$\int_{x+C_\epsilon} 1 \times |g(x)|\, dx \leqslant |C_\epsilon|^{1/q} \|g\|_{L^p(x+C_\epsilon)}, \tag{A7}$$

where $1/q + 1/p = 1$. Thus,

$$|A_\epsilon f(x)| \leqslant |C_\epsilon|^{1/q-1} \|f\|_{L^p(\mathbb{R}^n)},$$

independent of x. ■

4. The diagonal of a factorized kernel

Based on our intuition, we may now hypothesize that, given an arbitrary reduced density matrix $D(\mathbf{r},\mathbf{s}) \in \mathcal{D}_{N,1}$, the diagonal of $\tilde{D}$ is the proper definition of the density:

$$\rho(\mathbf{r}) = \tilde{D}(\mathbf{r},\mathbf{r}). \tag{A8}$$

This is indeed true, as we show. To this end, a slight reformulation and generalization of Theorem 3.5 in Ref. [17] is useful for us. The reformulation states that, if an operator kernel is factorized, then the diagonal of the regular representative is given by the diagonal of the factorization, almost everywhere. The proof carries over with only trivial modifications, but since it is important, we rephrase it here.

Theorem 11: Diagonal of factorization. Let (X,dx) and (Y,dy) be open subsets of Euclidean spaces equipped with the standard Lebesgue measures. For $P \in L^2(X \times Y)$ and $Q \in L^2(Y \times X)$, let $C : X \times X \to \mathbb{C}$ be given by

$$C(x,x') = (P * Q)(x,x') = \int_Y P(x,y)Q(y,x')\, dy. \tag{A9}$$

Then $C \in L^2(X \times X)$ (a pointwise representative) and

$$\tilde{C}(x,x) = C(x,x) \tag{A10}$$

for almost all $x \in X$. Moreover, the map $x \mapsto C(x,x) = (P * Q)(x,x)$ belongs to $L^1(X)$.

Proof. We now demonstrate that $C \in L^2(X \times X)$. For almost all $x \in X$ and for almost all $x' \in X$, it holds that $P(x,\cdot), Q(\cdot,x') \in L^2(Y)$. From the Cauchy-Schwarz inequality, we obtain

$$|C(x,x')| \leqslant \int |P(x,y)||Q(y,x')|\,dy \leqslant \|P(x,\cdot)\|_{L^2(Y)}\|Q(\cdot,x')\|_{L^2(Y)} < +\infty \quad \text{(A11)}$$

for almost all x and almost all x' and hence also for almost all $(x,x') \in X \times X$. Squaring and integrating, we obtain $\|C\|^2_{L^2(X\times X)} \leqslant \|P\|^2_{L^2(X\times Y)}\|Q\|^2_{L^2(Y\times X)} < +\infty$.

Next, we demonstrate that the diagonal is in $L^1(X \times X)$. For $\epsilon > 0$, let $A_{\epsilon,i}P(x,y)$ be the averaging operator acting on the ith argument and let $M_i P(x,y)$ be the maximal operator acting on the ith argument. For almost all x,x',y, we then obtain

$$|A_{\epsilon,1}P(x,y)A_{\epsilon,2}Q(y,x')| \leqslant M_1P(x,y)M_2Q(y,x'). \quad \text{(A12)}$$

By the Cauchy-Schwarz inequality, we obtain

$$\int |M_1P(x,y)M_2Q(y,x')|^2 dy \leqslant \left(\int |M_1P(x,y)|^2\,dy\right)\left(\int |M_2Q(y,x')|^2 dy\right), \quad \text{(A13)}$$

where both factors on the right-hand side are finite by the maximal theorem, for almost all x and almost all x'. These bounds justify the use of Fubini's theorem to write

$$A_\epsilon C(x,x') = \frac{1}{|C_\epsilon|^2}\int_{C_\epsilon\times C_\epsilon\times Y} P(x+t,y)Q(y,x'+t')dtdt'dy = \int_Y A_{\epsilon,1}P(x,y)\,A_{\epsilon,2}Q(y,x')\,dy, \quad \text{(A14)}$$

which holds for almost every x and x'.

We now observe that, for each factor on the right-hand side,

$$\lim_{\epsilon\to 0} A_{\epsilon,1}P(x,y) = P(x,y) \quad \text{a.a. } x \in X, \quad \text{(A15)}$$

$$\lim_{\epsilon\to 0} A_{\epsilon,2}Q(y,x') = Q(y,x') \quad \text{a.a. } x' \in X. \quad \text{(A16)}$$

The dominated convergence theorem together with the bounds in Eqs. (A12) and (A13) now imply that we can take the limit in Eq. (A14) to get

$$\tilde{C}(x,x) = \lim_{\epsilon\to 0} A_\epsilon C(x,x) = \int_Y P(x,y)Q(y,x)\,dy \quad \text{(A17)}$$

for almost all x. We have

$$\int_X (P * Q)(x,x)\,dx = \langle \hat{Q}, P\rangle_{L^2(X\times Y)}, \quad \text{(A18)}$$

with $\hat{Q}(x,y) = Q^*(y,x)$. Being an inner product on L^2, this expression is finite, completing the proof. ■

Remark 1. Although the diagonal of $P * Q$ is in L^1, we cannot conclude that $P * Q$ is trace class; see Ref. [17] for a counterexample. On the other hand, if $X = Y$ in Theorem 11, then $P * Q$ is by definition trace class and it is also true that $\mathrm{Tr}\, P * Q = \int_X (\mathrm{diag}\, P * Q)(x)\,dx$.

Remark 2. $C = P * Q$ is the kernel of a Hilbert-Schmidt operator over $L^2(X)$. We see that it is meaningful to define the diagonal $\mathrm{diag}\, C$ of any Hilbert-Schmidt operator on an explicitly factorized form from the expression

$$[\mathrm{diag}\, P * Q](x) := (P * Q)(x,x), \quad \text{(A19)}$$

and the theorem states that this function belongs to $L^1(X)$, independent of the factorization.

Remark 3. As a corollary, if $P \in L^2(\mathbb{R}^n_{\mathrm{loc}} \times \mathbb{R}^m)$, $Q \in L^2(\mathbb{R}^m \times \mathbb{R}^n_{\mathrm{loc}})$, then $P * Q \in L^1_{\mathrm{loc}}(\mathbb{R}^n)$.

Remark 4. If $P(x,y) = Q^*(y,x)$, then $P * Q$ is positive semidefinite. Since $\mathrm{diag}\, P * Q$ is integrable, it follows from a theorem in Ref. [17] that $P * Q$ is trace class over $L^2(X)$.

APPENDIX B: SOME PROOFS FROM SECTION III

1. Proof of Theorem 2

For this proof, we use Theorem 11 in Appendix A.

Proof. Let $\Gamma \in \mathcal{D}_N$ be given. Assume that $D_\Gamma(\mathbf{r},\mathbf{s}) = D(\mathbf{r},\mathbf{s})$ almost everywhere in $\mathbb{R}^3 \times \mathbb{R}^3$. It follows that $\tilde{D}_\Gamma(\mathbf{r},\mathbf{r}) = \tilde{D}(\mathbf{r},\mathbf{r}) = (G^\dagger * G)(\mathbf{r},\mathbf{r})$ for almost all $\mathbf{r}$, since the regular representative is unique, and by using Theorem 11, with $P(\mathbf{r},\mathbf{s}) = G(\mathbf{s},\mathbf{r})^*$ and $Q(\mathbf{r},\mathbf{s}) = G(\mathbf{s},\mathbf{r})$ ($X = Y = \mathbb{R}^3$).

We need to show that $\rho_\Gamma(\mathbf{r}) = (G^\dagger * G)(\mathbf{r},\mathbf{r})$ for almost all $\mathbf{r}$. Assume that $\Gamma = |\Psi\rangle\langle\Psi|$. Now, $\rho_\Gamma(\mathbf{r}) = \rho_\Psi(\mathbf{r}) = D_\Gamma(\mathbf{r},\mathbf{r})$ for almost every $\mathbf{r}$, by definition of $\rho_\Psi(\mathbf{r})$. Applying Theorem 11 to $P(\mathbf{r},\mathbf{r}_{2:N}) = \Psi(\mathbf{r},\mathbf{r}_{2:N})$ and $Q(\mathbf{r}_{2:N},\mathbf{r}) = \Psi(\mathbf{r},\mathbf{r}_{2:N})^*$, $X = \mathbb{R}^3$ and $Y = \mathbb{R}^{3N-3}$, we see that $\rho_\Gamma(\mathbf{r}) = \tilde{D}_\Gamma(\mathbf{r},\mathbf{r}) = (G^\dagger * G)(\mathbf{r},\mathbf{r})$.

We invite the reader to fill in the details when Γ is a general mixed state. ■

2. Proof for Theorem 3

Proof. $2 \Rightarrow 1$: Let a $G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ be given such that $\nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3_{\mathrm{loc}})$. Then, for every compact $K \subset \mathbb{R}^3$,

$$T_\alpha(\mathbf{r},\mathbf{s}) := \frac{1}{2}[\partial_{2,\alpha}G]^\dagger * [\partial_{2,\alpha}G](\mathbf{r},\mathbf{s}) = \frac{1}{2}\int d\mathbf{u}\,\partial_{2,\alpha}G(\mathbf{u},\mathbf{r})^*\partial_{2,\alpha}G(\mathbf{u},\mathbf{s}) \quad \text{(B1)}$$

is in $L^2(K \times K)$ by Theorem 11. T_α is positive semidefinite, so by Remark 4 after Theorem 11, T_α is trace class over $L^2(K)$.

By the definition of the weak derivative and Fubini's theorem, we easily verify that in fact $T_\alpha = \frac{1}{2}\partial_{1,\alpha}\partial_{2,\alpha}D$ almost everywhere. Thus $\nabla_1 \cdot \nabla_2 D$ is trace class, and D has locally finite kinetic energy.

$1 \Rightarrow 2$: Since $D \in \mathcal{D}_{N,1}$ there exists a spectral decomposition

$$B(\mathbf{r},\mathbf{s}) = \sum_k \lambda_k \phi_k(\mathbf{r})\phi_k(\mathbf{s})^*, \quad \text{(B2)}$$

where $\{\phi_k\} \subset L^2(\mathbb{R}^3)$ is a complete, orthonormal set, and where $0 \leqslant \lambda_k \leqslant 2$ such that $\sum_k \lambda_k = N$. Of course $B(\mathbf{r},\mathbf{s}) = D(\mathbf{r},\mathbf{s})$ almost everywhere, but they may be pointwise different.

Let $K \subset \mathbb{R}^3$ be compact. Restricted to $K \times K$, $\nabla_1 \cdot \nabla_2 D = \nabla_1 \cdot \nabla_2 B$ (almost everywhere) is trace class, and we compute

$$\nabla_1 \cdot \nabla_2 B(\mathbf{r},\mathbf{s}) = \sum_k \lambda_k \nabla\phi_k(\mathbf{r}) \cdot \nabla\phi_k(\mathbf{s})^* \quad \text{a.e.} \quad \text{(B3)}$$

Let $A_k(\mathbf{r},\mathbf{s}) = \nabla\phi_k(\mathbf{r}) \cdot \nabla\phi_k(\mathbf{s})^*$. By assumption,

$$\mathrm{Tr}(\nabla_1 \cdot \nabla_2 B) = \sum_k \lambda_k \,\mathrm{Tr}\, A_k = \sum_k \lambda_k \|\nabla\phi_k\|^2_{L^2(K)} < +\infty, \tag{B4}$$

implying that $\nabla\phi_k \in L^2(K)$ for every K; hence, $\nabla\phi_k \in L^2_{\mathrm{loc}}(\mathbb{R}^3)$.

Let G be given by

$$G(\mathbf{r},\mathbf{s}) = \sum_k \lambda_k^{1/2} \phi_k(\mathbf{r})\phi_k(\mathbf{s})^*. \tag{B5}$$

Clearly, $G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ and $D = G^\dagger * G$. Moreover,

$$\nabla_2 G(\mathbf{r},\mathbf{s}) = \sum_k \lambda_k^{1/2} \phi_k(\mathbf{r})\nabla\phi_k(\mathbf{s})^*. \tag{B6}$$

Computing the $L^2(\mathbb{R}^3 \times K)$ norm,

$$\begin{aligned}\|\nabla_2 G\|^2 &= \sum_{k\ell} \lambda_k^{1/2}\lambda_\ell^{1/2} \langle\phi_\ell,\phi_k\rangle_{L^2(\mathbb{R}^3)} \langle\nabla\phi_k,\nabla\phi_\ell\rangle_{L^2(K)} \\ &= \sum_k \lambda_k \|\nabla\phi_k\|^2_{L^2(K)}.\end{aligned} \tag{B7}$$

$3 \Leftrightarrow 1$: Let Γ be such that $D_\Gamma = D$ almost everywhere. We have

$$D_\Gamma(\mathbf{r},\mathbf{s}) = \sum_i p_i \int d\mathbf{r}_{2:N} \Psi_i(\mathbf{r},\mathbf{r}_{2:N})\Psi_i(\mathbf{s},\mathbf{r}_{2:N})^*. \tag{B8}$$

Furthermore,

$$\begin{aligned}T_\alpha(\mathbf{r},\mathbf{s}) &:= \frac{1}{2}\partial_{1,\alpha}\partial_{2,\alpha} D(\mathbf{r},\mathbf{s}) \\ &= \frac{1}{2}\sum_i p_i \int d\mathbf{r}_{2:N} \partial_{1,\alpha}\Psi_i(\mathbf{r},\mathbf{r}_{2:N})\partial_{1,\alpha}\Psi_i(\mathbf{s},\mathbf{r}_{2:N})^*,\end{aligned} \tag{B9}$$

using the definition of the weak derivative and Fubini's theorem. By Theorem 11,

$$\tilde{T}_\alpha(\mathbf{r},\mathbf{r}) = \frac{1}{2}\sum_i p_i \int |\partial_{1,\alpha}\Psi_i(\mathbf{r},\mathbf{r}_{2:N})|^2 d\mathbf{r}_{2:N} \tag{B10}$$

for almost all $\mathbf{r}$. For any compact $K \subset \mathbb{R}^3$, integration yields

$$\int_K d\mathbf{r}\tilde{T}_\alpha(\mathbf{r},\mathbf{r}) = \frac{1}{2}\sum_i p_i \|\partial_{1,\alpha}\Psi_i\|^2_{L^2(K\times\mathbb{R}^{3N-3})}. \tag{B11}$$

Since T_α is positive semidefinite, the left-hand side is the trace of $\frac{1}{2}\partial_{1,\alpha}\partial_{2,\alpha}D$. Thus, D has locally finite kinetic energy if and only if any representing $\Gamma \mapsto D$ has locally finite kinetic energy. ■

3. Proof for Theorem 4

Proof. Most of the proof is similar that of Theorem 3, so we skip some details.

Let Γ be such that $D_\Gamma = D$ almost everywhere. The state Γ has a locally finite kinetic energy by Theorem 3. By a reasoning similar to that of the proof of this lemma, we obtain

$$c_{\Gamma,\alpha}(\mathbf{r}) = c_\alpha(\mathbf{r}) = [\mathrm{diag}(-\mathrm{i}\partial_{2,\alpha}G)^\dagger * G](\mathbf{r},\mathbf{r}) \tag{B12}$$

almost everywhere, independently of Γ. Squaring and integrating over a compact $K \subset \mathbb{R}^3$ and applying the Cauchy-Schwarz inequality, we obtain the bound

$$\int_K |c_\alpha(\mathbf{r})|^2 d\mathbf{r} \leqslant \|\partial_{2,\alpha}G\|^2_{L^2(\mathbb{R}^3\times K)} \|G\|^2_{L^2(\mathbb{R}^3\times K)} < +\infty. \tag{B13}$$

■

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