Fermion \(N\)-representability for prescribed density and paramagnetic current density

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The \(N\)-representability problem is the problem of determining whether or not 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 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 Weizsäcker-like canonical kinetic energy density is locally integrable), we prove that there exist 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.

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 or not 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, 3]. For a one-particle reduced density matrix (1-rdm), Slater-determinantal representability is equivalent to idempotency; in general, however, pure-state \(N\)-representability of 1-rdm’s is a difficult and largely unsolved problem [2, 3]. On the other hand, any 1-rdm 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 note, we report the solution to the mixed-state \(N\)-representability problem when both the density and paramagnetic current density are prescribed. More precisely, we answer the following question: given a density \(\rho(\mathbf{r})\) and a paramagnetic current density \(j_p(\mathbf{r})\), does there exist a mixed state \(\Gamma\) with the prescribed density and current density, written \(\Gamma \mapsto (\rho,j_p)\)? We answer this question affirmatively by constructing an explicit 1-rdm, from which the existence of the \(N\)-particle state follows.

We note that standard constructions demonstrating the Slater-determinantal \(N\)-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 reproduce both densities and currents. However, such solutions have limited scope, since the vorticity vanishes when orbitals give rise to the same density.

During the preparation of the present work, one of us (S. Kvaal) met E.H. Lieb during a trimester at Institut Henri Poincaré in Paris in July 2013, and became aware that together with R. Schrader, he had shown a Slater determinant representability result for \((\rho,j_p)\) and \(N \geq 4\). This work has now been published [11].

Clearly, \(N\)-representability via a Slater determinant implies representability via a mixed state. However, Lieb and Schrader’s result requires \(N \geq 4\), 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 \((\rho,j_p)\) 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 1-rdm. 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 Section [III] 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 Section [III] we construct in Sections [IV] and [IV C] a reduced density matrix for a prescribed density and paramagnetic current density. Section [V] contains some concluding remarks. Finally, two appendices 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 Section [III]. Appendix B contains proofs of theorems in Section [III].
II. BACKGROUND

The $N$-representability problem with prescribed density $\rho$ and paramagnetic current density $j_p$ arises in current-density functional theory (CDFT) [12]. In CDFT, a magnetic vector potential $A$, in addition to the scalar potential $v$, enters the (spin-free) N-electron Hamiltonian. In atomic units,

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

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

$$E[v, A] = \inf_{\Gamma} \text{Tr}(\Gamma H[v, A]) \quad (2)$$

where the minimization is over all mixed states $\Gamma$ with a finite canonical kinetic energy

$$T[\Gamma] := \frac{1}{2} \text{Tr}(\nabla \Gamma \nabla^\dagger). \quad (3)$$

Introducing the constrained-search universal functional

$$F[\rho, j_p] = \inf_{\Gamma \rightarrow \rho, j_p} \text{Tr}(\Gamma H[0, 0]), \quad (4)$$

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

$$E[v, A] = \inf_{\rho, j_p} \left( F[\rho, j_p] + \int \left( \rho \left( v + \frac{1}{2} A^2 + j_p \cdot A \right) \right) dr \right). \quad (5)$$

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

In Kohn–Sham theory, the idea is to express the densities in Eq. (5) in terms of a single Slater determinant of non-interacting particles and to approximate the kinetic-energy contributions to $F[\rho, j_p]$ by the non-interacting kinetic energy,

$$T_0[\rho, j_p] = \inf_{\{\phi_k\} \leftarrow \rho, j_p} \frac{1}{2} \sum_{k=1}^{N} \langle \nabla \phi_k, \nabla \phi_k \rangle, \quad (6)$$

where the infimum is over an orthonormal set of orbitals $\phi_k$ or, equivalently, the corresponding Slater determinants or idempotent 1-rdms. At this point, the Slater-determinantal $N$-representability problem arises.

In general, densities $\rho$ and $j_p$ arising from a single orbital have a vanishing paramagnetic vorticity,

$$\nu = \nabla \times \frac{j_p}{\rho} = 0, \quad (7)$$

except for possible Dirac-delta singularities at points $r$ where $\rho(r) = 0$. 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. [13–14] for work in this direction),

$$\bar{T}_s[\rho, j_p] = \inf_{\{\phi_k\} \rightarrow \rho, j_p} \frac{1}{2} \sum_{k=1}^{N} n_k \langle \nabla \phi_k, \nabla \phi_k \rangle, \quad (8)$$

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

For a mixed state $\Gamma \rightarrow (\rho, j_p)$, it is known that

$$T_W[\rho] + T_P[\rho, j_p] \leq T[\Gamma], \quad (9)$$

where the von Weizsäcker kinetic-energy functionals are given by

$$T_W[\rho] := \frac{1}{8} \int \rho(r)^{-1} \nabla \rho(r)^2 dr, \quad (10)$$

$$T_P[\rho, j_p] := \frac{1}{2} \int \rho(r)^{-1} j_p(r)^2 dr. \quad (11)$$

A necessary condition for a finite-kinetic-energy representability is therefore that $T_W[\rho] + T_P[\rho, j_p] < +\infty$. For the case $j_p = 0$, this is also a sufficient condition. It is of interest to know whether this sufficiency generalizes to $j_p \neq 0$. In this paper, we shall 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:

$$\Gamma(r_{1:N}, s_{1:N}) = \sum_i p_i \Psi_i(r_{1:N})\Psi_i^\dagger(s_{1:N}). \quad (12)$$

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

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

and is symmetric with respect to permutations $\pi$ of the coordinate labels, $(r_k, s_k) \overset{\pi}{\rightarrow} (r_{\pi(k)}, s_{\pi(k)})$. 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 $p_i \geq 0$ such that $\sum_i p_i = 1$. Moreover, each (spin-free) pure state is either totally symmetric or anti-symmetric. In what follows, it does not matter whether each pure state is required to be anti-symmetric, symmetric, or either anti-symmetric or symmetric with respect to index permutations $\pi$ of the spatial coordinates. For simplicity, we shall occasionally simplify the presentation by taking $\Gamma$ to be a pure state.
A. Density matrices

The 1-rdm belonging to a pointwise defined $\Gamma$ on the form \([12]\) is given by the convex combination

$$D_T(r, s) := N \sum_i p_i \Psi_i^T(r, r_{2:N}) \Psi_i^*(s, r_{2:N}), \quad dr_{2:N}$$ \quad (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 $D_{\Psi}$. Due to permutation symmetry, the 1-rdm is independent of which $N - 1$ coordinates that have been integrated out.

Given that $D_T \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$, $D_T$ 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, $\Gamma$ and $D_T$ 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

$$\text{Tr } A := \sum_k \langle \phi_k, A \phi_k \rangle$$ \quad (15)

has a finite value, independent of the orthonormal basis.

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

$$(B \ast C)(x, y) := \int_X B(x, z)C(z, y) \, dz,$$ \quad (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 $A = B \ast C$ with $B$ and $C$ Hilbert–Schmidt. (Indeed, this is often taken as an alternative definition of trace-class operators.) The trace is then given by \([17]\) the integral of the diagonal,

$$\text{Tr } A = \int_X (A \ast B)(x, x) \, dx.$$ \quad (17)

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

We denote by $D_N$ the set of mixed $N$-electron states $\Gamma$ and by $D_{N,1}$ the set of 1-rdm's that belong to some mixed $N$-electron state. The set $D_{N,1}$ has the following well-known characterization:

**Theorem III.1.** $D_{N,1}$ 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: $D(r, s) = D^*(s, r)$ for almost all $(r, s)$.
3. $D$ is positive semidefinite: $0 \leq \int \phi^*(r)D(r, s)\phi(s) \, ds$ for all $\phi \in L^2(\mathbb{R}^3)$
4. $D$ has no eigenvalues greater than two: $2 \geq \int \phi^*(r)D(r, s)\phi(s) \, ds$ for all $\phi \in L^2(\mathbb{R}^3)$
5. $D$ has eigenvalues that add up to $N$: $\text{Tr } D = N$.

**Proof.** See Ref. \([19]\), Section 2.6. \qed

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

Since $D \in D_{N,1}$ 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

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

which plays an important role in the following.

B. Density

We now define the density $\rho_{\Psi}$ associated with the wave function $\Psi$ as

$$\rho_{\Psi}(r) := D_{\Psi}(r, r) = N \int_{\mathbb{R}^{3N-3}} |\Psi(r, r_{2:N})|^2 \, dr_{2:N}.$$ \quad (19)

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

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 $D_{\Gamma} = D$ everywhere, only that $D_{\Gamma}$ and $D$ are equal as elements of $L^2(\mathbb{R}^3 \times \mathbb{R}^3)$. Consequently, $D_{\Gamma}$ 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 “$\rho_{\Gamma}(r) = D_{\Gamma}(r, r)$” for a state and density matrix related by $\Gamma \mapsto D$.

Suppose next that we are able to assign a diagonal $D$ to $D$ in some unambiguous way and let $\Gamma, \Gamma' \in D_N$ 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 $\rho_{\Gamma} = \rho_{\Gamma'} = \text{diag } D$ almost everywhere in $\mathbb{R}^3$? Intuitively, this should be so.

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

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

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

$$\rho_\Gamma(r) = (G^\dagger \ast G)(r, r)$$

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

Since the factorization of $D = G^\dagger \ast G$ does exist following the discussion in Section III.1A it is indeed meaningful to talk about “the density $\rho$ of $D$” without reference to a specific $\Gamma \mapsto D$:

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

In particular, it follows that

$$\text{Tr} D = \int \rho_D(r) dr. \quad (21)$$

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 $g = \partial_a f \in L^1_{\text{loc}}(X)$ if, for all smooth, compactly supported “test functions” $u \in C_c^\infty(X)$,

$$\int_X g(x) u(x) dx = - \int_X f(x) \partial_a u(x) dx. \quad (22)$$

Thus, the weak derivative acts just like the standard derivative $\partial f / \partial x_a$ 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 $c_\Psi$ of a state $\Psi$ as

$$c_\Psi(r) := N \int_{\mathbb{R}^{3N-3}} [i \nabla_r \Psi(r, r_{2:N})] \Psi^*(r, r_{2:N}) dr_{2:N}$$

$$= -i \nabla_r D_\Psi(r, s)|_{r=s}, \quad (23)$$

whose real part is the paramagnetic current density:

$$j_{p,\Psi}(r) = \text{Re} c_\Psi(r) \quad (24)$$

with an analogous definitions for a mixed state $\Gamma$. We note, however, that this definition may not make sense without additional assumptions on the wave function $\Psi$, 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 $\nabla_r D_\Psi(r, s)|_{r=s}$ is only defined pointwise almost everywhere and since integration may not commute with differentiation.

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

**Definition III.1** (Locally finite kinetic energy). We say that $D \in \mathcal{D}_{N,1}$ has a locally finite kinetic energy if the weak derivative $\nabla_1 \cdot \nabla_2 D$ 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 $\nabla_1 \Psi \in L^2(\mathbb{R}^{3N-3})$ for every compact $K \subset \mathbb{R}^3$—that is, $\nabla_1 \Psi \in L^2(\mathbb{R}^{3N-3})$. (See Appendix C.4.) A mixed state $\Gamma \in \mathcal{D}_N$ has a locally finite kinetic energy if $\sum_i |p_i|_3 \|\nabla_1 \Psi_i\|_{L^2(K \times \mathbb{R}^{3N-3})}$ 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 III.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 $D = G^\dagger \ast G$ (a.e.) with $G$ Hilbert–Schmidt and $\nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}_3^{\text{loc}})$.

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

$$\tau_D(r) := \frac{1}{2} \|\nabla_2 G(\cdot, r)\|_{L^2(\mathbb{R}^3)}^2$$

$$= \frac{1}{2} \int_{\mathbb{R}^3} [\nabla_2 G(u, r)]^* \cdot [\nabla_2 G(u, r)] du \quad (25)$$

which is finite almost everywhere. From the proof of the Theorem III.3 in Appendix B, it follows that $\tau_D$ is in fact the kinetic energy density of any $\Gamma \mapsto D$. We also see that $\tau_D \in L^1_{\text{loc}}(\mathbb{R}^3)$ 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 III.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 $D = G^\dagger \ast G$ and $\nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3_{\text{loc}})$. For each $\Gamma \in \mathcal{D}_N$ with $\Gamma \mapsto D$, it then holds that $c_\Gamma \in L^1_{\text{loc}}(\mathbb{R}^3)$ and that

$$c_\Gamma(r) = (\pm i \nabla_2 G^\dagger \ast G) \ (r, \ r) \ a.e.$$
D. Summary

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

**Theorem III.5.** Let \( D = G^4 \ast G \) with \( G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3) \) \( \in \mathcal{D}_{N,1}, \nabla_2 G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3_{\text{vec}}) \). For every \( \Gamma \in \mathcal{D} \) such that \( \Gamma \rightarrow D \), then holds that the density \( \rho_{\Gamma} = \rho \in L^1(\mathbb{R}^3) \), the momentum density \( \mathbf{c}_{\Gamma} = \mathbf{c} \in L^1_{\text{vec}}(\mathbb{R}^3) \), and the kinetic energy density \( \tau_{\Gamma} = \tau \in L^1_{\text{loc}}(\mathbb{R}^3) \) are given almost everywhere by the expressions

\[
\rho(r) = \int_{\mathbb{R}^3} G^*(u, r)G(u, s) \, du,
\]

\[
c(r) = \int_{\mathbb{R}^3} [-i\nabla_2 G^*(u, r)] G(u, s) \, du,
\]

\[
\tau(r) = \frac{1}{2} \int_{\mathbb{R}^3} [\nabla_2 G^*(u, r) \cdot [\nabla_2 G(u, s)]] \, du.
\]

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

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

\[
\rho(r) \geq 0, \quad \rho \in L^1(\mathbb{R}^3) \cap L^q(\mathbb{R}^3),
\]

for some \( q > 1 \). The latter condition amounts to

\[
N := \|\rho\|_1 = \int |\rho(r)| \, dr < +\infty,
\]

\[
\|\rho\|_q^q = \int |\rho(r)|^q \, dr < +\infty,
\]

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

\[
\mathbf{j}_p(r) = \frac{1}{2} \rho(r) \kappa(r).
\]

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

(a) \( \rho(r) = \rho_D(r) = (\text{diag} \, D)(r) \)

(b) \( \mathbf{j}_p(r) = \text{Re} \, \mathbf{c}_D(r) = \frac{-1}{2} (\text{diag} \, \nabla_1 D)(r) + \text{c.c.} \),

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

A. Factorized elements \( P_\lambda \) and \( Q_\mu \)

Our strategy is to construct explicitly factorized elements \( P_\lambda = G^4_{\lambda} \ast G_\lambda \) and \( Q_\mu = H^4_{\mu} \ast H_\mu \) 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 \( D_\mu = (P_\lambda + Q_\mu)/2 \) remains in \( \mathcal{D}_{N,1} \), also with a locally finite kinetic energy. The flexibility of having several independent factorized reduced density matrices \( P_\lambda \) and \( Q_\mu \) allows the convex combination to reproduce the desired current.

The two terms are defined by the factorized expressions

\[
P_\lambda(r, s) = \sqrt{\rho(r)\rho(s)} \int_{\mathbb{R}^3} g^*(u, r)g(u, s) \, du,
\]

\[
Q_\mu(r, s) = \sqrt{\rho(r)\rho(s)} \int_{\mathbb{R}^3} h^*(u, r)h(u, s) \, du,
\]

where

\[
g(u, v) = \frac{\sqrt{8\lambda^{3/4}}}{\pi^{3/4}} e^{-i\cdot(v \cdot \kappa)} e^{-2\lambda(u \cdot v)^2},
\]

\[
h(u, v) = \frac{\sqrt{8\mu^{3/4}}}{\pi^{3/4}} e^{i\cdot(u \cdot \kappa)} e^{-2\mu(u \cdot v)^2}.
\]

Clearly, these operators may be written in the form

\[
P_\lambda = G_\lambda^4 \ast G_\lambda, \quad G_\lambda(r, s) = g(r, s)\sqrt{\rho(s)},
\]

\[
Q_\mu = H_\mu^4 \ast H_\mu, \quad H_\mu(r, s) = h(r, s)\sqrt{\rho(s)},
\]

It is straightforward to verify that \( G_\lambda, H_\mu \in L^2(\mathbb{R}^3 \times \mathbb{R}^3) \). The integration over \( u \) may be performed analytically, yielding the alternative expressions

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

\[
Q_\mu(r, s) = \sqrt{\rho(r)\rho(s)} e^{-\mu|\kappa(r) - \kappa(s)|^2} \times e^{-\frac{i}{2} (r \cdot s - \kappa(r) - \kappa(s))} - |\kappa(r) - \kappa(s)|^2/16\mu.
\]

These operators were found by making the initial ansatz \( \phi(r) = \sqrt{\rho(r)} e^{i\mathbf{r} \cdot \mathbf{k}(r)} \) 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 \( e^{i\mathbf{r} \cdot \mathbf{k}(s)} \). Since the elements of \( \mathcal{D}_{N,1} \) and their properties are conveniently described if an explicit factorization is available (see Theorem III.3), Gaussian kernels are suitable since since they allow mixed phase factors of the type \( e^{i\mathbf{r} \cdot \mathbf{k}(s)} \) to survive the integration.

B. The density of \( P_\lambda \) and \( Q_\mu \)

We now need to verify that \( P_\lambda \) and \( Q_\mu \) are elements of \( \mathcal{D}_{N,1} \) by checking points (1)–(5) of Theorem III.1.

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

\[
\lambda, \mu \geq \frac{2p}{\pi} \left( \frac{4}{3} N \|\rho\|_q \right)^{2p/3}
\]
where $1/p + 1/q = 1$. Then $P_\lambda$ and $Q_\mu$ in Eqs. 30–33 are elements of $\mathcal{D}_{N,1}$, with

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

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 semi-definite. From the expressions in Eqs. (36) and (37),

$$\text{tr} P_\lambda = \text{tr} Q_\lambda = \int \rho(r) dr = N.$$

It remains to compute a bound on the largest eigenvalues, demonstrating point (4) of Theorem 11 for the corresponding parameter values $\lambda$ and $\mu$. For an arbitrary normalized orbital,

$$n^2 \leq \left| \int \phi^*(r) P_\lambda(r,s) \phi(s) dr ds \right|^2 \leq \left( \int |\phi(r)| \sqrt{\rho(r) \rho(s)} e^{-\lambda |r-s|^2} |\phi(s)| ds \right)^2 \leq \left( \int |\phi(r)| \sqrt{\rho(r)} \left( \int \sqrt{\rho(s)} e^{-\lambda |r-s|^2} |\phi(s)| ds \right) dr \right)^2. \quad (38)$$

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

$$n^2 \leq \left( \int |\phi(r)|^2 dr \right) \times \left( \int \rho(r) \left( \int \sqrt{\rho(s)} e^{-2\lambda |r-s|^2} |\phi(s)| ds \right)^2 dr \right) \leq \int \rho(r) \left( \int |\phi(s)|^2 ds \int \rho(s) e^{-2\lambda |r-s|^2} ds \right) dr \leq \int \rho(r) \left( \sup_e \int \rho(s) e^{-2\lambda |e-s|^2} ds \right) dr \leq N \sup_e \int \rho(s) e^{-2\lambda |e-s|^2} ds \quad (39)$$

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

$$n^2 \leq N \|\rho\|_q \sup_e \|e^{-2\lambda |e-s|^2}\|_p = N \|\rho\|_q \left( \frac{\pi}{2\rho^2} \right)^{3/2p} \quad (40)$$

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

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

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

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

### C. The canonical kinetic energy of $P_\lambda$ and $Q_\lambda$

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

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

and

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

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

The kinetic energy density of $P_\lambda$ is

$$\tau_P(r) = \frac{1}{2} \|\nabla G(\cdot, r)\|_{L^2(\mathbb{R}^3)}^2 = \frac{1}{2} \nabla_r \cdot \nabla_s P_\lambda(r,s) \big|_{s=r} = \frac{\|
abla_\mathbf{r} \cdot \nabla_\mathbf{s} P_\lambda(r,s)\|_{L^2(\mathbb{R}^3)}}{8\rho(r)} + \frac{1}{2} (\nabla_\mathbf{r} \cdot \mathbf{k}(\mathbf{r}))^2 \rho(r) + \lambda \rho(r). \quad (44)$$

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

$$\tau_Q(r) = \frac{1}{2} \nabla_\mathbf{r} \cdot \nabla_\mathbf{s} Q_\mu(r,s) \big|_{s=r} = \left| \nabla_\mathbf{r} \cdot \mathbf{k}(\mathbf{r}) \right|^2 \rho(r) + \mu \rho(r).$$
The total kinetic energy density becomes

\[
\tau_D(r) = \frac{\lvert \nabla \rho(r) \rvert^2}{8\rho(r)} + \frac{1}{4} \sum_{\alpha=1}^{3} \left[ \left( \frac{\partial}{\partial r_\alpha} r \cdot \kappa(r) \right)^2 + \left( r \cdot \frac{\partial \kappa(r)}{\partial r_\alpha} \right)^2 \right] \rho(r) + \frac{1}{2}(\lambda + \mu)\rho(r),
\]

where we have used the fact that the kinetic energy density is linear in the density matrix. Using the special form of Young’s inequality, with \(a = \kappa_\alpha(r)\) and \(b = r \cdot \frac{\partial \kappa(r)}{\partial r_\alpha}\), we find that

\[
\left| \frac{\partial}{\partial r_\alpha} r \cdot \kappa(r) \right|^2 = |\kappa_\alpha(r) + r \cdot \frac{\partial \kappa(r)}{\partial r_\alpha}|^2 
\leq 2|\kappa_\alpha(r)|^2 + 2\left| r \cdot \frac{\partial \kappa(r)}{\partial r_\alpha} \right|^2.
\]

Hence, the canonical kinetic energy density is bounded by

\[
\tau_D(r) \leq \frac{\lvert \nabla \rho(r) \rvert^2}{8\rho(r)} + \frac{1}{2} \left[ |\kappa(r)|^2 + \frac{2}{3} \left( \frac{\partial \kappa(r)}{\partial r_\alpha} \right)^2 \right]/\sum_{\beta=1}^{3} \left( \frac{\partial \kappa_\beta(r)}{\partial r_\alpha} \right)^2 \rho(r) + \frac{1}{2}(\lambda + \mu)\rho(r)
\]

where the second inequality was obtained by using \(|r_{i,j}| \leq |r|\). A finite canonical kinetic energy of \(P_\lambda, Q_\mu\) and \(D_{\lambda\mu}\) is ensured if

\[
T_W[\rho] = \int \frac{\lvert \nabla \rho \rvert^2}{8\rho} \, dr = \frac{1}{2} \int \lvert \nabla \sqrt{\rho} \rvert^2 \, dr < \infty,
\]

\[
T_p[\rho, \mathbf{j}_\rho] = \int \frac{\lvert \mathbf{j}_\rho \rvert^2}{2\rho} \, dr = \frac{1}{8} \int \rho \kappa^2 \, dr < \infty,
\]

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

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

\[
\int (1 + r^2)\rho \kappa^2 \, dr < \infty.
\]

We have thus proved the following result:

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

\[
\frac{1}{2} \text{Tr}(\nabla_1 \cdot \nabla_2 D) \leq T_W[\rho] + 4T_p[\rho, \frac{1}{2}\rho \kappa] + \frac{1}{2}(\mu + \nu)N + \int \rho(1 + r^2)\rho \kappa^2 \, dr < \infty.
\]

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

**D. Lifting the global integrability condition**

Theorem IV.2 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 \(\rho\) and \(\kappa\) are then seen to be reproducible, albeit with merely a locally finite kinetic energy.

**Theorem IV.3.** Let \(\rho\) and \(\kappa\) be given such that \(\rho \geq 0\), \(\rho \in L^1_{\text{loc}}(\mathbb{R}^3)\), \(\rho \kappa_\alpha^2 \in L^1_{\text{loc}}(\mathbb{R}^3)\), \((1 + r^2)\rho(\partial \kappa_\beta/\partial r_\alpha)^2 \in L^1_{\text{loc}}(\mathbb{R}^3)\) for all Cartesian components \(\alpha, \beta \in \{1, 2, 3\}\). Then there exist a 1-rdm \(D\) with density \(\rho\) and current \(\mathbf{j}_\rho = \frac{1}{2}\rho \kappa\).
V. DISCUSSION

We have provided an explicit construction of a 1-rdm 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 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 $T_p[\rho, J_p]$ is not gauge invariant; on the contrary, it can make to be become infinite by applying a gauge transformation $J_p \mapsto J_p + \rho \nabla \chi$ with a rapidly growing gauge function $\chi$. Our results establish that such gauge transformations do not affect $N$-representability, as long as $\chi$ exhibits some minimal regularity.

The explicit constructions of density matrices can be used to provide orbital-free upper bounds on the canonical kinetic energy $T_r[\rho, J_p]$ for an extended Kohn–Sham formalism. Combining the above results with the standard lower bound $T_W + T_p$ on the kinetic energy, we get the following orbital-free bounds on the extended Kohn–Sham kinetic energy,

$$T_W + T_p \leq \overline{T}[\rho, J_p] \leq T_W + 4T_p + (\lambda + \mu)N$$
$$+ \int \rho(r) \left( \frac{|r|^2}{\lambda} + \frac{1}{4\lambda} \right) |\nabla a^{\alpha\beta}(r)|^2 \, dr. \quad (52)$$

Noting that several authors, following Vignale and Rauolf [21], have discussed CDFT formulations in terms of spin-resolved densities $(\rho_\uparrow, \rho_\downarrow, J_{p\uparrow}, J_{p\downarrow})$, we also remark that our 1-rdm construction is easily modified for spin-resolved 1-rdms $D^{\uparrow\uparrow}$ 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 III.1 becomes that no occupation is larger than 1, and the factors $1/4$ consequently disappear from Eqs. (IV.1) and (III.1).

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Appendix A: The regular representation of trace-class operators

The density matrix $D(r, s)$ 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 $D(r, r)$” 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

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

The vector space $L^p(X)$ consists of all functions $f$ such that $\|f\|_p < +\infty$. The space $L^0(X)$ 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 $[f] = \{g \in L^p(X) : \|f - g\|_p = 0\}$ 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 $\Psi$ and $\Phi$ describe the same physical state if and only if $\|\Psi - \Phi\|_2 = 0$. Thus, $[\Psi] \in L^2(\mathbb{R}^3)$ is the wave function. Similarly, a reduced density matrix $D \in L^2(\mathbb{R}^3)$ is not defined pointwise: its formal diagonal $D(r, r)$ 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 $D(r,s)$ 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 $L^p_{\text{loc}} \subset L^q_{\text{loc}}$ for $q > p$, and

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

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

Definition A.1. 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 $u : X \times Y \rightarrow \mathbb{C}$ 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 $\hat{f}$ 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 $A_{\epsilon}$:

Definition A.2 (Local averaging operator $A_{\epsilon}$). 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 $A_{\epsilon} : L^1_{\text{loc}} \rightarrow L^1_{\text{loc}}$ is defined by

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

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

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

$$\lim_{\epsilon \rightarrow 0} A_{\epsilon}f(x) = f(x).$$ (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 A.3 (Regular representative). The regular representative $\hat{f}$ of $f \in L^1_{\text{loc}}(\mathbb{R}^n)$ is defined by

$$\hat{f}(x) := \lim_{\epsilon \rightarrow 0} A_{\epsilon}f(x)$$ (A5)

whenever the limit in Eq. (A4) exists.

Since $\hat{f}(x) = f(x)$ almost everywhere, $\hat{f}$ and $f$ represent the same element $[f] \in L^1_{\text{loc}}$. Moreover, it is easy to see that $\hat{f}$ 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, $\hat{f}$ 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 A.4 (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

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

The following theorem is also called the Maximal Theorem:

Theorem A.2 (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

$$\|Mf\|_p \leq C_p \|f\|_p.$$ Proof. See Ref. [22].

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

Lemma A.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 $f \in L^p$,

$$\|A_{\epsilon}f\|_\infty \leq K(\epsilon) \|f\|_p.$$ We have

$$|A_{\epsilon}f(x)| \leq \frac{1}{|C_{\epsilon}|} \|f\|_{L^1(x + C_{\epsilon})}.$$ (A7)

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

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

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

$$|A_{\epsilon}f(x)| \leq |C_{\epsilon}|^{1/q - 1} \|f\|_{L^p(\mathbb{R}^n)},$$ 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 \( D(r,s) \) in \( \mathcal{D}_{N,1} \), the diagonal of \( D \) is the proper definition of the density:

\[
\rho(r) = \hat{D}(r,r).
\]

(A8)

This is indeed true, as we shall 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 A.3** (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

\[
C(x, x') = (P * Q)(x, x') = \int P(x, y)Q(y, x') dy.
\]

(A9)

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

\[
\hat{C}(x, x) = C(x, x)
\]

(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 \( P(x,)Q(,)x' \in L^2(Y) \). From the Cauchy–Schwarz inequality, we obtain

\[
|C(x, x')| \leq \int |P(x, y)Q(y, x')| dy \leq \|P(x, \cdot)||Q(\cdot, x')\|_{L^2(Y)} < +\infty
\]

(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 \( \|C\|^2_{L^2(X \times X)} \leq \|P\|^2_{L^2(X \times Y)} \|Q\|^2_{L^2(Y \times X)} < +\infty \).

Next, we demonstrate that the diagonal is in \( L^1(X \times X) \). For \( \epsilon > 0 \), let \( \epsilon A_{\epsilon, 1}P(x, y) \) be the averaging operator acting on the \( \epsilon \)th argument and let \( M_1P(x, y) \) be the maximal operator acting on the \( \epsilon \)th argument. For almost all \( x, x', y \), we then obtain

\[
|A_{\epsilon, 1}P(x, y)A_{\epsilon, 2}Q(y, x')| \leq M_1P(x, y)M_2Q(y, x'),
\]

(A12)

By the Cauchy–Schwarz inequality, we obtain

\[
\int |M_1P(x, y)M_2Q(y, x')|^2 dy \leq \left( \int |M_1P(x, y)|^2 dy \right) \left( \int |M_2Q(y, x')|^2 dy \right)
\]

(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

\[
A_{\epsilon, 1}C(x, x') = \frac{1}{|C_\epsilon|} \int P(x + t, y)Q(y, x' + t') dt'dy = \int Y M_1P(x, y)A_{\epsilon, 2}Q(y, x') dy,
\]

(A14)

which holds for almost every \( x \) and \( x' \).

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

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

(A15)

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

(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

\[
\hat{C}(x, x) = \lim_{\epsilon \to 0} A_{\epsilon}C(x, x) = \int Y P(x, y)Q(y, x) dy
\]

(A17)

for almost all \( x \). We have

\[
\int X (P * Q)(x, x) dx = \langle \hat{Q}, P \rangle_{L^2(X \times Y)},
\]

(A18)

with \( \hat{Q}(y, x) = Q^*(y, x) \). 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 A.3, then \( P * Q \) is by definition trace class and it is also true that \( \text{Tr} \ P * Q = \int_X (\text{diag} \ P * Q)(x) dx \).

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 \( C \) of any Hilbert–Schmidt operator on an explicitly factorized form from the expression

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

(A19)

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

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

Remark 4: If \( P(x, y) = Q^*(y, x) \), then \( P * Q \) is positive semidefinite. Since 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 III.2

For this proof, we use Theorem A.3 in Appendix A.
Proof. Let $\Gamma \in \mathcal{D}_N$ be given. Assume that that $D_T(r, s) = D(r, s)$ almost everywhere in $\mathbb{R}^3 \times \mathbb{R}^3$. It follows that $D_T(r, r) = D(r, r) = (G^\dagger \ast G)(r, r)$ for almost all $r$, since the regular representative is unique, and by using Theorem A.3 with $P(r, s) = G(s, r)^*$ and $Q(r, s) = G(s, r)$ ($X = Y = \mathbb{R}^3$).

We need to show that $\rho_\tau(r) = (G^\dagger \ast G)(r, r)$ for almost all $r$. Assume that $\Gamma = [\Psi] \langle \psi \rangle$. Now, $\rho_\tau(r) = \rho_\Psi(r) = D_T(r, r)$ for almost every $r$, by definition of $\rho_\Psi(r)$. Applying Theorem A.3 to $P(r, r; N) = \Psi(r, r; N)$ and $Q(r; N) = \Psi(r; N)^*$, $X = \mathbb{R}^3$ and $Y = \mathbb{R}^{3N-3}$, we see that $\rho_\tau(r) = D_T(r, r) = (G^\dagger \ast G)(r, r)$.

We invite the reader to fill in the details when $\Gamma$ is a general mixed state. $\square$

2. Proof of Theorem III.3

Proof. $2 \Rightarrow 1$: Let a $G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$ be given such that $\nabla G \in L^2(\mathbb{R}^3 \times \mathbb{R}^3)$.

Then, for every compact $K \subset \mathbb{R}^3$,

$$T_\alpha(r, s) := \frac{1}{2} [\partial_{\alpha, \alpha} G] \ast [\partial_{\alpha, \alpha} G](r, s)$$

$$= \frac{1}{2} \int du \partial_{\alpha, \alpha} G(u, r) \ast \partial_{\alpha, \alpha} G(u, s) \quad (B1)$$

is in $L^2(K \times K)$ by Theorem A.3. $T_\alpha$ is positive semidefinite, so by Remark 4 after Theorem A.3 $T_\tau$ is trace class over $L^2(K)$.

By the definition of the weak derivative and Fubini’s Theorem, we easily verify that in fact $T_\alpha = \rho_{\tau, \alpha} \partial_{\alpha, \alpha} D$ 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

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

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

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

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

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

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

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

Let $G$ be given by

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

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

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

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

$$\| \nabla_2 G \|^2 = \sum_{k, \ell} \lambda_k^{1/2} \lambda_\ell^{1/2} \langle \phi_k, \phi_\ell \rangle_{L^2(\mathbb{R}^3)} \langle \nabla \phi_k, \nabla \phi_\ell \rangle_{L^2(K)}$$

$$= \sum_k \lambda_k \| \nabla \phi_k \|^2_{L^2(K)}. \quad (B7)$$

3 $\Rightarrow 1$:

Let $\Gamma$ be such that $D_T = D$ a.e. We have,

$$D_T(r, s) = \sum_i p_i \int dr_N \Psi_i(r, r; N) \Psi_i(s, r; N)^*. \quad (B8)$$

Furthermore,

$$T_\tau(r, s) := \frac{1}{2} \partial_{\alpha, \alpha} \partial_{\alpha, \alpha} D(r, s)$$

$$= \frac{1}{2} \sum_i p_i \int dr_N \partial_{\alpha, \alpha} \Psi_i(r, r; N) \partial_{\alpha, \alpha} \Psi_i(s, r; N)^*, \quad (B9)$$

using the definition of the weak derivative and Fubini’s theorem. By Theorem A.3,

$$\tilde{T}_\tau(r, r) = \frac{1}{2} \sum_i p_i \int |\partial_{\alpha, \alpha} \Psi_i(r, r; N)|^2 dr_N \quad (B10)$$

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

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

Since $T_\tau$ is positive semidefinite, the left hand side is the trace of $\sum_i \partial_{\alpha, \alpha} \partial_{\alpha, \alpha} D$. Thus, $D$ has locally finite kinetic energy if and only if any representing $\Gamma \mapsto D$ has locally finite kinetic energy. $\square$

3. Proof of Theorem III.4

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

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

$$c_{\tau, \alpha}(r) = c_{\alpha}(r) = |\text{diag}(-i \partial_{\alpha, \alpha} G)| \ast G(r, r) \quad (B12)$$

almost everywhere, independently of $\Gamma$. Taking the absolute value, integrating over a compact $K \subset \mathbb{R}^3$ and
applying the Cauchy–Schwarz inequality, we obtain the bound

\[
\int_K |c_\alpha(r)| dr \leq \| \partial_{2,\alpha} G \|_{L^2(\mathbb{R}^3 \times K)} \| G \|_{L^2(\mathbb{R}^3 \times K)} < +\infty.
\]

\[\text{(B13)}\]

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