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Letter

Lower Semicontinuity of the Universal Functional in Paramagnetic Current–Density Functional Theory

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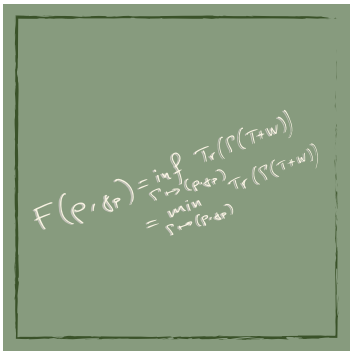
ABSTRACT: A cornerstone of current–density functional theory (CDFT) in its paramagnetic formulation is proven. After a brief outline of the mathematical structure of CDFT, the lower semicontinuity and expectation-valuedness of the CDFT constrained-search functional is proven, meaning that there is always a minimizing density matrix in the CDFT constrained-search universal density functional. These results place the mathematical framework of CDFT on the same footing as that of standard DFT.



Density functional theory (DFT) is at present the most widely used tool for first-principles electronic structure calculations in solid-state physics and quantum chemistry. DFT was put on a solid mathematical ground by Lieb in a landmark paper[1] from 1983, where he introduced the universal density functional $F(\rho)$ as the convex conjugate to the concave ground-state energy $E(v)$ for an electronic system in the external scalar potential v.

For electronic systems under the influence of a classical external magnetic potential $\mathbf{A}$, current–density functional theory (CDFT) was introduced by Vignale and Rasolt in 1987.[2] In addition to the density ρ, the paramagnetic current density $\mathbf{j}_p$ becomes a basic variable. The mathematical foundation of CDFT was put in place by Tellgren et al.[3] and Laestadius[4,5] in the 2010s on the basis of Lieb's treatment of the field-free standard case. However, a central piece of the puzzle has been missing—namely, whether the CDFT constrained-search functional $F(\rho, \mathbf{j}_p)$ is lower-semicontinuous and expectation-valued,[6] i.e., whether the infimum in its definition (see eq 3 below) is in fact attained. These foundational issues are important because CDFT is the natural extension of DFT to treat general magnetic systems and several numerical implementations have been reported, although the development of practical functionals lags behind standard DFT.[7−13]

In this Letter, we provide proofs of the above assertions. The CDFT constrained-search functional is indeed convex lower-semicontinuous and can therefore be identified with the CDFT Lieb functional—that is, the Legendre–Fenchel transform of the energy. *Without* this fact, the ground-state energy functional $E(v, \mathbf{A})$ and the constrained-search functional $F(\rho, \mathbf{j}_p)$ contain *different* information. If $F(\rho, \mathbf{j}_p)$ were not expectation-valued, one would lose the interpretation of the universal functional as intrinsic energy, which is very useful in standard DFT. For the interested reader, suggested further reading for convex analysis are van Tiel's excellent introductory text[14] and the monograph by Ekeland and Témam.[15] Also, the monograph by Barbu and Precupanu,[16] which treats convex analysis in Banach spaces, and the one by Bauschke and Combettes,[17] which focuses on the Hilbert space formulation, are highly recommended. For more details on trace-class operators, the monograph by Weidmann is an accessible starting point,[18] as well as the now classic volume by Reed and Simon.[19]

For an N-electron system in sufficiently regular external potentials v and $\mathbf{A}$, the ground-state energy is given by the Rayleigh–Ritz variation principle as

$$E(v, \mathbf{A}) = \inf_{\Gamma} \mathrm{Tr}(\Gamma H(v, \mathbf{A})) \tag{1}$$

where $H(v, \mathbf{A}) = T(\mathbf{A}) + W + \sum_{i=1}^{N} v(\mathbf{r}_i)$ is the electronic Hamiltonian with kinetic energy operator $T(\mathbf{A}) = \frac{1}{2}\sum_{i=1}^{N}[-\nabla_i + \mathbf{A}(\mathbf{r}_i)]^2$ and two-electron repulsion operator W. The

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minimization is over all N-electron density matrices Γ of finite kinetic energy, for which the one-electron density is $\rho \in X_L = L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3)$ and an element $\mathbf{j}_p \in X_p = \mathbf{L}^1(\mathbb{R}^3) \cap \mathbf{L}^{3/2}(\mathbb{R}^3)$.[20] (The boldface notation indicates a space of vector fields.) The external potential energy $(v|\rho) = \int_{\mathbb{R}^3} v(\mathbf{r})\rho(\mathbf{r})\,d\mathbf{r}$, the paramagnetic and diamagnetic terms $\frac{1}{2}(|\mathbf{A}|^2|\rho)$ and $(\mathbf{A}|\mathbf{j}_p) = \int_{\mathbb{R}^3} \mathbf{A}(\mathbf{r})\cdot\mathbf{j}_p(\mathbf{r})\,d\mathbf{r}$ and thus the Hamiltonian $H(v, \mathbf{A})$ are well-defined for any $v \in X_L' = L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ and $\mathbf{A} \in X_p' = \mathbf{L}^3(\mathbb{R}^3) + \mathbf{L}^\infty(\mathbb{R}^3)$, where X_L' and X_p' are the dual spaces of X_L and X_p, respectively. Examples of such potentials are the nuclear Coulomb potentials and uniform magnetic fields inside bounded domains. The symbol X_L for the space of densities is so chosen to indicate that it is the density space of Lieb's analysis, while X_p indicates "paramagnetic" current densities.

By a well-known reformulation of eq 1, we obtain the CDFT Hohenberg–Kohn variation principle:

$$E(v, \mathbf{A}) = \inf_{(\rho,\mathbf{j}_p)\in X_L\times X_p} \left\{ F(\rho, \mathbf{j}_p) + \left(v + \frac{1}{2}|\mathbf{A}|^2 \middle| \rho\right) + (\mathbf{A}|\mathbf{j}_p) \right\} \tag{2}$$

Here the Vignale–Rasolt constrained-search density functional F: $X_L \times X_p \to [0, +\infty]$ is defined by

$$F(\rho, \mathbf{j}_p) = \inf_{\Gamma\to(\rho,\mathbf{j}_p)} \mathrm{Tr}(\Gamma H_0) \tag{3}$$

where $H_0 = T(\mathbf{0}) + W$ is the intrinsic electronic Hamiltonian, and $\Gamma \to (\rho, \mathbf{j}_p)$ means that the infimum is taken over all N-electron density matrices Γ with density–current pair $(\rho, \mathbf{j}_p) \in L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3)$. Thus, if $(\rho, \mathbf{j}_p)$ is not N-representable, we have $F(\rho, \mathbf{j}_p) = +\infty$. The universal density functional F is the central quantity in any flavor of DFT, and its mathematical properties and approximation are of utmost importance to the field.

Although E in eq 2 is not concave, it is readily seen that the reparametrized energy

$$\tilde{E}(u, \mathbf{A}) = E\left(u - \frac{1}{2}|\mathbf{A}|^2, \mathbf{A}\right) \tag{4}$$

is concave. This reparametrization relies on a technical notion of *compatibility* of function spaces for the scalar and vector potentials,[20] which is satisfied for the potentials we consider here.

From the concavity and upper semicontinuity of the modified ground-state energy $\tilde{E}$, one can deduce the existence of an alternative universal density functional $\hat{F}$: $X_L \times X_p \to [0, +\infty]$ that is related to the ground-state energy by Legendre–Fenchel transformations in the manner

$$\tilde{E}(u, \mathbf{A}) = \inf_{(\rho,\mathbf{j}_p)} \{\hat{F}(\rho, \mathbf{j}_p) + (u|\rho) + (\mathbf{A}|\mathbf{j}_p)\} \tag{5}$$

$$\hat{F}(\rho, \mathbf{j}_p) = \sup_{(u,\mathbf{A})} \{\tilde{E}(u, \mathbf{A}) - (u|\rho) - (\mathbf{A}|\mathbf{j}_p)\} \tag{6}$$

where the optimizations are over the space $X_L \times X_p$ and its dual $X_L' \times X_p'$, respectively. As a Legendre–Fenchel transform, the functional $\hat{F}$ is convex and lower-semicontinuous. In this formulation of CDFT, the ground-state energy $\tilde{E}$ and the universal density functional $\hat{F}$ contain precisely *the same information*: each functional can be obtained from the other and therefore contains the same of the information about ground-state electronic systems in external scalar and vector fields.

From a comparison of the Hohenberg–Kohn variation principles in eqs 2 and 5, it is tempting to conclude that $\hat{F}$ and F are the same functional (i.e., $\hat{F} = F$), producing the same ground-state energy for each $(v, \mathbf{A})$. However, there exist infinitely many functionals $\tilde{F}$: $X_L \times X_p \to [0, +\infty]$ that give the correct ground-state energy $E(v, \mathbf{A})$ (but not necessarily the same minimizing densities, if any) for each $(v, \mathbf{A})$ in the Hohenberg–Kohn variation principle. Each such $\tilde{F}$ is said to be an *admissible* density functional.[6] Among these, the functional $\hat{F}$ stands out as being the only lower-semicontinuous and convex universal density functional and a lower bound to all other admissible density functionals (i.e., $\hat{F} \le \tilde{F}$). The functional $\hat{F}$, the closed convex hull of all admissible density functionals, is thus the most well-behaved admissible density functional. Indeed, we may view it as a regularization of all admissible density functionals, known as the Γ regularization in convex analysis. (This name is unrelated to our notation of density matrices.)

A fundamental result of Lieb's analysis of DFT is the identification of the transparent constrained-search density functional with the mathematically well-behaved closed convex hull $\hat{F}$. The identification follows since F is convex and lower-semicontinuous. Whereas convexity follows easily for the CDFT Vignale–Rasolt functional F, the proof of lower semicontinuity is nontrivial. For standard DFT it is given in ref 1, and for CDFT it is provided in the present Letter.

We simplify our analysis by merely assuming that the density–current pairs are $(\rho,\mathbf{j}_p) \in L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3) = [L^1(\mathbb{R}^3)]^4$, which we denote as X. With this topology, the potentials must be taken to be bounded functions, $(v, \mathbf{A}) \in X' = L^\infty(\mathbb{R}^3) \times \mathbf{L}^\infty(\mathbb{R}^3) = [L^\infty(\mathbb{R}^3)]^4$. This simplification is irrelevant in this context: if F can be shown to be lower-semicontinuous in the $[L^1(\mathbb{R}^3)]^4$ topology, it will be lower-semicontinuous in any stronger topology, as required if we enlarge the potential space to include more singular functions such as those in $X_L' \times X_p'$. Indeed, the original proof of lower semicontinuity of the standard DFT Levy–Lieb functional (eq 3) was with respect to the $L^1(\mathbb{R}^3)$ topology, from which the same property with respect to the X_L topology immediately follows.

Theorem and Proof. The intrinsic Hamiltonian H_0 is self-adjoint ($H_0 = H_0^\dagger$) over L^2_N, the Hilbert space of square-integrable N-electron wave functions (with spin and permutational antisymmetry built in). The expectation values of H_0 and $H(v, \mathbf{A})$ are well-defined on the Sobolev space H^1_N, the subset of L^2_N with finite kinetic energy.

We denote by $\mathcal{D}_N$ the convex set of N-electron mixed states with finite kinetic energy. We have the mathematical characterization[21]

$$\mathcal{D}_N = \{\Gamma \in \mathrm{TC}(L^2_N) | \Gamma^\dagger = \Gamma \ge 0,\ \mathrm{Tr}\Gamma = 1,\ \nabla_i \Gamma \nabla_i^\dagger \in \mathrm{TC}(L^2_N)\} \tag{7}$$

where $\mathrm{TC}(L^2_N)$ is the set of *trace-class operators* over L^2_N, the largest set of operators to which a basis-independent trace can be assigned. An operator A is trace-class if and only if the positive square root $|A| := \sqrt{A^\dagger A}$ is trace-class.[18,19] A self-

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adjoint operator A is trace-class if and only if it has a spectral decomposition of the form $A = \sum_{k=1}^{\infty} \lambda_k |\phi_k\rangle\langle\phi_k|$, where $\{\phi_k\}$ forms an orthonormal basis and $\sum_k \lambda_k$ is absolutely convergent. Now $A = \Gamma \in \mathcal{D}_N$ if and only if $\lambda_k \geq 0$, $\sum_k \lambda_k = 1$, and $\{\phi_k\} \subset H_N^1$ and if the total kinetic energy is finite (i.e., $\sum_k \lambda_k \langle\phi_k|T|\phi_k\rangle < +\infty$).

For any $\psi \in H_N^1$, the density−current pair $(\rho, \mathbf{j}_p) \in L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3)$ is defined by

$$\rho(\mathbf{r}_1) := N \int |\psi(\mathbf{r}_1; \tau_{-1})|^2 \, d\tau_{-1} \tag{8}$$

$$\mathbf{j}_p(\mathbf{r}_1) := N \, \mathrm{Im} \int \psi^*(\mathbf{r}_1; \tau_{-1}) \nabla_1 \psi(\mathbf{r}_1; \tau_{-1}) \, d\tau_{-1} \tag{9}$$

where we integrate over all spin variables and over $N - 1$ spatial coordinates, $\tau_{-1} = (\sigma_1, x_2, ..., x_N)$. For $A = \Gamma \in \mathcal{D}_N$, we can for instance compute $\rho = \rho_\Gamma$ from $\sum_k \lambda_k \rho_k$, where ρ_k is obtained from eq 8 with $\psi = \phi_k$ (and similarly for $\mathbf{j}_p$).

The theorem involves the *weak topology* on $X = L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3)$. Weak convergence of a sequence $\{x_n\} \subset X$, written as $x_n \rightharpoonup x \in X$, means that for any bounded linear functional $\omega \in X'$, we have $\omega(x_n) \to \omega(x)$ as a sequence of numbers—that is, weak convergence is the pointwise convergence of all bounded linear functionals. Recall that the dual space of $L^1(\mathbb{R}^3)$ is $L^\infty(\mathbb{R}^3)$, so $\rho_n \rightharpoonup \rho \in L^1(\mathbb{R}^3)$ if and only if $(f|\rho_n) \to (f|\rho)$ for every $f \in L^\infty(\mathbb{R}^3)$. Likewise, $(\rho_n, \mathbf{j}_{pn}) \rightharpoonup (\rho, \mathbf{j}_p) \in X$ if and only if $(f|\rho_n) \to (f|\rho)$ and $(\mathbf{a}|\mathbf{j}_{pn}) \to (\mathbf{a}|\mathbf{j}_p)$ for every $(f, \mathbf{a}) \in X'$.

The trace-class operators over a separable Hilbert space $\mathcal{H}$ are examples of *compact operators*, an infinite-dimensional generalization of finite-rank operators. Indeed, the set of compact operators $K(\mathcal{H})$ is the closure of the set of finite-rank operators in the norm topology and thus a Banach space. The dual space of $K(\mathcal{H})$ is in fact $\mathrm{TC}(\mathcal{H})$. For $B \in K(\mathcal{H})$ and $A \in \mathrm{TC}(\mathcal{H})$, the dual pairing is $\mathrm{Tr}(BA)$. Similar to the weak topology for a Banach space, the dual of a Banach space can be equipped with the *weak-* topology*. A sequence of trace-class operators $\{A_n\}$ converges weak-* to $A \in \mathrm{TC}(\mathcal{H})$ if, for each $B \in K(\mathcal{H})$, $\mathrm{Tr}(B_n A) \to \mathrm{Tr}(BA)$.

We now state and prove our main result, from which lower semicontinuity follows in Corollary 1 and expectation-valuedness in Corollary 2. The theorem is the CDFT analogue of Theorem 4.4 in ref 1.

Theorem 1. *Suppose that $(\rho, \mathbf{j}_p) \in X$ and $\{(\rho_n, \mathbf{j}_{pn})\} \subset X$ are such that $F(\rho, \mathbf{j}_p) < +\infty$ and $F(\rho_n, \mathbf{j}_{pn}) < +\infty$ for each $n \in \mathbb{N}$ and further suppose that $(\rho_n, \mathbf{j}_{pn}) \rightharpoonup (\rho, \mathbf{j}_p)$. Then there exists $\Gamma \in \mathcal{D}_N$ such that $\Gamma \to (\rho, \mathbf{j}_p)$ and $\mathrm{Tr}(H_0\Gamma) \leq \liminf_n F(\rho_n, \mathbf{j}_{pn})$.*

Proof of Theorem 1. The initial setup follows ref 1, which we here restate. Without loss of generality, we may replace $H_0 = T + W$ by $h^2 = T + W + 1$, which is self-adjoint and positive-definite. The operator h is taken to be the unique positive self-adjoint square root of $T + W + 1$.

Consider the sequence $\{g_n\}$ with elements $g_n := F(\rho_n, \mathbf{j}_{pn})$. If $g_n \to +\infty$, then the statement of the theorem is trivially true. We therefore assume that $\{g_n\}$ is bounded. There then exists a subsequence such that $g := \lim_n g_n$ exists. Furthermore, for each n there exists $\Gamma_n \in \mathcal{D}_N$ such that $\Gamma_n \to (\rho_n, \mathbf{j}_{pn})$ and $\mathrm{Tr}(h\Gamma_n h) = \mathrm{Tr}(h^2\Gamma_n) \leq g + 1/n$. To demonstrate this, we select for each n a density matrix $\Gamma_n \to (\rho_n, \mathbf{j}_{pn})$ that satisfies $\mathrm{Tr}(h^2\Gamma_n) < g_n + 1/2n$ and choose m such that $|g - g_n| < 1/2n$ for each $n > m$ (by taking a subsequence if necessary); for each $n > m$, we then have

$$0 \leq \mathrm{Tr}(h\Gamma_n h) - g = |\mathrm{Tr}(h\Gamma_n h) - g| \leq |\mathrm{Tr}(h\Gamma_n h) - g_n| + |g_n - g| \leq 1/n \tag{10}$$

Using the sequence $\{h\Gamma_n h\}$, we next establish a candidate limit density operator $\Gamma \in \mathcal{D}_N$.

The dual-space sequence of (positive-semidefinite) operators $y_n := h\Gamma_n h \in \mathrm{TC}(L_N^2)$ is uniformly bounded in the trace norm: $\|y_n\|_{\mathrm{TC}} \leq g + 1$. By the Banach−Alaoglu theorem, a norm-closed ball of finite radius in the dual space is compact in the weak-* topology. Thus, there exists $y \in \mathrm{TC}(L_N^2)$ such that for a subsequence, $\mathrm{Tr}(By_n) \to \mathrm{Tr}(By)$ for each $B \in K(L_N^2)$, meaning that y is the (possibly nonunique) weak-* limit of a subsequence of $\{y_n\}$. The limit is positive-definite, since the orthogonal projector P_Φ onto $\Phi \in L_N^2$ is a compact operator, which gives

$$\langle\Phi|y|\Phi\rangle = \mathrm{Tr}(yP_\Phi) = \lim_n \mathrm{Tr}(y_n P_\Phi) = \lim_n \langle\Phi|y_n|\Phi\rangle \geq 0 \tag{11}$$

We now define $\Gamma = h^{-1}yh^{-1}$, which fulfills all of the criteria for being an element of $\mathcal{D}_N$, except possibly $\mathrm{Tr}\Gamma = 1$, although $\mathrm{Tr}\Gamma \leq 1$ is already implied by the weak convergence. (It should be noted that Γ has finite kinetic energy since $\mathrm{Tr}(h^2\Gamma) < +\infty$.) If we can show that $\Gamma \to (\rho, \mathbf{j}_p)$, then we are done with the complete proof since $\Gamma \in \mathcal{D}_N$ follows from $\mathrm{Tr}\Gamma = N^{-1}\int_{\mathbb{R}^3} \rho(\mathbf{r}) \, d\mathbf{r} = 1$ and since

$$\mathrm{Tr}(h^2\Gamma) = \mathrm{Tr}\, y \leq \liminf_n \mathrm{Tr}\, y_n = \liminf_n \mathrm{Tr}(h^2\Gamma_n) \leq \liminf_n \{F(\rho_n, \mathbf{j}_{pn}) + 1/2n\} = \liminf_n F(\rho_n, \mathbf{j}_{pn}) \tag{12}$$

Let $(\rho', \mathbf{j}_p') \leftarrow \Gamma$ be the density associated with Γ. To demonstrate that $(\rho', \mathbf{j}_p') = (\rho, \mathbf{j}_p)$, we recall that $(\rho_n, \mathbf{j}_{pn}) \rightharpoonup (\rho, \mathbf{j}_p)$ by assumption. Since weak limits are unique, our proof is complete if we can show that $(\rho_n, \mathbf{j}_{pn}) \rightharpoonup (\rho', \mathbf{j}_p')$ in $L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3)$. The proof that $\rho_n \rightharpoonup \rho'$ is given in ref 1. Here we demonstrate that $\mathbf{j}_{pn} \rightharpoonup \mathbf{j}_p'$ by showing for each $\mathbf{a} \in \mathbf{L}^\infty(\mathbb{R}^3)$ that $(\mathbf{j}_{pn} - \mathbf{j}_p' \mid \mathbf{a}) \to 0$.

Let $\Omega \subset \mathbb{R}^3$ be a bounded domain with characteristic function χ equal to 1 on Ω and 0 elsewhere. Since $\rho, \rho' \in L^1(\mathbb{R}^3)$, for a given $\varepsilon > 0$ we may choose Ω to be sufficiently large that $\int(1 - \chi)\rho \, d\mathbf{r} < \varepsilon$ and $\int(1 - \chi)\rho' \, d\mathbf{r} < \varepsilon$. Since $\rho_n \rightharpoonup \rho$, we also have $\int(1 - \chi)(\rho_n - \rho) \, d\mathbf{r} < \varepsilon$ for sufficiently large n. From the triangle inequality, we obtain $\int(1 - \chi)\rho_n \, d\mathbf{r} \leq \int (1 - \chi)(\rho_n - \rho) \, d\mathbf{r} + \int (1 - \chi)\rho' \, d\mathbf{r}$, implying that $\int(1 - \chi)\rho_n \, d\mathbf{r} < 2\varepsilon$ for sufficiently large n.

In the notation $\tau = (\mathbf{r}_1, \tau_{-1}) = (x_1, x_2, ..., x_N)$ and $\tau_{-1} = (\sigma_1, x_2, ..., x_N)$ with space−spin coordinates $x_i = (\mathbf{r}_i, \sigma_i)$, we define

$$U_\alpha = N \, \mathrm{Im} \, \mathrm{diag} \, \partial_{1\alpha}\Gamma = N \, \mathrm{Im} \sum_\mu \lambda_\mu \overline{\psi_\mu(\tau)} \partial_{1\alpha}\psi_\mu(\mathbf{r}_1, \tau_{-1})$$

where α denotes a Cartesian component and we have introduced the spectral decomposition $\Gamma = \sum_\mu \lambda_\mu |\psi_\mu\rangle\langle\psi_\mu| \in \mathcal{D}_N$ with $\psi_\mu \in H_N^1$. We note that if $\Gamma \to (\rho_\Gamma, \mathbf{j}_{p\Gamma})$, then integration of U_α over τ_{-1} gives the current component $j_{p\alpha\Gamma}(\mathbf{r}) = \int U_\alpha(\mathbf{r}, \tau_{-1}) \, d\tau_{-1}$.

We now let $S = \prod_{i=1}^N \chi(\mathbf{r}_i)$ be the characteristic function of $\Omega^N \subset \mathbb{R}^{3N}$. By the definition of U_α, we then have

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$$I(U_\alpha) := \left|\int (1-S)U_\alpha \, d\tau\right| \le N\int (1-S)\sum_\mu \lambda_\mu |\psi_\mu||\partial_{1,\alpha}\psi_\mu| \, d\tau$$

Applying the Cauchy−Schwarz inequality twice, we obtain

$$I(U_\alpha) \le N\int (1-S)\left(\sum_\mu \lambda_\mu |\psi_\mu|^2\right)^{1/2}\left(\sum_\mu \lambda_\mu |\partial_{1,\alpha}\psi_\mu|^2\right)^{1/2} d\tau$$
$$\le (2N)^{1/2}\left(\int (1-S)\sum_\mu \lambda_\mu |\psi_\mu|^2 d\tau\right)^{1/2} \times \left(\frac{N}{2}\int \sum_\mu \lambda_\mu |\partial_{1,\alpha}\psi_\mu|^2 d\tau\right)^{1/2}$$

Noting that $1 - S \le \sum_{i=1}^N [1 - \chi(\mathbf{r}_i)]$ and using the symmetry of $|\psi_\mu|^2$, we obtain for the two factors

$$\int (1-S)\sum_\mu \lambda_\mu |\psi_\mu|^2 \, d\tau \le \int (1-\chi)\rho' \, d\mathbf{r} < \varepsilon$$

and

$$\frac{N}{2}\int \sum_\mu \lambda_\mu |\partial_\alpha \psi_\mu|^2 \, d\tau = \mathrm{Tr}(T\Gamma) \le g$$

We conclude that $I(U_\alpha)^2 \le 2Ng\varepsilon$. Introducing $U_{n,\alpha} = N$ Im diag $\partial_{1\alpha}\Gamma_n$ and proceeding in the same manner, we arrive at the bound $I(U_{n,\alpha})^2 \le 4Ng\varepsilon$, assuming that n has been chosen to be sufficiently large that $\int (1-\chi)\rho_n \, d\mathbf{r} < 2\varepsilon$ holds.

We are now ready to consider the weak convergence $\mathbf{j}_{pn} \rightharpoonup \mathbf{j}'_p$ in $\mathbf{L}^1(\mathbb{R}^3)$. For each $\mathbf{a} \in \mathbf{L}^\infty(\mathbb{R}^3)$ and for sufficiently large n, using the Cauchy−Schwarz inequality and the Hölder inequality in combination with the bounds $I(U_\alpha)^2 \le 2Ng\varepsilon$ and $I(U_{n,\alpha})^2 \le 4Ng\varepsilon$, we obtain the inequality

$$\left|\int (\mathbf{j}_{pn} - \mathbf{j}'_p)\cdot \mathbf{a} \, d\mathbf{r}\right| \le \sum_\alpha \left|\int (j_{pn\alpha} - j'_{p\alpha})a_\alpha \, d\mathbf{r}\right|$$
$$= \sum_\alpha \left|\int (U_{n,\alpha} - U_\alpha)a_\alpha(\mathbf{r}_1) \, d\tau\right|$$
$$\le \sum_\alpha \left|\int (1-S)(U_{n,\alpha} - U_\alpha)a_\alpha(\mathbf{r}_1) \, d\tau\right| + \sum_\alpha \left|\int S(U_{n,\alpha} - U_\alpha)a_\alpha(\mathbf{r}_1) \, d\tau\right|$$
$$\le \sum_\alpha \|a_\alpha\|_\infty (6Ng\varepsilon)^{1/2} + \sum_\alpha \left|\int (U_{n,\alpha} - U_\alpha)a_\alpha(\mathbf{r}_1)S \, d\tau\right| \quad (13)$$

Since $\varepsilon > 0$ is arbitrary, it only remains to show that we have $\int (U_{n,\alpha} - U_\alpha)a_\alpha(\mathbf{r}_1)S \, d\tau \to 0$ as $n \to \infty$.

Let M be the compact multiplication operator associated with $a_\alpha(\mathbf{r}_1)S(\tau)$, a bounded function with compact support over $\mathbb{R}^{3N}$. Let $\Omega_\sigma = \{\uparrow, \downarrow\}$ be the set consisting of the two spin states of the electrons. We note that

$$\int U_{n,\alpha}a_\alpha S \, d\tau = \int_{(\Omega\times\Omega_\sigma)^N} U_{n,\alpha}a(\mathbf{r}_1) \, d\tau$$
$$= N \ \mathrm{Im}\ \mathrm{Tr}(\partial_{1\alpha}\Gamma_n M)$$
$$= N \ \mathrm{Im}\ \mathrm{Tr}(h^{-1}M\partial_{1\alpha}h^{-1}y_n) \quad (14)$$

viewing Γ_n as an operator over $L^2((\Omega\times\Omega_\sigma)^N)$ by domain restriction of the spectral decomposition elements—that is, $\psi_\mu \in H^1((\Omega\times\Omega_\sigma)^N)$, meaning that the 2^N spin components of ψ_μ are in $H^1(\Omega^N)$. For simplicity, the spaces used here are not antisymmetrized.

Our next task is to demonstrate that $B = h^{-1}M\partial_{1\alpha}h^{-1}$ is compact over $L^2((\Omega\times\Omega_\sigma)^N)$. We first show that h^{-1} is compact with range $H^1((\Omega\times\Omega_\sigma)^N)$. We have $h = \sqrt{T + W + 1}$ with domain $H^1((\Omega\times\Omega_\sigma)^N)$. Now h^{-1} exists and is bounded since -1 is not in the spectrum of $T + W$—that is, h^{-1}: $L^2((\Omega\times\Omega_\sigma)^N) \to H^1((\Omega\times\Omega_\sigma)^N)$ is bounded. By the Rellich−Kondrachov theorem, $H^1(\Omega^N)$ (the standard Sobolev space without spin) is a compact subset of $L^2(\Omega^N)$. It follows that $H^1((\Omega\times\Omega_\sigma)^N)$ is a compact subset of $L^2((\Omega\times\Omega_\sigma)^N)$, since the tensor product of compact sets is compact. Hence, h^{-1} is compact.

Next, the operator $\partial_{1\alpha}$ is, by the definition of the Sobolev space $H^1(\Omega^N)$, bounded from $H^1((\Omega\times\Omega_\sigma)^N)$ to $L^2((\Omega\times\Omega_\sigma)^N)$. Thus, $\partial_{1\alpha}h^{-1}$ is bounded over $L^2((\Omega\times\Omega_\sigma)^N)$. It follows that $B \in K(L^2((\Omega\times\Omega_\sigma)^N))$ because it is a product of the compact operator h^{-1} and the bounded operator $M\partial_{1\alpha}h^{-1}$.

From the compactness of B, it follows that

$$\int U_{n,\alpha}a_\alpha S \, d\tau = N \ \mathrm{Im}\ \mathrm{Tr}(By_n) \to N \ \mathrm{Im}\ \mathrm{Tr}(By) = \int U_\alpha a_\alpha S \, d\tau \quad (15)$$

by the weak-* convergence of y_n to y. We conclude that $\mathbf{j}_{pn} \rightharpoonup \mathbf{j}'_p$ and hence that $(\rho, \mathbf{j}_p) = (\rho', \mathbf{j}'_p)$, completing the proof.

Corollary 1. *F: $L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3) \to [0, +\infty]$ is lower-semicontinuous and also weakly lower-semicontinuous.*

Proof. Let $(\rho_n, \mathbf{j}_{pn}) \rightharpoonup (\rho, \mathbf{j}_p) \in L^1(\mathbb{R}^3) \times \mathbf{L}^1(\mathbb{R}^3)$. From Theorem 1, we then obtain

$$F(\rho, \mathbf{j}_p) \le \mathrm{Tr}(H_0\Gamma) \le \liminf_n F(\rho_n, \mathbf{j}_{pn}) \quad (16)$$

where $\Gamma \to (\rho, \mathbf{j}_p)$. Hence, F is weakly lower-semicontinuous. By Mazur's lemma,[15] weak lower semicontinuity of a convex function implies strong lower semicontinuity.

Corollary 2. *If $F(\rho, \mathbf{j}_p) < +\infty$, then the infimum in the CDFT constrained-search functional is a minimum:*

$$F(\rho, \mathbf{j}_p) = \min_{\Gamma\to(\rho,\mathbf{j}_p)} \mathrm{Tr}(\Gamma H_0) \quad (17)$$

Proof. One simply takes $(\rho_n, \mathbf{j}_{pn}) = (\rho, \mathbf{j}_p)$ for all n and applies Theorem 1.

In conclusion, we have extended Theorem 4.4 of ref 1 to CDFT. As immediate corollaries, the constrained-search functional $F(\rho, \mathbf{j}_p)$ is lower-semicontinuous and expectation-valued, that is, if $F(\rho, \mathbf{j}_p) < +\infty$, then there exists $\Gamma \to (\rho, \mathbf{j}_p)$ such that $F(\rho, \mathbf{j}_p) = \mathrm{Tr}(H_0\Gamma)$. These mathematical results are the final pieces in the puzzle of placing CDFT on a solid mathematical ground in a similar manner as done by Lieb for standard DFT.

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Notes

The authors declare no competing financial interest.

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