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ORIGINAL PAPER

Kohn–Sham theory in the presence of magnetic field

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Abstract In the well-known Kohn–Sham theory in density functional theory, a fictitious non-interacting system is introduced that has the same particle density as a system of N electrons subjected to mutual Coulomb repulsion and an external electric field. For a long time, the treatment of the kinetic energy was not correct and the theory was not well-defined for N-representable particle densities. In the work of (Hadjisavvas and Theophilou in Phys Rev A 30:2183, 1984), a rigorous Kohn–Sham theory for N-representable particle densities was developed using the Levy–Lieb functional. Since a Levy–Lieb-type functional can be defined for current density functional theory formulated with the paramagnetic current density, we here develop a rigorous N-representable Kohn–Sham approach for interacting electrons in magnetic field. Furthermore, in the one-electron case, criteria for N-representable particle densities to be v-representable are given.

Keywords Density functional theory · Kohn–Sham theory · Levy–Lieb functional · Current density functional theory · N-representable

1 Introduction

In the fundamental paper by Hohenberg and Kohn [1], the theoretical foundation of density functional theory (DFT) was established. The Hohenberg–Kohn theorem states that, for a quantum mechanical system, the particle density ρ determines the scalar potential v of the system up to a constant. From this, in principle, the ground-state wavefunction can be computed. For particle densities that come from a unique

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ground-state, the so-called v-representable particle densities, an energy functional was defined and proven to satisfy a variational principle [1]. Subsequently, Kohn and Sham provided an algorithm [2], the Kohn–Sham equations, for computing the density. These equations bear much resemblance to the Hartree–Fock integro-differential equations. The idea of Kohn and Sham was to introduce a fictitious system of non-interacting particles that has the same particle density as the real interacting system. The particle density can then be computed from a determinant wavefunction. This was achieved by means of the exchange-correlation functional, which accounts for the non-classical two-particle interactions and the residual between the interacting and non-interacting kinetic energy. The domain of this functional is the intersection of the set of v-representable and non-interacting v-representable particle densities. However, this functional remains unknown.

The Hohenberg–Kohn–Sham theory relies on, when minimizing the energy, one does not go outside the domain of the exchange-correlation functional. Since for a well-behaved wavefunction ψ, the corresponding N-representable particle density ρ can be non-v-representable [3,4], one can not minimize freely over determinant wavefunctions (see also [5]). This means, in principle, that any v-representable formalism is unjustified. However, as was proven by Lieb in [3] (see also the related work of Levy [6]), for any N-representable particle density ρ there exists a wavefunction ψ, with particle density ρ, that minimizes the potential-free Hamiltonian (kinetic energy and repulsive two-particle interactions) under the constraint that ρ is fixed. Using the existence of such a minimizer, Hadjisavvas and Theophilou [7] developed a mathematically rigorous N-representable Kohn–Sham approach. The importance of this work relies on the fact that N-representability can be guaranteed for a proper wavefunction, whereas v-representability cannot.

In the presence of a magnetic field, no general Hohenberg–Kohn theorem has been proven to exist that is valid for any number of electrons. (In the special case ψ real-valued a Hohenberg–Kohn theorem can be proven, see [8]). For the formulation of current density functional theory (CDFT) that uses the paramagnetic current density j^p, it is well-known that the density pair (ρ, j^p) does not determine the scalar potential and vector potential of the system [9]. Counterexamples have been constructed that show that a ground-state can come from two different Hamiltonians [9,10]. Thus, the particle density ρ and the paramagnetic current density j^p do not fully determine the Hamiltonian. For a many-electron system, neither proof nor counterexample exists so far in the literature for a general Hohenberg–Kohn theorem formulated with the total current density j [10,11]. In the one-electron case, on the other hand, it is possible to give a direct proof that ρ and j determine the scalar and vector potential up to a gauge transformation [10,11].

However, since the density pair (ρ, j^p) determines the (possibly degenerate) ground-state(s) of the system [10,12], this work aims at continue the N-representable approach of [7] and develop a rigorous Kohn–Sham approach for CDFT formulated with the paramagnetic current density j^p. The N-representable Kohn–Sham approach outlined here does not use any variational principle for densities. Instead, the approach relies on the existence of minimizers for certain (Levy–Lieb-type) density functionals.

2 Current density functional theory

We will in this paper consider a system of N interacting electrons subjected to both an electric and a magnetic field. The system's Hamiltonian is given by (in suitable units)

$$H(v, A) = \sum_{k=1}^{N} \Big((i\nabla_k - A(x_k))^2 + v(x_k) \Big) + \sum_{1\le k<l\le N} |x_k - x_l|^{-1},$$

where $v(x)$ is the scalar potential and $A(x)$ the vector potential. The magnetic field is computed from $B(x) = \nabla \times A(x)$. Throughout we will assume that the ground-state is non-degenerate, i.e., $\dim \ker(e_0 - H(v, A)) = 1$, where e_0 is the lowest eigenvalue of $H(v, A)$.

2.1 Preliminaries

To begin with, some mathematical concepts needed for the forthcoming discussion are introduced. We first mention some relevant function spaces. If for some $p \in [1, \infty)$ a function f satisfies $\int_{\mathbb{R}^n} |f|^p < \infty$, then f belongs to the normed space $L^p(\mathbb{R}^n)$ with norm $||f||_{L^p(\mathbb{R}^n)} = (\int_{\mathbb{R}^n} |f|^p)^{1/p}$. In the case $p = \infty$, we say $f \in L^\infty(\mathbb{R}^n)$ if

$$||f||_{L^\infty(\mathbb{R}^n)} = \operatorname{ess\,sup}\{|f|\,|x \in \mathbb{R}^n\} < \infty.$$

Furthermore, $f \in L^2(\mathbb{R}^n)$ is said to belong to the Hilbert space $\mathcal{H}^1(\mathbb{R}^n)$ if

$$||f||^2_{\mathcal{H}^1(\mathbb{R}^n)} = \int_{\mathbb{R}^n} |f|^2 + \int_{\mathbb{R}^n} |\nabla f|^2 < \infty.$$

Let $B_R = \{x \in \mathbb{R}^n |\, |x| \le R\}$ for $R > 0$. Then $f \in L^1_{\mathrm{loc}}(\mathbb{R}^n)$ whenever $\int_{B_R} |f| < \infty$ for any B_R. For a vector u such that $(u)_l \in L^p$, $l = 1, 2, 3$, we write $u \in (L^p)^3$.

We say that a sequence $\{\psi_k\} \subset L^p(\mathbb{R}^n)$ converges in $L^p(\mathbb{R}^n)$-norm to $\psi \in L^p(\mathbb{R}^n)$ if $\int_{\mathbb{R}^n} |\psi_k - \psi|^p \to 0$ as $k \to \infty$, and we write $\psi_k \to \psi$. For the Hilbert space $L^2(\mathbb{R}^n)$, with inner product $(\psi, \phi)_{L^2(\mathbb{R}^n)} = \int_{\mathbb{R}^n} \psi^* \phi$, we say that $\{\psi_k\} \subset L^2(\mathbb{R}^n)$ converges weakly to $\psi \in L^2(\mathbb{R}^n)$ if $(\psi_k, \phi)_{L^2(\mathbb{R}^n)} \to (\psi, \phi)_{L^2(\mathbb{R}^n)}$ as $k \to \infty$ for all $\phi \in L^2(\mathbb{R}^n)$, and we write $\psi_k \rightharpoonup \psi$. For weak convergence in $\mathcal{H}^1(\mathbb{R}^n)$, we require $(\psi_k, \phi)_{\mathcal{H}^1(\mathbb{R}^n)} \to (\psi, \phi)_{\mathcal{H}^1(\mathbb{R}^n)}$ as $k \to \infty$ for all $\phi \in \mathcal{H}^1(\mathbb{R}^n)$, where the inner product of $\mathcal{H}^1(\mathbb{R}^n)$ is given by $(\psi, \phi)_{\mathcal{H}^1(\mathbb{R}^n)} = \int_{\mathbb{R}^n} \psi^* \phi + \int_{\mathbb{R}^n} \nabla\psi^* \cdot \nabla\phi$. Weak convergence on $\mathcal{H}^1(\mathbb{R}^n)$ implies weak convergence in the $L^2(\mathbb{R}^n)$ sense. A functional f is said to be weakly lower semi continuous if $\psi_k \rightharpoonup \psi$ implies $\liminf_{k\to\infty} f(\psi_k) \ge f(\psi)$. In particular, $\liminf_{k\to\infty} ||\psi_k||_{L^2(\mathbb{R}^n)} \ge ||\psi||_{L^2(\mathbb{R}^n)}$ if $\psi_k \rightharpoonup \psi$ weakly in $L^2(\mathbb{R}^n)$.

For a fixed particle number N, define the set of proper wavefunctions to be

$$W_N = \{\psi \in \mathcal{H}^1(\mathbb{R}^{3N}) | \psi \text{ antisymmetric and } ||\psi||_{L^2(\mathbb{R}^{3N})} = 1\}$$

and let the ground-state energy of $H(v, A)$ be given by

$$e_0(v, A) = \inf\{\mathcal{E}_{v,A}(\psi) | \psi \in W_N\},$$

where

$$\mathcal{E}_{v,A}(\psi) = \sum_{k=1}^{N} \left(\int_{\mathbb{R}^{3N}} |(i\nabla_k - A(x_k))\psi|^2 + \int_{\mathbb{R}^{3N}} |\psi|^2 v(x_k) \right) + \sum_{1 \le k < l \le N} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1}.$$

We will define the inner-product $(\psi, H(v, A)\psi)_{L^2}$ as the number $\mathcal{E}_{v,A}(\psi)$ for $\psi \in W_N$, even if $H(v, A)\psi \notin L^2$.

The particle and paramagnetic current density for $\psi \in W_N$ are computed from

$$\rho_\psi(x) = N \int_{\mathbb{R}^{3(N-1)}} |\psi(x, x_2, \ldots, x_N)|^2 dx_2 \ldots dx_N,$$
$$j^p_\psi(x) = N \,\mathrm{Im} \int_{\mathbb{R}^{3(N-1)}} \psi^*(x, x_2, \ldots, x_N) \nabla_x \psi(x, x_2, \ldots, x_N) dx_2 \ldots dx_N,$$

respectively. We will use the notation $\psi \mapsto (\rho, j^p)$ to mean $\rho_\psi = \rho$ and $j^p_\psi = j^p$. Furthermore, we shall use the notation H_0 for the Hamiltonian $H(v, A)$ when the potential terms are set to zero, i.e.,

$$(\psi, H_0\psi)_{L^2} = \sum_{k=1}^{N} \int_{\mathbb{R}^{3N}} |\nabla_k \psi|^2 + \sum_{1 \le k < l \le N} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1}.$$

Note that

$$\mathcal{E}_{v,A}(\psi) = (\psi, H(v, A)\psi)_{L^2} = (\psi, H_0\psi)_{L^2} + 2 \int_{\mathbb{R}^3} j^p_\psi \cdot A + \int_{\mathbb{R}^3} \rho_\psi (v + |A|^2),$$

which follows from a direct computation.

2.2 N-representable DFT

A v-representable particle density is a density ρ that satisfies $\rho = \rho_\psi$ and where ψ is the ground-state of some $H(v)$. (We will use the notation $H(v) = H(v, 0)$ and $e_0(v) = e_0(v, 0)$ when not considering magnetic fields). The set of N-representable particle densities is given by [3]

$$I_N = \left\{ \rho | \rho \ge 0, \int_{\mathbb{R}^3} \rho = N, \rho^{1/2} \in \mathcal{H}^1(\mathbb{R}^3) \right\}.$$

As demonstrated by Englisch and Englisch in [4], not every N-representable particle density is v-representable. For $\rho \in I_N$, the Levy–Lieb functional [3,6]

$$F_{LL}(\rho) = \inf\{(\psi, H_0\psi)_{L^2} | \psi \in W_N, \psi \mapsto \rho\}$$

is well-defined. As was proven in [3] (Theorem 3.3), there exists a $\psi_0 \in W_N$ such that $F_{LL}(\rho) = (\psi_0, H_0\psi_0)_{L^2}$ and $\rho_{\psi_0} = \rho$. The functional $F_{LL}(\rho)$ extends the Hohenberg–Kohn functional to N-representable densities, and for the ground-state energy we have

$$e_0(v) = \inf\left\{F_{LL}(\rho) + \int_{\mathbb{R}^3} \rho v | \rho \in I_N\right\}.$$

Note that the number $e_0(v)$ is well-defined for $v \in L^{3/2}(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ even if $H(v)$ does not have a ground-state. ($\int_{\mathbb{R}^3} \rho v$ is finite for all $\rho \in I_N$, since $I_N \subset L^1(\mathbb{R}^3) \cap L^3(\mathbb{R}^3)$, see [3].)

2.3 N-representable CDFT

A density pair (ρ, j^p) is said to be v-representable if there exists a ψ that is the ground-state of some Hamiltonian $H(v, A)$ such that $\rho = \rho_\psi$ and $j^p = j^p_\psi$. We denote this set of densities $\mathcal{A}_N$, i.e.,

$$\mathcal{A}_N = \{(\rho, j^p) | \text{there exists a } H(v, A) \text{ with ground-state } \psi \text{ such that } \psi \mapsto (\rho, j^p)\}.$$

Now, assume that $H(v_1, A_1)$ and $H(v_2, A_2)$ have the ground-states ψ and ϕ, respectively. Then from Theorem 9 in [10], if $\psi \mapsto (\rho, j^p)$ and $\phi \mapsto (\rho, j^p)$, it follows that $\psi = \text{const.}\,\phi$. For $(\rho, j^p) \in \mathcal{A}_N$, let ψ_{ρ, j^p} denote the ground-state of some $H(v, A)$ such that $\psi \mapsto (\rho, j^p)$. Then the generalized Hohenberg–Kohn functional

$$F_{HK}(\rho, j^p) = (\psi_{\rho, j^p}, H_0\psi_{\rho, j^p})_{L^2}$$

is well-defined on $\mathcal{A}_N$. Furthermore (Theorem 2 in [13]),

$$e_0(v, A) = \min\left\{F_{HK}(\rho, j^p) + 2\int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \Big| (\rho, j^p) \in \mathcal{A}_N\right\}$$

for $(v, A) \in V_N$, where

$$V_N = \{(v, A) | \, H(v, A) \text{ has a unique ground-state}\}.$$

However, a $\psi \in W_N$ may be such that $(\rho_\psi, j^p_\psi) \notin \mathcal{A}_N$. From Proposition 3 in [13], $\psi \in W_N$ implies that $\psi \mapsto (\rho, j^p) \in Y_N$, where

$$Y_N = \Big\{(\rho, j^p) | \rho \geq 0, \int_{\mathbb{R}^3} \rho = N, \rho^{1/2} \in \mathcal{H}^1(\mathbb{R}^3), j^p \in (L^1(\mathbb{R}^3))^3, \int_{\mathbb{R}^3} |j^p|^2 \rho^{-1} < \infty \Big\}.$$

The set Y_N is referred to as the set of N-representable density pairs (ρ, j^p). It is a convex set and $\mathcal{A}_N \subsetneq Y_N$ (Proposition 4 in [13]). For $(\rho, j^p) \in Y_N$, define as in [13]

$$Q(\rho, j^p) = \inf\{(\psi, H_0\psi)_{L^2} | \psi \in W_N, \psi \mapsto (\rho, j^p)\}.$$

The functional $Q(\rho, j^p)$ is the generalization of the Levy–Lieb functional $F_{LL}(\rho)$. It also depends on the paramagnetic current density j^p. The functional $Q(\rho, j^p)$ inherits many properties of $F_{LL}(\rho)$: by Theorems 5 and 6 in [13], we have (i) $Q(\rho, j^p) = F_{HK}(\rho, j^p)$ for $(\rho, j^p) \in \mathcal{A}_N$, (ii) there exists a $\psi_m \in W_N$ such that $Q(\rho, j^p) = (\psi_m, H_0\psi_m)_{L^2}$ and where $\psi_m \mapsto (\rho, j^p)$, and (iii)

$$e_0(v, A) = \inf \Big\{ Q(\rho, j^p) + 2\int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \Big| (\rho, j^p) \in Y_N \Big\}.$$

In [7], $F_{LL}(\rho)$ was used to obtain a rigorous Kohn–Sham theory for N-representable densities. Before generalizing this to CDFT formulated with j^p, we shall discuss the following question raised in [7]: since a $\psi_0 \in W_N$ exists such that $F_{LL}(\rho) = (\psi_0, H_0\psi_0)_{L^2}$ and $\psi_0 \mapsto \rho$, does ψ_0 satisfy any Schrödinger equation, i.e., is there a $v(x)$ such that $H(v)\psi = e\psi$?

3 Characterization of V-representable particle densities

We start be stating the mentioned result of Lieb (Theorem 3.3 in [3]) for the functional $F_{LL}(\rho)$.

Theorem 1 *There exists a ψ_0 in W_N such that for $\rho \in I_N$, $F_{LL}(\rho) = (\psi_0, H_0\psi_0)_{L^2}$ and $\rho_{\psi_0} = \rho$.*

Let $\rho \in I_N$. In light of Theorem 1, if the minimizer ψ_0 would be the ground-state of some Hamiltonian $H(v)$, then ρ would be v-representable. However, since the v-representable densities are a proper subset of the N-representable ones [4], there exists $\rho \in I_N$ such that the corresponding minimizer ψ_0 is not the ground-state of any Hamiltonian $H(v)$. Also note that, if ρ is v-representable, then the minimizer ψ_0 is also the ground-state associated with ρ. This so since if ρ is v-representable, then by the definition of the minimizer ψ_0, we have

$$(\psi_0, H_0\psi_0)_{L^2} + \int_{\mathbb{R}^3} \rho v = e_0(v)$$

for some v, i.e., ψ_0 is the ground-state of $H(v)$. (A similar result holds for a minimizer of $Q(\rho, j^p)$, see Proposition 5.)

Now, let $N = 1$. Note the following: $(\psi, H_0\psi)_{L^2} = \int_{\mathbb{R}^3} |\nabla\psi|^2 dx \geq \int_{\mathbb{R}^3} |\nabla|\psi||^2 dx$. Thus, for $F_{LL}(\rho)$, it is enough to minimize over the non-negative functions of W_1, i.e.,

$$F_{LL}(\rho) = \inf\left\{ \int_{\mathbb{R}^3} |\nabla\psi|^2 dx \,|\psi \in W_1, \psi \geq 0, \psi^2 = \rho \right\}.$$

We now give criteria when ψ_0 in Theorem 1 is an eigenfunction of some $H(v)$.

Proposition 2 *(i) Let $N = 1$ and $\rho \in I_1$ be such that ψ_0 fulfills $\Delta\psi_0 \in L^2(\mathbb{R}^3)$ and $\psi_0 \neq 0$ almost everywhere (a.e.), where $\psi_0 \geq 0$ minimizes $\int_{\mathbb{R}^3} |\nabla\psi|^2$ subject to the constraint $\psi^2 = \rho$. Then there exists a $\phi_0 \in L^2(\mathbb{R}^3)$ and a constant e such that, with $v - e = \phi_0/\rho^{1/2}$, ψ_0 satisfies*

$$-\Delta\psi_0 + v\psi_0 = e\psi_0,$$

and where $\int_{\mathbb{R}^3} v|\psi_0|^2 > -\infty$.
(ii) For $N = 1$, there exists $\rho_0 \in I_1$ such that $\Delta\psi_0 \notin L^2(\mathbb{R}^3)$, and $-\Delta\psi_0 + v\psi_0 = 0$ implies $\int_{\mathbb{R}^3} v|\psi_0|^2 = -\infty$.

Proof By assumption, $\psi_0 > 0$ a.e. and $\psi_0 = \rho^{1/2}$. Now, set $\phi_0 = \Delta\psi_0$, which is in $L^2(\mathbb{R}^3)$. Then with $v - e = \phi_0/\rho^{1/2}$ the conclusion of the first part follows, since

$$\int_{\mathbb{R}^3} v|\psi_0|^2 = \int_{\mathbb{R}^3} \phi_0\rho^{1/2} + e \geq -||\phi_0||_{L^2} + e.$$

For the second part, set, for small $|x_1|$, $\rho_0(x) = \rho_1(x_1)\tilde{\rho}(x_2, x_3)$, where $\tilde{\rho}(x_2, x_3)$ is regular and $\rho_1(x_1) = (a + b|x_1|^{\varepsilon+1/2})^2$, $a, b > 0$ and $0 < \varepsilon < 1/2$. Then $\Delta\psi_0 \notin L^2(\mathbb{R}^3)$. Furthermore, $-\Delta\psi_0 + v\psi_0 = 0$ implies $\int_{\mathbb{R}^3} v|\psi_0|^2 = -\infty$. (The density ρ_0 is the counterexample of Englisch and Englisch that shows that not every N-representable density is v-representable, see [4].) □

Note that ψ_0 is not proven to be the ground-state of $-\Delta + v$. However, we have

Corollary 3 *Let ρ, ψ_0 and ϕ_0 be as in Proposition 2 (i). In addition, assume that $\phi_0 \leq C\rho^{1/2}$ for some constant C and that $\rho^{-1} \in L^1_{loc}(\mathbb{R}^3)$. Then ψ_0 is the ground-state of $-\Delta + v$.*

Proof From Proposition 2, we know that $-\Delta\psi_0 + v\psi_0 = e\psi_0$, where $v = \phi_0/\rho^{1/2} + e$. By Schwarz's inequality, it follows that $v \in L^1_{\text{loc}}(\mathbb{R}^3)$. Since v is also bounded above, we have by Corollary 11.9 in [14] that $\psi_0 > 0$ is the ground-state of $-\Delta + v$. □

We can thus conclude with the following characterization: if $\rho \in I_1$ satisfies (i) $\rho > 0$ (a.e.), (ii) $\Delta\rho^{1/2} \in L^2(\mathbb{R}^3)$ and bounded above by a constant times $\rho^{1/2}$, and (iii) $\rho^{-1} \in L^1_{\text{loc}}$, then ρ is v-representable.

4 Rigorous Kohn–Sham theory for CDFT

By means of the Levy–Lieb-type density functional $Q(\rho, j^p)$ we can formulate a rigorous N-representable Kohn–Sham approach for CDFT as that of Ref. [7] for DFT. Now, fix the particle number N. We say that a wavefunction $\phi \in W_N$ is a determinant if there exist N orthonormal one-particle functions f^k such that

$$\phi(x_1, \ldots, x_N) = (N!)^{-1/2} \det[f^k(x_l)]_{k,l}.$$

Let the space of all normalized determinants of finite kinetic energy be denoted W_S, i.e.,

$$W_S = \{\phi | \phi \text{ is a determinant}, ||\phi||_{L^2(\mathbb{R}^{3N})} = 1, (\phi, K\phi)_{L^2(\mathbb{R}^{3N})} < \infty\},$$

where $K = -\sum_{k=1}^{N} \Delta_k$. Note that, in particular, for a $\phi \in W_S$, we have $\rho_\phi = \sum_{k=1}^{N} |f^k|^2$ and

$$(\phi, K\phi)_{L^2(\mathbb{R}^{3N})} = \sum_{k=1}^{N} \int_{\mathbb{R}^3} |\nabla f^k|^2 dx.$$

Thus, $||\phi||_{L^2(\mathbb{R}^{3N})} = 1$ and $(\phi, K\phi)_{L^2(\mathbb{R}^{3N})} < \infty$ are equivalent to $f^k \in \mathcal{H}^1(\mathbb{R}^3)$ for all k. Also note that a $\psi \in W_N$ is not in general an element of W_S, i.e., $W_S \subsetneq W_N$.

Furthermore, define, for a non-interacting system, the non-interacting Hamiltonian

$$H'(v, A) = \sum_{k=1}^{N} \left((i\nabla_k - A(x_k))^2 + v(x_k) \right).$$

The non-interacting ground-state energy is then given by

$$e_0'(v, A) = \inf\{\mathcal{E}'_{v,A}(\psi) | \psi \in W_N\},$$

where $\mathcal{E}'_{v,A}(\psi)$ is given by the relation

$$\mathcal{E}'_{v,A}(\psi) + \sum_{1 \le k < l \le N} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1} = \mathcal{E}_{v,A}(\psi).$$

This motivates: set, for $(\rho, j^p) \in Y_N$,

$$Q'(\rho, j^p) = \inf\{(\psi, K\psi)_{L^2} | \psi \in W_N, \psi \mapsto (\rho, j^p)\}.$$

For $Q(\rho, j^p)$ and $Q'(\rho, j^p)$ we have the following.

Theorem 4 *Fix $(\rho, j^p) \in Y_N$, then (i) there exists a $\psi_m \in W_N$ such that $\psi_m \mapsto (\rho, j^p)$ and $Q(\rho, j^p) = (\psi_m, H_0\psi_m)_{L^2}$, and (ii) there exists a $\psi'_m \in W_N$ such that $\psi'_m \mapsto (\rho, j^p)$ and $Q'(\rho, j^p) = (\psi'_m, K\psi'_m)_{L^2}$.*

Remark Part (i) above is just Theorem 5 in [13]. However, for (ii), we can use the same proof. For the sake of completeness we will give the proof in [13] here applied to $Q'(\rho, j^p)$. See also the related work of Higuchi and Higuchi [15], where Theorem 5 in [13] was first suggested.

Proof Let $\{\psi^j\}_{j=1}^{\infty}$ be a minimizing sequence, i.e., $\psi^j \in W_N$, $\psi^j \mapsto (\rho, j^p)$ and

$$\lim_{j\to\infty} (\psi^j, K\psi^j)_{L^2} = Q'(\rho, j^p). \tag{1}$$

Since $\{\psi^j\}_{j=1}^{\infty}$ is bounded in $\mathcal{H}^1(\mathbb{R}^{3N})$, by the Banach–Alaoglu theorem there exists a subsequence and a $\psi_m' \in \mathcal{H}^1(\mathbb{R}^{3N})$ such that $\psi^{j_k} \rightharpoonup \psi_m'$ weakly in $\mathcal{H}^1(\mathbb{R}^{3N})$ as $k \to \infty$. Since the functional $\psi \mapsto (\psi, K\psi)_{L^2}$ is weakly lower semi continuous, we know that

$$(\psi_m', K\psi_m')_{L^2} \leq Q'(\rho, j^p).$$

However, it remains to prove that $\psi_m' \mapsto (\rho, j^p)$. In the proof of Theorem 3.3 in [3], it is shown that $\psi^{j_k} \to \psi_m'$ in $L^2(\mathbb{R}^{3N})$ and $\psi_m' \mapsto \rho$. Now, let g be the characteristic function of any measurable set in $\mathbb{R}^3$. For $l = 1, 2, 3$ and $k = 1, 2, \ldots,$ let

$$I_l(k) = \left| \int_{\mathbb{R}^{3N}} [(\psi^{j_k})^* \partial_l \psi^{j_k} - (\psi_m')^* \partial_l \psi_m'] g \right|.$$

Then

$$\begin{aligned} I_l(k) &\leq \left| \int_{\mathbb{R}^{3N}} (\psi^{j_k} - \psi_m')^* (\partial_l \psi^{j_k}) g \right| + \left| \int_{\mathbb{R}^{3N}} (\psi_m')^* (\partial_l \psi^{j_k} - \partial_l \psi_m') g \right| \\ &\leq ||\psi^{j_k} - \psi_m'||_{L^2} ||(\partial_l \psi^{j_k}) g||_{L^2} + \left| \int_{\mathbb{R}^{3N}} (\psi_m' g^*)^* (\partial_l \psi^{j_k} - \partial_l \psi_m') \right|. \end{aligned}$$

Thus $I_l(k)$ tends to zero as $k \to \infty$ (because $\psi^{j_k} \to \psi_m'$ in $L^2(\mathbb{R}^{3N})$-norm and $\psi^{j_k} \rightharpoonup \psi_m'$ weakly in $\mathcal{H}^1(\mathbb{R}^{3N})$ as $k \to \infty$). Since $\psi^{j_k} \mapsto j^p$ for all k, we have $\int_{\mathbb{R}^3} (j^p)_l g = \int_{\mathbb{R}^3} (j^p_{\psi_m'})_l g$, i.e., $j^p_{\psi_m'}(x) = j^p(x)$ a.e. □

Proposition 5 *Assume that* $(\rho, j^p) \in \mathcal{A}_N$*, i.e., there exists a* $H(v, A)$ *with ground-state* ψ *such that* $\psi \mapsto (\rho, j^p)$*. Then the minimizer* ψ_m *is the ground-state of* $H(v, A)$*.*

Proof Since $\psi \mapsto (\rho, j^p)$, we have $(\psi, H_0\psi)_{L^2} \geq (\psi_m, H_0\psi_m)_{L^2}$. The conclusion then follows from

$$\begin{aligned} e_0(v, A) &\leq (\psi_m, H(v, A)\psi_m)_{L^2} = (\psi_m, H_0\psi_m)_{L^2} + 2\int_{\mathbb{R}^3} j^p \cdot A \\ &\quad + \int_{\mathbb{R}^3} \rho(v + |A|^2) \\ &\leq (\psi, H_0\psi)_{L^2} + 2\int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) \end{aligned}$$

$$= (\psi, H(v, A)\psi)_{L^2} = e_0(v, A).$$

□

Note that when H_0 is replaced by K, $Q'(\rho, j^p)$ is the minimal kinetic energy for $\psi \in W_N$ such that $\rho_\psi = \rho$ and $j^p_\psi = j^p$. Next we will introduce another kinetic energy density functional.

4.1 Non-interacting kinetic energy density functional

Set, for $(\rho, j^p) \in Y_N$,

$$T_{\text{det}}(\rho, j^p) = \inf\{(\phi, K\phi)_{L^2} | \phi \in W_S, \phi \mapsto (\rho, j^p)\}.$$

For $(\rho, j^p) \in Y_N$, we remark that the set $\{\phi \in W_S | \phi \mapsto (\rho, j^p)\}$ is not empty, at least when $N \geq 4$. This follows from the determinant construction in [16]. However, for all N, the set $\{\phi \in W_S | \phi \mapsto (\rho, j^p), \nabla \times (j^p/\rho) = 0\}$ is non-empty (see [13,16]).

We have that $T_{\text{det}}(\rho, j^p) \geq Q'(\rho, j^p)$ on Y_N. Now, let the set of non-interacting v-representable densities be denoted $\mathcal{A}'_N$,

$$\mathcal{A}'_N = \{(\rho, j^p) | H'(v, A) \text{ has a unique ground-state}\}.$$

If $(\rho, j^p) \in \mathcal{A}'_N$, by the same argument as in the proof of Proposition 5, we can conclude that ψ'_m is the ground-state of some $H'(v, A)$. Clearly, ψ'_m is in this case a determinant. Thus, $T_{\text{det}}(\rho, j^p) = Q'(\rho, j^p)$ on $\mathcal{A}'_N$.

An important property of $T_{\text{det}}(\rho, j^p)$ is that the infimum actually is a minimum. For the proof, we need the following:

(i) For $k = 1, \ldots, N$, assume that $f^k_j \to f^k$ in L^2-norm as $j \to \infty$ and for each j, $(f^k_j, f^l_j)_{L^2} = \delta_{kl}$. Then $f^1, \ldots, f^N$ are orthonormal. This so since

$$(f^k, f^l)_{L^2} = \lim_{j\to\infty} (f^k_j, f^l)_{L^2} = \lim_{j\to\infty} [(f^k_j, f^l - f^l_j)_{L^2} + (f^k_j, f^l_j)_{L^2}] = \delta_{kl},$$

where we used that $|(f^k_j, f^l - f^l_j)_{L^2}| \leq ||f^k_j||_{L^2} ||f^l - f^l_j||_{L^2} \to 0$ as $j \to \infty$.

(ii) If $f_j \rightharpoonup f$ weakly in L^2 as $j \to \infty$ and $||f_j||_{L^2} \to ||f||_{L^2}$ as $j \to \infty$, then $f_j \to f$ in L^2-norm as $j \to \infty$. (This is an elementary fact and can be checked by expanding $||f_j - f||^2_{L^2} = (f_j - f, f_j - f)_{L^2}$.)

Theorem 6 *Let* $(\rho, j^p) \in Y_N$. *If* $N < 4$ *we also assume* $\nabla \times (j^p/\rho) = 0$. *Then there exists a determinant* ϕ_m *such that* $\phi_m \mapsto (\rho, j^p)$ *and* $T_{det}(\rho, j^p) = (\phi_m, K\phi_m)_{L^2}$.

Proof Fix $(\rho, j^p) \in Y_N$ and let $\{D^j\}_{j=1}^\infty \subset W_S$ be a sequence of minimizing determinants, i.e., $D^j \mapsto (\rho, j^p)$ and $\lim_{j\to\infty}(D^j, KD^j)_{L^2} = T_{\text{det}}(\rho, j^p)$. From the proof of Theorem 4, there exists a subsequence D^{j_n} and a $\phi_m \in W_N$ such that $\phi_m \mapsto (\rho, j^p)$,

$$T_{\text{det}}(\rho, j^p) = (\phi_m, K\phi_m)_{L^2}$$

and $D^{j_n} \to \phi_m$ in L^2-norm. It remains to show that $\phi_m \in W_S$. To meet that end, let

$$D^j(x_1,\ldots,x_N) = (N!)^{-1/2}\det[f_j^k(x_l)]_{k,l},$$

where for each j the N one-particle functions f_j^k are orthonormal. By the Banach–Alaoglu theorem, there exist N functions f^k such that (for a subsequence) $f_j^k \rightharpoonup f^k$ weakly in L^2 as $j \to \infty$. We furthermore claim that $f^1,\ldots,f^N$ are orthonormal. If we could prove that $f_j^k \to f^k$ in L^2-norm, it would follow that $(f^k, f^l)_{L^2} = \delta_{kl}$.

We shall prove $f_j^k \to f^k$ by demonstrating that $||f_j^k||_{L^2} \to ||f^k||_{L^2}$. This together with the fact that $f_j^k \rightharpoonup f^k$ weakly in L^2 gives the desired result. Let $\varepsilon > 0$ and choose a characteristic function χ such that $\int_{\mathbb{R}^3}\rho(1-\chi) < \varepsilon$. Since for each j, $D^j \mapsto \rho$, we have for each k,

$$\int_{\mathbb{R}^3}|f_j^k|^2(1-\chi) \le \sum_{k=1}^N \int_{\mathbb{R}^3}|f_j^k|^2(1-\chi) = \int_{\mathbb{R}^3}\rho(1-\chi) < \varepsilon.$$

By the Rellich-Kondrachov theorem, we can choose a subsequence such that $\chi f_{j_n}^k \to \chi f^k$ in L^2-norm. But this implies

$$\int_{\mathbb{R}^3}|f^k|^2 \ge \int_{\mathbb{R}^3}\chi|f^k|^2 = \lim_{n\to\infty}\int_{\mathbb{R}^3}\chi|f_{j_n}^k|^2 \ge 1-\varepsilon.$$

Conversely, by the lower semi continuity of the L^2-norm, $1 = \liminf_{j\to\infty}||f_j^k||_{L^2} \ge ||f^k||_{L^2}$, and we have $||f^k||_{L^2} = 1$.

Returning to the fact that $f_{j_n}^k \rightharpoonup f^k$ weakly in L^2, we note that $\Pi_{k=1}^N f_{j_n}^k(x_k) \rightharpoonup \Pi_{k=1}^N f^k(x_k)$ weakly in $L^2(\mathbb{R}^{3N})$ (since product-functions are dense in $L^2(\mathbb{R}^{3N})$). But then

$$D^{j_n} \rightharpoonup (N!)^{-1/2}\det[f^k(x_l)]_{k,l},$$

where $f^1,\ldots,f^N$ are orthonormal. However, since $D^{j_n} \to \phi_m$, we have $\phi_m \in W_S$. □

4.2 N-representable Kohn–Sham theory

In the Kohn–Sham approach [2], a non-interacting system is introduced that has the same ground-state density as the fully interacting system. The idea is then to use an element of W_S, i.e., a determinant, to compute the ground-state density. On $\mathcal{A}'_N$, the (generalized) Kohn–Sham density functional $T_{KS}(\rho, j^p)$ satisfies

$$T_{KS}(\rho, j^p) = T_{\rm det}(\rho, j^p) = Q'(\rho, j^p).$$

Moreover, T_{KS} defines an exchange-correlation functional $E_{xc}(\rho, j^p)$ on $\mathcal{A}_N \cap \mathcal{A}'_N$ according to

$$E_{xc}(\rho, j^p) = F_{HK}(\rho, j^p) - \frac{1}{2}\int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \frac{\rho(x)\rho(y)}{|x-y|}dxdy - T_{KS}(\rho, j^p).$$

Now, to obtain an N-representable Kohn–Sham scheme, define two functionals on W_S,

$$\mathcal{G}_K(\phi) = \inf\{(f, Kf)_{L^2} | f \in W_S, f \mapsto (\rho_\phi, j^p_\phi)\},$$
$$\mathcal{G}_{H_0}(\phi) = \inf\{(f, H_0 f)_{L^2} | f \in W_N, f \mapsto (\rho_\phi, j^p_\phi)\}.$$

Note that, by Theorems 4 and 6, there exists a $\psi_m \in W_N$ and a $\phi_m \in W_S$ such that $\mathcal{G}_{H_0}(\phi) = (\psi_m, H_0\psi_m)_{L^2}$ and $\mathcal{G}_K(\phi) = (\phi_m, K\phi_m)_{L^2}$ and where $\psi_m, \phi_m \mapsto (\rho_\phi, j^p_\phi)$. Furthermore, we can use the existence of the minimizers ψ_m and ϕ_m and define, for $\phi \in W_S$,

$$\Delta T(\phi) = (\psi_m, K\psi_m)_{L^2} - (\phi_m, K\phi_m)_{L^2},$$
$$E^W_{xc}(\phi) = (\psi_m, \sum_{1\le k<l\le N} |x_k - x_l|^{-1}\psi_m)_{L^2} - \frac{1}{2}\int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \frac{\rho_\phi(x)\rho_\phi(y)}{|x-y|}dxdy.$$

On W_S, we now introduce the following energy functional

$$\mathcal{G}_{v,A}(\phi) = (\phi, K\phi)_{L^2} + \Delta T(\phi) + 2\int_{\mathbb{R}^3} j^p_\phi \cdot A + \int_{\mathbb{R}^3} \rho_\phi(v + |A|^2) + E^W_{xc}(\phi) + \frac{1}{2}\int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \frac{\rho_\phi(x)\rho_\phi(y)}{|x-y|}dxdy.$$

We then have

Theorem 7 *Assume that $H(v, A)$ has a unique ground-state ψ_0. Let $e_0(v, A)$, ρ_0 and j^p_0 denote the ground-state energy, ground-state particle density and ground-state paramagnetic current density, respectively. If $N < 4$ we assume that $\nabla \times (j^p_0/\rho_0) = 0$. Then*

$$e_0(v, A) = \inf\{\mathcal{G}_{v,A}(\phi) | \phi \in W_S\} = \mathcal{G}_{v,A}(\phi_m)$$

for some $\phi_m \in W_S$. Moreover, $\rho_{\phi_m} = \rho_0$ and $j^p_{\phi_m} = j^p_0$, i.e., the ground-state densities can be computed from the determinant ϕ_m that minimizes $\mathcal{G}_{v,A}$.

Proof First note, for any $\phi \in W_S$, we have

$$\mathcal{G}_{v,A}(\phi) = (\phi, K\phi)_{L^2} + \big((\psi_m, K\psi_m)_{L^2} - (\phi_m, K\phi_m)_{L^2}\big) + 2\int_{\mathbb{R}^3} j^p_\phi \cdot A + \int_{\mathbb{R}^3} \rho_\phi(v + |A|^2) + (\psi_m, \sum_{1\le k<l\le N} |x_k - x_l|^{-1}\psi_m)_{L^2}$$

$$\geq (\psi_m, (K + \sum_{1\leq k<l\leq N} |x_k - x_l|^{-1})\psi_m)_{L^2} + 2\int_{\mathbb{R}^3} j_\phi^p \cdot A$$
$$+ \int_{\mathbb{R}^3} \rho_\phi(v + |A|^2)$$
$$= \mathcal{E}_{v,A}(\psi_m) \geq e_0(v, A),$$

where we used that $(\phi, K\phi)_{L^2} - (\phi_m, K\phi_m)_{L^2} \geq 0$ and $\psi_m \mapsto (\rho_\phi, j_\phi^p)$. In the next step, we want to show that there exists a $\phi_m \in W_S$ such that $\mathcal{G}_{v,A}(\phi_m) = e_0(v, A)$ and $\phi_m \mapsto (\rho_0, j_0^p)$.

Let $\phi \in W_S$ be a determinant such that $\phi \mapsto (\rho_0, j_0^p)$ (if $N < 4$, we need the assumption $\nabla \times (j_0^p/\rho_0) = 0$). By Theorem 6, we then have

$$\mathcal{G}_K(\phi) = T_{\text{det}}(\rho_0, j_0^p) = (\phi_m, K\phi_m)_{L^2},$$

for some $\phi_m \in W_S$. Note that ϕ_m is a determinant such that $\phi_m \mapsto (\rho_0, j_0^p)$ and

$$\mathcal{G}_K(\phi_m) = (\phi_{m,m}, K\phi_{m,m})_{L^2} = (\phi_m, K\phi_m)_{L^2}.$$

Furthermore,

$$\mathcal{G}_{H_0}(\phi_m) = Q(\rho_0, j_0^p) = (\psi_m, H_0\psi_m)_{L^2},$$

for some $\psi_m \in W_N$, which follows from Theorem 4. Note that $\psi_m \mapsto (\rho_0, j_0^p) = (\rho_{\phi_m}, j_{\phi_m}^p)$. We have,

$$e_0(v, A) = (\psi_m, H(v, A)\psi_m)_{L^2}$$
$$= (\psi_m, H_0\psi_m)_{L^2} + 2\int_{\mathbb{R}^3} j_0^p \cdot A + \int_{\mathbb{R}^3} \rho_0(v + |A|^2)$$
$$= (\psi_m, K\psi_m)_{L^2} + (\psi_m, \sum_{1\leq k<l\leq N} |x_k - x_l|^{-1}\psi_m)_{L^2} + 2\int_{\mathbb{R}^3} j_{\phi_m}^p \cdot A$$
$$+ \int_{\mathbb{R}^3} \rho_{\phi_m}(v + |A|^2),$$

where the first equality follows from Proposition 5. Since

$$\Delta T(\phi_m) = (\psi_m, K\psi_m)_{L^2} - (\phi_m, K\phi_m)_{L^2}$$

and

$$E_{xc}^W(\phi_m) = (\psi_m, \sum_{1\leq k<l\leq N} |x_k - x_l|^{-1}\psi_m)_{L^2} - \frac{1}{2}\int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \frac{\rho_{\phi_m}(x)\rho_{\phi_m}(y)}{|x-y|}dxdy,$$

it follows that

$$e_0(v, A) = (\phi_m, K\phi_m)_{L^2} + 2\int_{\mathbb{R}^3} j^p_{\phi_m} \cdot A + \int_{\mathbb{R}^3} \rho_{\phi_m}(v + |A|^2)$$
$$+ \frac{1}{2}\int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \frac{\rho_{\phi_m}(x)\rho_{\phi_m}(y)}{|x-y|}dxdy + E^W_{xc}(\phi_m) + \Delta T(\phi_m) = \mathcal{G}_{v,A}(\phi_m).$$

□

Remarks.

(i) Any density pair (ρ, j^p) computed from a $\phi \in W_S$ is N-representable, but not necessarily (non-interacting) v-representable. So Theorem 7 establishes a Kohn–Sham approach for N-representable densities (whereas T_{KS} is only defined on $\mathcal{A}'_N$).

(ii) Recall that no Hohenberg–Kohn theorem can exist for CDFT formulated with the paramagnetic current density. On the other hand, since ρ and j^p determine the ground-state, the Hohenberg–Kohn variational principle continues to hold for CDFT formulated with these densities. However, the N-representable Kohn–Sham approach outlined here does not use any variational principle for densities.

(iii) If we set $\phi(x_1, \dots, x_N) = (N!)^{-1/2}\det[f^k(x_l)]_{k,l}$ and define on $(\mathcal{H}^1(\mathbb{R}^3))^N$ the functional

$$\mathcal{E}(f^1, \dots, f^N) = \sum_{k=1}^N \int_{\mathbb{R}^3} |\nabla f^k|^2 + 2\sum_{k=1}^N \int_{\mathbb{R}^3} \mathrm{Im}(f^{k*}\nabla f^k) \cdot A$$
$$+ \sum_{k=1}^N \int_{\mathbb{R}^3} |f^k|^2 (v + |A|^2)$$
$$+ \frac{1}{2}\sum_{k,l=1}^N \int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \frac{|f^k(x)|^2|f^l(y)|^2}{|x-y|}dxdy + E_{xc},$$

where $E_{xc} = \Delta T + E^W_{xc}$, we can obtain the usual Kohn–Sham equations by minimizing $\mathcal{E}(f^1, \dots, f^N)$ subject to the constraint $(f^k, f^l)_{L^2} = \delta_{kl}$.

5 Summary

In this paper, a rigorous N-representable Kohn–Sham approach has been developed. In Theorem 6, it is proven that a minimizing determinant ϕ_m exists such that

$$T_{\mathrm{det}}(\rho, j^p) = \inf\{(\phi, K\phi)_{L^2} | \phi \mapsto (\rho, j^p)\} = (\phi_m, K\phi_m)_{L^2}.$$

From this, in addition to the fact that

$$Q(\rho, j^p) = \inf\{(\psi, H_0\psi)_{L^2} | \psi \mapsto (\rho, j^p)\} = (\psi_m, H_0\psi_m)_{L^2},$$

for some wavefunction ψ_m, $T_{\text{det}}(\rho, j^p)$ and $Q(\rho, j^p)$ have been used to define functionals that account for the exchange-correlation energy and the residual energy between an interacting kinetic energy and a non-interacting one. In Theorem 7, the main result is given. Here it is shown that the ground-state energy and ground-state densities can be obtained by minimizing an energy functional over the set of normalized determinant wavefunctions with finite kinetic energy. Since any density pair (ρ, j^p) computed from such a determinant wavefunction is N-representable, but not necessarily (non-interacting) v-representable, Theorem 7 establishes a Kohn–Sham approach for N-representable densities.

Furthermore, in the one-electron case, the question when a minimizer ψ_0 of the Levy–Lieb functional $F_{LL}(\rho) = \inf\{(\psi, H_0\psi)_{L^2} | \psi \mapsto \rho\}$ is an eigenstate of some Hamiltonian $H(v) = -\Delta + v(x)$ has been addressed (Proposition 2). In Corollary 3, criteria are given for ρ when this eigenstate ψ_0 also is the ground-state. Thus, these criteria become sufficient conditions for a particle density to be v-representable.

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