Kohn-Sham Theory in the Presence of Magnetic Field

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(Dated: April 14, 2014)

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, Phys. Rev. A, 1984, 30, 2183], 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.
I. 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 $\rho$ determines the scalar potential of the system up to a constant. Subsequently, Kohn and Sham provided an algorithm [2], the so-called 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. This is 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. However, this functional remains unknown.

In the work of Hadjisavvas and Theophilou [3], a mathematically rigorous Kohn-Sham approach was developed. The importance of this work relies on the fact that $N$-representability can be guaranteed for a proper wavefunction, whereas $\nu$-representability cannot. This means, in principle, that any $\nu$-representable formalism is unjustified.

In the presence of a magnetic field, no Hohenberg-Kohn theorem exists at the present time (that is valid for any number of electrons). 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 $(\rho, j_p)$ does not determine the scalar potential and vector potential of the system [4]. Counterexamples have been constructed that show that a ground-state can come from two different Hamiltonians [4, 5]. Thus, the particle density $\rho$ 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 Hohenberg-Kohn theorem formulated with the total current density $j$ [5, 6]. In the one-electron case, on the other hand, it is possible to give a direct proof that $\rho$ and $j$ determine the scalar and vector potential up to a gauge transformation [5, 6].

However, since the density pair $(\rho, j_p)$ determines the (possibly degenerate) ground-state(s) of the system [3, 7], this work aims at continue the $N$-representable approach of [3] and develop a rigorous Kohn-Sham approach for CDFT formulated with the paramagnetic current density $j_p$. 

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II. 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)

$$H(v,A) = \sum_{k=1}^{N} \left( (i\nabla_{k} - A(x_{k}))^{2} + v(x_{k}) \right) + \sum_{1 \leq k < l \leq N} |x_{k} - x_{l}|^{-1},$$

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 $H(v,A)$.

A. 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

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

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

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

Let $B_{R} = \{x \in \mathbb{R}^{n} : |x| \leq R\}$ for $R > 0$. Then $f \in L^{1}_{\text{loc}}(\mathbb{R}^{n})$ 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 $\liminf_{k \to \infty} f(\psi_{k}) \geq f(\psi)$. In particular, $\liminf_{k \to \infty} ||\psi_{k}||_{L^{2}(\mathbb{R}^{n})} \geq ||\psi||_{L^{2}(\mathbb{R}^{n})}$ if $\psi_{k} \rightharpoonup \psi$ weakly in $L^{2}(\mathbb{R}^{n})$. 

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For a fixed particle number \( N \), define the set of proper wavefunctions to be
\[
W_N = \{ \psi \in \mathcal{H}^1(\mathbb{R}^{3N}) \mid \psi \text{ antisymmetric and } ||\psi||_{L^2(\mathbb{R}^{3N})} = 1 \}
\]
and let the ground-state energy of \( H(v,A) \) be given by
\[
e_0(v,A) = \inf \{ \mathcal{E}_{v,A}(\psi) \mid \psi \in W_N \},
\]
where
\[
\mathcal{E}_{v,A}(\psi) = \sum_{k=1}^{N} \left( \int_{\mathbb{R}^{3N}} |(i\nabla_k - A(x_k))\psi|^2 + \int_{\mathbb{R}^{3N}} |\psi|^2 v(x_k) \right) + \sum_{1 \leq k < l \leq N} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1}.
\]
We will define the inner-product \( \langle \psi, H(v,A)\psi \rangle_{L^2} \) 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
\[
\rho_\psi(x) = N \int_{\mathbb{R}^{3(N-1)}} |\psi(x,x_2,\ldots,x_N)|^2 dx_2 \ldots dx_N,
\]
\[
j^p_\psi(x) = N \text{ Im} \int_{\mathbb{R}^{3(N-1)}} \psi^*(x,x_2,\ldots,x_N)\nabla x_2 \psi(x,x_2,\ldots,x_N) dx_2 \ldots dx_N,
\]
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.,
\[
(\psi, H_0\psi)_{L^2} = \sum_{k=1}^{N} \int_{\mathbb{R}^{3N}} |\nabla k \psi|^2 + \sum_{1 \leq k < l \leq N} \int_{\mathbb{R}^{3N}} |\psi|^2 |x_k - x_l|^{-1}.
\]
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.

\[\text{B. } N\text{-representable DFT}\]

A \( v \)-representable particle density is a density \( \rho \) that satisfies \( \rho = \rho_\psi \) and where \( \psi \) 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
\[
I_N = \{ \rho \mid \rho \geq 0, \int_{\mathbb{R}^3} \rho = N, \rho^{1/2} \in \mathcal{H}^1(\mathbb{R}^3) \}.
\]
As demonstrated by Englisch and Englisch in [9], not every $N$-representable particle density is $v$-representable. For $\rho \in I_N$, the Levy-Lieb functional

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

is well-defined. As was proven in [8] (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 [8].)

### C. $N$-representable CDFT

A density pair $(\rho, j^p)$ is said to be $v$-representable if there exists a $\psi$ 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 $A_N$, i.e.,

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

Now, assume that $H(v_1, A_1)$ and $H(v_2, A_2)$ have the ground-states $\psi$ and $\phi$, respectively. Then from Theorem 9 in [5], if $\psi \mapsto (\rho, j^p)$ and $\phi \mapsto (\rho, j^p)$, it follows that $\psi = \text{const.} \phi$. For $(\rho, j^p) \in A_N$, let $\psi_{\rho,j^p}$ 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 $A_N$. Furthermore (Theorem 2 in [10]),

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

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 $(\rho_\psi, j^p_\psi) \notin A_N$. From Proposition 3 in [10], $\psi \in W_N$ implies that $\psi \mapsto (\rho, j^p) \in Y_N$, where

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

The set $Y_N$ is referred to as the set of $N$-representable density pairs $(\rho, j^p)$. It is a convex set and $A_N \subset Y_N$ (Proposition 4 in [10]). For $(\rho, j^p) \in Y_N$, define as in [10]

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

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 Theorem 5 and Theorem 6 in [10], we have (i) $Q(\rho, j^p) = F_{HK}(\rho, j^p)$ for $(\rho, j^p) \in A_N$, (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)

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

In [3], $F_{LL}(\rho)$ 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 [3]: 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 $\psi_0$ satisfy any Schrödinger equation, i.e., is there a $v(\cdot)$ such that $H(v)\psi = e\psi$?

III. CHARACTERIZATION OF $V$-REPRESENTABLE PARTICLE DENSITIES

We start by stating the mentioned result of Lieb (Theorem 3.3 in [8]) for the functional $F_{LL}(\rho)$.

**Theorem 1** There exists a $\psi_0$ 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 $\psi_0$ would be the ground-state of some Hamiltonian $H(v)$, then $\rho$ would be $v$-representable. However, since the $v$-representable densities are a proper subset of the $N$-representable ones [9], there exists $\rho \in I_N$ such that the corresponding minimizer $\psi_0$ is not the ground-state of any Hamiltonian $H(v)$. Also note
that, if \( \rho \) is \( v \)-representable, then the minimizer \( \psi_0 \) is also the ground-state associated with \( \rho \). This so since if \( \rho \) is \( v \)-representable, then by the definition of the minimizer \( \psi_0 \), we have

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

for some \( v \), i.e., \( \psi_0 \) 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 \( \psi_0 \) in Theorem 1 is an eigenfunction of some \( H(v) \).

**Proposition 2**

(i) Let \( N = 1 \) and \( \rho \in I_1 \) be such that \( \psi_0 \) 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 \( v - e = \phi_0/\rho^{1/2} \), \( \psi_0 \) 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 \( \rho_1(x_1) = (a + b|x_1|^{\epsilon+1/2})^2 \), \( a,b < 0 \) and \( 0 < \epsilon < 1/2 \). 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 \( \rho_0 \) is the counterexample of Englisch and Englisch that shows that not every \( N \)-representable density is \( v \)-representable, see [9].) ■

Note that \( \psi_0 \) is not proven to be the ground-state of \( -\Delta + v \). However, we have
Corollary 3 Let $\rho, \psi_0$ and $\phi_0$ 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 $\psi_0$ 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 $v \in L^1_{loc}(\mathbb{R}^3)$. Since $v$ is also bounded above, we have by Corollary 11.9 in [11] 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) $\rho^{-1} \in L^1_{loc}$, then $\rho$ is $v$-representable.

IV. 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. [3] 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

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

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

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

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 $Q'(\rho, j^p) = (\psi'_m, K\psi'_m)_{L^2}$.

**Proof.** Part (i) above is just Theorem 5 in [10]. However, for (ii), we can use the same proof. For the sake of completeness we include the proof in [10] here applied to $Q'(\rho, j^p)$.

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

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

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 $\psi^j \rightharpoonup \psi'_m$ 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 [8], it is shown that $\psi^j \rightharpoonup \psi'_m$ 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

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

Then

$$I_l(k) \leq \left| \int_{\mathbb{R}^{3N}} (\psi^{jk} - \psi'_m)^* (\partial_l \psi^{jk}) g \right| + \left| \int_{\mathbb{R}^{3N}} (\psi'_m)^* (\partial_l \psi^{jk} - \partial_l \psi'_m) g \right|$$ 

$$\leq ||\psi^{jk} - \psi'_m||_{L^2} ||(\partial_l \psi^{jk}) g||_{L^2} + \left| \int_{\mathbb{R}^{3N}} (\psi'_m g^*)^* (\partial_l \psi^{jk} - \partial_l \psi'_m) g \right|.$$ 

Thus $I_l(k)$ tends to zero as $k \to \infty$ (because $\psi^{jk} \to \psi'_m$ in $L^2(\mathbb{R}^{3N})$-norm and $\psi^{jk} \to \psi'_m$ weakly in $\mathcal{H}^1(\mathbb{R}^{3N})$ as $k \to \infty$). Since $\psi^{jk} \mapsto j^p$ for all $k$, we have $\int_{\mathbb{R}^3} (j^p) g = \int_{\mathbb{R}^3} (j'_{\psi'_m}) g$, 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 \(\psi\) such that \(\psi \mapsto (\rho, j^p)\). Then the minimizer \(\psi_m\) 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

\[
e_0(v, A) \leq (\psi_m, H(v, A)\psi_m)_{L^2} = (\psi_m, H_0\psi_m)_{L^2} + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2)
\leq (\psi, H_0\psi)_{L^2} + 2 \int_{\mathbb{R}^3} j^p \cdot A + \int_{\mathbb{R}^3} \rho(v + |A|^2) = (\psi, H(v, A)\psi)_{L^2} = e_0(v, A).\]

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.

A. 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 [12]. 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 [10, 12]).

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 \(\psi'_m\) is the ground-state of some \(H'(v, A)\). Clearly, \(\psi'_m\) 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

\[ (f^k, f^l)_{L^2} = \lim_{j \to \infty} (f^k_j, f^l_j)_{L^2} = \lim_{j \to \infty} (f^k_j, f^l_j - f^k_j)_{L^2} + (f^k_j, f^l_j)_{L^2} = \delta_{kl}, \]
where we used that \( |(f_j^k, f^l - f_j^l)_L^2| \leq ||f_j^k||_L^2 ||f^l - f_j^l||_L^2 \to 0 \) as \( j \to \infty \).

(ii) If \( f_j \to f \) 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 \( ||f_j - f||_L^2 = (f_j - f, f_j - f)_L^2 \).

**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 \( \phi_m \) 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 \( \{D^j\}_j^{\infty} \subset W_S \) be a sequence of minimizing determinants, i.e., \( D^j \mapsto (\rho, j^p) \) and \( \lim_{j \to \infty} (D^j, K D^j)_L^2 = T_{det}(\rho, j^p) \). From the proof of Theorem 4 there exists a subsequence \( D^{j_n} \) and a \( \phi_m \in W_N \) such that \( \phi_m \mapsto (\rho, j^p) \),

\[
T_{det}(\rho, j^p) = (\phi_m, K\phi_m)_L^2
\]

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

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

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

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

\[
\int_{\mathbb{R}^3} |f^k_j|^2 (1 - \chi) \leq \sum_{k=1}^N \int_{\mathbb{R}^3} |f^k_j|^2 (1 - \chi) = \int_{\mathbb{R}^3} \rho(1 - \chi) < \varepsilon.
\]

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

\[
\int_{\mathbb{R}^3} |f^k|^2 \geq \int_{\mathbb{R}^3} \chi |f^k|^2 = \lim_{n \to \infty} \int_{\mathbb{R}^3} \chi |f^k_{j_n}|^2 \geq 1 - \varepsilon.
\]

Conversely, by the lower semi continuity of the \( L^2 \)-norm, \( 1 = \liminf_{j \to \infty} ||f^k_j||_L^2 \geq ||f^k||_L^2 \), and we have \( ||f^k||_L^2 = 1 \).
Returning to the fact that \( f^k \rightharpoonup f \) weakly in \( L^2 \), we note that \( \Pi^{N}_{k=1} f^k(x_k) \rightharpoonup \Pi^{N}_{k=1} f(x_k) \) 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 \( \phi_m \in W_S \). ■

B. 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 \( A'_N \), the (generalized) Kohn-Sham density functional \( T_{KS}(\rho, j^p) \) satisfies
\[
T_{KS}(\rho, j^p) = T_{\det}(\rho, j^p) = Q'(\rho, j^p).
\]
Moreover, \( T_{KS} \) defines an exchange-correlation functional \( E_{xc}(\rho, j^p) \) on \( A_N \cap A'_N \) according to
\[
E_{xc}(\rho, j^p) = \frac{1}{2} \int \int |x - y| dxdy - T_{KS}(\rho, j^p).
\]
Now, to obtain an \( N \)-representable Kohn-Sham scheme, define two functionals on \( W_S \),
\[
\mathcal{G}_K(\phi) = \inf \{(f,Kf)_{L^2} | f \in W_S, f \mapsto (\rho_\phi, j^p_\phi)\},
\]
\[
\mathcal{G}_{H_0}(\phi) = \inf \{(f,H_0f)_{L^2} | f \in W_N, f \mapsto (\rho_\phi, j^p_\phi)\}.
\]
Note that, by Theorem 4 and Theorem 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 \( \psi_m, \phi_m \) and define, for \( \phi \in W_S \),
\[
\Delta T(\phi) = (\psi_m, K\psi_m)_{L^2} - (\phi_m, K\phi_m)_{L^2},
\]
\[
E_{xc}^W(\phi) = (\psi_m, \sum_{1 \leq k < l \leq N} |x_k - x_l|^{-1}\psi_m)_{L^2} - \frac{1}{2} \int \int |x - y| \frac{\rho_\phi(x)\rho_\phi(y)}{|x - y|} dxdy.
\]
On \( W_S \), we now introduce the following energy functional
\[
\mathcal{G}_{v,A}(\phi) = (\phi, K\phi)_{L^2} + \Delta T(\phi) + 2 \int \int j^p_\phi \cdot A
\]
\[
+ \int \rho_\phi(v + |A|^2) + E_{xc}^W(\phi) + \frac{1}{2} \int \int \frac{\rho_\phi(x)\rho_\phi(y)}{|x - y|} dxdy.
\]
We then have
Theorem 7 Assume that $H(v, A)$ has a unique ground-state $\psi_0$. Let $e_0(v, A)$, $\rho_0$ and $j_0^p$ denote the ground-state energy, ground-state particle density and ground-state paramagnetic current density, respectively. If $N < 4$ we assume that $\nabla \times (j_0^p/\rho_0) = 0$. Then

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

for some $\phi_m \in W_S$. Moreover, $\rho_{\phi_m} = \rho_0$ and $j_{\phi_m}^p = j_0^p$, i.e., the ground-state densities can be computed from the determinant $\phi_m$ that minimizes $G_{v, A}$.

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

$$G_{v, A}(\phi) = (\phi, K\phi)_{L^2} + ((\psi_m, K\psi_m)_{L^2} - (\phi_m, K\phi_m)_{L^2})$$

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

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

$$= \mathcal{E}_{v, A}(\psi_m) \geq e_0(v, A),$$

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 $\phi_0 \in W_S$ such that $G_{v, A}(\phi_0) = e_0(v, A)$ and $\phi_0 \mapsto (\rho_0, j_0^p)$.

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

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

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

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

Furthermore,

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

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,

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

$$= (\psi_m, H_0\psi_m)_{L^2} + 2 \int_{\mathbb{R}^3} j_0^p \cdot A + \int_{\mathbb{R}^3} \rho_0 (v + |A|^2)$$

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

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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

\[ e_0(v, A) = (\phi_m, K\phi_m)_{L^2} + 2 \int_{\mathbb{R}^3} j^p_{\phi_m} \cdot A + \int_{\mathbb{R}^3} \rho_{\phi_m}(v + |A|^2) \]

\[ + \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho_{\phi_m}(x)\rho_{\phi_m}(y)}{|x - y|} dxdy + E_{xc}^W(\phi_m) + \Delta T(\phi_m) = G_{v,A}(\phi_m). \]

**Remarks.** (i) Any density pair \((\rho, j^p)\) 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 \(A'_N\)).

(ii) Recall that no Hohenberg-Kohn theorem can exist for CDFT formulated with the paramagnetic current density. On the other hand, since \(\rho\) 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. Instead, the approach relies on the existence of minimizers for certain functionals.

(iii) If we set \(\phi(x_1, \ldots, x_N) = (N!)^{-1/2} \det[f^k(x_i)]_{k,l}\) and define on \((\mathcal{H}_1(\mathbb{R}^3))^N\) the functional

\[ \mathcal{E}(f^1, \ldots, f^N) = \sum_{k=1}^N \int_{\mathbb{R}^3} |\nabla f^k|^2 + 2 \sum_{k=1}^N \int_{\mathbb{R}^3} \text{Im}(\overline{f^k} \nabla f^k) \cdot A + \sum_{k=1}^N \int_{\mathbb{R}^3} |f^k|^2(v + |A|^2) \]

\[ + \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{|f^k(x)|^2|f^l(y)|^2}{|x - y|} dxdy + E_{xc}, \]

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

**ACKNOWLEDGMENTS**

The author is thankful to Michael Benedicks for useful comments and discussions.
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