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# On the existence of effective potentials in time-dependent density functional theory

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**Abstract**
We investigate the existence and properties of effective potentials in time-dependent density functional theory. We outline conditions for a general solution of the corresponding Sturm–Liouville boundary value problems. We define the set of potentials and $v$-representable densities, give a proof of existence of the effective potentials under certain restrictions and show the set of $v$-representable densities to be independent of the interaction.

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## 1. Introduction

The calculation of the wavefunction of a fully interacting many-body quantum system is a formidable challenge [1]. Hence, feasible approaches to the quantum many-body problem are very important if one is interested in the properties of complex multi-particle systems. One such approach is Nobel-Prize-winning density functional theory (DFT) [1, 2]. The main theorem of DFT is the Hohenberg–Kohn theorem [3], which proves that the external potential of an interacting $N$-body system uniquely defines the one-particle density of the ground state via minimization of the corresponding energy functional. The mathematical foundations of DFT were extensively investigated and a rigorous formulation is available [4, 5]. Substantial extensions of the ground-state theory can be found in the literature [2], e.g., to excited states or to relativistic systems.

The mainstay of applications of the minimization principle of DFT is due to Kohn and Sham [6]. The so-called Kohn–Sham scheme uses an auxiliary system of noninteracting particles which has the same energy and one-particle density as the interacting system. To make contact with the physical system, one introduces the so-called exchange-correlation energy functional [2]. It accounts for the difference between the combined kinetic and interaction

  

energy of the interacting system and the kinetic energy of the noninteracting Kohn–Sham system. The variational minimization of the energy with respect to the density leads to a set of coupled, nonlinear single-particle differential equations, the so-called Kohn–Sham equations. It can be rigorously proven that a self-consistent solution of these equations will generate the exact one-particle ground-state density of the corresponding interacting system [2]. In practice, however, the exchange-correlation energy functional is not known and has to be approximated.

An exact extension of DFT to time-dependent systems was given in [7] by Runge and Gross. By assuming the external potentials to be Taylor expandable in time about $t = t_0$ they could prove a one-to-one correspondence between time-dependent densities and external potentials. However, the straightforward extension of the Kohn–Sham scheme to the time-dependent case as shown in [7] led to the so-called symmetry-causality paradox [8]. This flaw in the time-dependent extension of DFT was soon realized to be connected to the naive application of the usual variational principle of time-dependent quantum mechanics to TDDFT [9]. Only with an extension of the Runge–Gross theorem by van Leeuwen in [10] one was able to justify a time-dependent Kohn–Sham scheme. There it was shown that by the successive solution of Sturm–Liouville boundary value problems one can formally construct a unique effective potential governing the time evolution of the noninteracting system such that it reproduces the interacting one-particle density. In view of this theorem, one does not need a variational approach analogously to time-independent DFT and can obtain the exact one-particle density via propagation of the time-dependent Kohn–Sham equations.

Note that, although the extended Runge–Gross theorem [10] shows the uniqueness of the effective potential, the conditions of existence were not investigated.

The intention of this work is not to give an introduction to TDDFT, for this we refer to [11], but to consider the mathematical foundations of the theory. In contrast to DFT, a mathematically rigorous formulation of TDDFT is missing. Here we will take a first step toward this goal. We will give conditions for the existence of a solution to the Sturm–Liouville boundary value problems at hand. We will introduce the set of external potentials and time-dependent densities under consideration. We will prove that all orders of the Taylor expansions of the effective potentials exist, if the initial configurations and the different two-particle interactions as well as the external potential of the interacting system are spatially infinitely differentiable. Given that these conditions hold, we will show the set of $v$-representable time-dependent densities being purely determined by the initial one-particle density and its first derivative in time at $t = t_0$.

Section 2 summarizes the basic theorems of TDDFT so far. Section 3 outlines fundamental properties of the potentials and investigates the existence of general solutions of the corresponding Sturm–Liouville problems. In section 4, we introduce a certain set of potentials and $v$-representable densities and prove the existence of the solutions of all considered Sturm–Liouville boundary value problems, and hence all orders of the Taylor expansion of the effective potentials, under certain restrictions. Assuming these conditions we can evidence properties of the set of $v$-representable densities. Finally we conclude in section 5.

## 2. Time-dependent density functional theory

We consider a general many-body Hamiltonian in atomic units of the form

$$\hat{H}(t) = \hat{T} + \hat{V}_{\text{int}} + \hat{V}([v]; t). \tag{1}$$

The operator $\hat{T} = \sum_\sigma \int \mathrm{d}^3 r\, \hat{\psi}^\dagger_\sigma(r) \left(-\frac{1}{2}\nabla^2\right) \hat{\psi}_\sigma(r)$ is the kinetic term, $\hat{V}_{\mathrm{int}} = \frac{1}{2}\sum_{\sigma,\sigma'} \iint \mathrm{d}^3 r\, \mathrm{d}^3 r' v_{\mathrm{int}}(|r-r'|) \hat{\psi}^\dagger_\sigma(r) \hat{\psi}^\dagger_{\sigma'}(r') \hat{\psi}_{\sigma'}(r') \hat{\psi}_\sigma(r)$ is the interaction and $\hat{V}([v];t) = \sum_\sigma \int \mathrm{d}^3 r\ v(r,t)\ \hat{\psi}^\dagger_\sigma(r) \hat{\psi}_\sigma(r)$ is the external potential, where $\hat{\psi}^\dagger_\sigma(r)$ and $\hat{\psi}_\sigma(r)$ are the creation and annihilation operators with spin $\sigma$ [12]. The interaction potential $v_{\mathrm{int}}(|r-r'|)$ is arbitrary but will usually be chosen to be equal to the Coulomb interaction. The external one-particle potential $v(r, t)$ typically consists of a static part, e.g. the attractive Coulomb potential of a fixed nucleus, and a time-dependent part, e.g. a laser pulse in dipole approximation and length gauge. The time-dependent one-particle density is defined by the expectation value of

$$\hat{n}(r) := \sum_\sigma \hat{\psi}^\dagger_\sigma(r) \hat{\psi}_\sigma(r) \tag{2}$$

with the time-dependent density matrix $\hat{\rho}(t)$, i.e.

$$n(r,t) = \langle \hat{n}(r) \rangle = \mathrm{tr}\left(\hat{\rho}(t)\, \hat{n}(r)\right). \tag{3}$$

With the current-density operator

$$\hat{\jmath}(r) := \frac{1}{2\,\mathrm{i}} \sum_\sigma \left\{ \hat{\psi}^\dagger_\sigma(r) \nabla \hat{\psi}_\sigma(r) - \left[\nabla \hat{\psi}^\dagger_\sigma(r)\right] \hat{\psi}_\sigma(r) \right\} \tag{4}$$

it is straightforward to find the usual continuity equation via application of the Heisenberg equation

$$\partial_t n(r,t) = -\mathrm{i} \langle [\hat{n}(r), \hat{H}(t)]_- \rangle = -\nabla \cdot j(r,t), \tag{5}$$

where $\partial_t \equiv \partial/\partial t$ and $[\cdot,\cdot]_-$ is the usual commutator. The time derivative of the current-density leads to the local force balance equation [11]

$$\partial_t j_\nu(r,t) = -n(r,t) \partial_\nu v(r,t) - \partial_\mu \langle \hat{T}_{\mu\nu}(r) \rangle - \langle \hat{W}_\nu(r) \rangle, \tag{6}$$

where we made use of the Einstein summation convention, i.e. summing over multiple indices, the momentum-stress-tensor

$$\hat{T}_{\mu\nu}(r) := \frac{1}{2} \sum_\sigma \left\{ \left(\partial_\mu \hat{\psi}^\dagger_\sigma(r)\right) \partial_\nu \hat{\psi}_\sigma(r) + \left(\partial_\nu \hat{\psi}^\dagger_\sigma(r)\right) \partial_\mu \hat{\psi}_\sigma(r) - \frac{1}{2} \partial_\mu \partial_\nu \left(\hat{\psi}^\dagger_\sigma(r) \hat{\psi}_\sigma(r)\right) \right\}, \tag{7}$$

and the divergence of the interaction-stress-tensor [13]

$$\hat{W}_\nu(r) := \sum_{\sigma,\sigma'} \int \mathrm{d}^3 r'\ \left(\partial_\nu v_{\mathrm{int}}(|r-r'|)\right) \hat{\psi}^\dagger_\sigma(r) \hat{\psi}^\dagger_{\sigma'}(r') \hat{\psi}_{\sigma'}(r') \hat{\psi}_\sigma(r). \tag{8}$$

Now we will shortly sketch the idea underlying the Runge–Gross proof. Assume two external potentials $v(r, t)$ and $v'(r, t)$, both Taylor expandable about the initial time $t = t_0$, which differ by more than a merely time-dependent function $c(t)$, i.e. $v(r,t) - v'(r,t) \neq c(t)$. If we evolve an initial configuration $\hat{\rho}(t_0) = \hat{\rho}_0$ of a finite multi-particle system in time with the two different external potentials, we can investigate the difference of the time derivatives of the current-densities at time $t = t_0$ via the application of equation (6). If the time derivatives of the current-densities differ for some order, then the corresponding densities will be different after an infinitesimal time step. This leads to the Runge–Gross theorem [7].

**Theorem 1.** *For every single-particle potential $v(r, t)$ which can be expanded into a Taylor series with respect to the time coordinate around $t = t_0$, a map $G : v(r,t) \mapsto n(r,t)$ is defined by solving the time-dependent Schrödinger equations with fixed initial configuration $\hat{\rho}(t_0) = \hat{\rho}_0$ and calculating the corresponding density $n(r, t)$. This map can be inverted up to an additive, merely time-dependent function in the potential.*

Now let us fix the additive merely time-dependent function in the potential to be equal to zero by the boundary condition $v(r,t) \to 0$ for $|r| \to \infty$. Hence, we have an invertible mapping $G$ which obviously depends on the initial configuration $\hat{\rho}_0$. The proof is not restricted to Coulombic interactions and can be applied to any reasonable interaction. The domain and the range of this mapping are not further investigated in the original paper [7]. With the mapping $F : v(r,t) \mapsto \hat{\rho}(t)$ defined by the solutions of the associated Schrödinger equations, we further find via $F \circ G^{-1} : n(r,t) \mapsto \hat{\rho}(t)$ that every expectation value of an operator $\hat{O}$, i.e. $O = \mathrm{tr}[\hat{O}\hat{\rho}(t)]$, is uniquely determined by the density alone. Although this theorem holds also for noninteracting systems, it is not clear that every density subject to the Runge–Gross theorem in an interacting system can be reproduced by some effective Kohn–Sham potential in a noninteracting system. In other words, it is not known if the interacting $v$-representable density is also noninteracting $v$-representable. Initial attempts to construct such connections in the original paper [7] led to the symmetry-causality paradox [8].

In order to overcome these problems, the extended Runge–Gross theorem was introduced in [10]. A sketch of the proof reads as follows.

We apply the continuity equation (5) to the local force balance equation (6), leading to

$$\frac{\partial^2}{\partial t^2} n(r,t) = \nabla \cdot [n(r,t) \nabla v(r,t)] + \left\langle \underbrace{\partial_\nu (\partial_\mu \hat{T}_{\mu\nu}(r) + \hat{W}_\nu(r))}_{=:\hat{q}(r)} \right\rangle. \tag{9}$$

If we now assume the external potential $v(r,t)$ as well as the density $n(r,t)$ to be analytic about $t = t_0$, the different orders of the Taylor expansion are connected via equation (9) leading to [11]

$$n^{(k+2)}(r) = q^{(k)}(r) + \sum_{l=0}^{k} \binom{k}{l} \nabla \cdot [n^{(k-l)}(r) \nabla v^{(l)}(r)], \tag{10}$$

for $k > 1$ where we used $n^{(k)}(r) = \partial_t^k n(r,t)|_{t=t_0}$ and $q^{(k)}(r)$ is defined by applying the Heisenberg equation $k$ times to $\hat{q}(r)$ at time $t = t_0$. Hence, $q^{(k)}(r)$ contains terms $v^{(l)}(r)$ up to order $l = k-1$. For a second system with Hamiltonian

$$\hat{H}'(t) = \hat{T} + \hat{V}'_{\mathrm{int}} + \hat{V}'([v'];t) \tag{11}$$

and initial state $\hat{\rho}'(t_0) = \hat{\rho}'_0$ and accordingly redefined operator $\hat{q}'(r)$ we can rederive equation (10) for the primed system. Given the time-dependent density $n(r,t)$ of the unprimed system we can define the potential $v'(r,t)$ leading to the same density in the primed system in terms of its Taylor expansion in the form of a Sturm–Liouville problem:

$$\nabla \cdot [n^{(0)}(r) \nabla v'^{(k)}(r)] = n^{(k+2)}(r) - q'^{(k)}(r) - \sum_{l=0}^{k-1} \binom{k}{l} \nabla \cdot [n^{(k-l)}(r) \nabla v'^{(l)}(r)]. \tag{12}$$

With this we can state the extended Runge–Gross theorem [10].

**Theorem 2.** *For a Hamiltonian $\hat{H}(t)$ with an analytic potential $v(r,t)$ about $t = t_0$ we assume the density $n(r,t)$ generated via propagation of the initial configuration $\hat{\rho}_0$ to be analytic about $t = t_0$ as well. For a second system $\hat{H}'(t)$ with initial configuration $\hat{\rho}_0$ subject to the conditions*

$$n(r,t_0) = n^{(0)}(r) = n'(r,t_0), \tag{13a}$$

$$\mathrm{tr}\,(\hat{\rho}_0 \nabla \cdot \hat{\jmath}(r)) = n^{(1)}(r) = \mathrm{tr}(\hat{\rho}'_0 \nabla \cdot \hat{\jmath}(r)), \tag{13b}$$

*and an interaction $\hat{V}'_{\mathrm{int}}$ assumed such that its expectation value and its derivatives are finite, the analytic potential $v'(r,t)$ leading to the same density $n(r,t)$ is uniquely defined up to a purely time-dependent function.*

Again we can fix the purely time-dependent function of the potentials to be equal to zero by choosing the boundary condition $v'(r,t) \to 0$ for $|r| \to \infty$. The existence of this potential $v'(r,t)$ is investigated in [9] by minimization of a corresponding functional. However, a rigorous proof of existence was not given.

Note, in the above schematically depicted proof of the extended Runge–Gross theorem one made use of the knowledge of the density $n(r,t)$. However, we want a theory *predicting* the density. That TDDFT is a predictive theory can be seen if we use equation (10) for the primed and the unprimed system and assume both to yield the same density $n(r,t)$. With the definition

$$v'(r,t) = v(r,t) + v_\Delta(r,t) \tag{14}$$

we thus infer the Sturm–Liouville problem

$$\begin{aligned}\nabla \cdot \left[n^{(0)}(r)\nabla v_\Delta^{(k)}([n];r)\right] &= \zeta^{(k)}(r)\\ &:= q^{(k)}(r) - q'^{(k)}(r) - \sum_{l=0}^{k-1}\binom{k}{l}\nabla\cdot\left[n^{(k-l)}(r)\nabla v_\Delta^{(l)}([n];r)\right].\end{aligned} \tag{15}$$

Via equation (15) one can generate $v'^{(k)}(r)$ which can then be used in equation (10) to find the corresponding higher terms of the Taylor expansion of $n(r,t)$. In return, the higher terms of the density Taylor expansion again determine the next term in the Taylor expansion of the potential. Hence, only the initial configurations and the external potential of the unprimed system are needed to generate the expansion of the density about $t = t_0$. However, the special form of $q^{(k)}(r)$ defined by successive application of the Heisenberg equation led to the question whether a simultaneous solution of the unprimed system is required in principle [14, 15].

## 3. Properties of the potentials and the Sturm–Liouville problem

In order to define the sets of one-particle potentials and the associated one-particle densities, i.e. the $v$-representable densities, we start by looking at the properties of the corresponding Hamiltonian $\hat{H}(t)$. We demand the Hamiltonian to be self-adjoint for every time $t$ on the domain of the kinetic energy operator $\mathrm{dom}(\hat{T})$. The free Hamiltonian $\hat{H}_0 = \hat{T} + \hat{V}_{\mathrm{int}}$ with $\hat{V}_{\mathrm{int}}$ being the Coulomb interaction can be shown to fulfill this constraint [5], and any other interaction $\hat{V}'_{\mathrm{int}}$ under consideration will be assumed to do so as well. This condition is trivially fulfilled if we choose $\hat{V}'_{\mathrm{int}} \equiv 0$. With the theory of Kato perturbations [5] we find the simple condition

$$v(t) \in L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3) \tag{16}$$

for every time $t$. Here $L^2$ and $L^\infty$ are the usual Lebesgue quotient spaces with norm $\|\cdot\|_2$ and $\|\cdot\|_\infty$, respectively. $L^2(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$ is a Banach space with the norm

$$\|v(t)\| = \inf\{\|v_1(t)\|_2 + \|v_2(t)\|_\infty \,|\, v_1(t) \in L^2(\mathbb{R}^3),\ v_2(t) \in L^\infty(\mathbb{R}^3),\ v(t) = v_1(t) + v_2(t)\}. \tag{17}$$

The effective potentials $v'(r,t)$ leading to the same density as in the Coulombic system exist in accordance with the extended Runge–Gross proof if all orders of equation (12) have an existing solution. Accordingly, this is true if all orders of equation (15) have a solution with $v'(r,t) = v(r,t) + v_\Delta(r,t)$. A way to investigate the existence of such solutions to such Sturm–Liouville boundary value problems is within the following framework.

Let us first introduce the bounded open domain $\Omega \subset \mathbb{R}^3$ with piecewise $\mathcal{C}^1$ boundary $\partial\Omega$. For the real Hilbert space $L^2(\Omega)$ with scalar product denoted as $\langle\cdot,\cdot\rangle_2$ and norm $\|\cdot\|_2$ we define the Sobolev space [5]

$$W^{1,2}(\Omega) = \{u \in L^2(\Omega) |\ \partial_j u \in L^2(\Omega),\ j = 1,\ldots,3\} \tag{18}$$

with norm $\|u\|_{1,2}^2 = \|u\|_2^2 + \|\nabla u\|_2^2$. Here the partial derivatives $\partial_j u$ are understood in the distributional sense. Further the real Hilbert space $H_0^1(\Omega)$ is defined to be the closure in $W^{1,2}(\Omega)$ of the infinitely differentiable functions compactly supported in $\Omega$. Hence, functions $u \in H_0^1(\Omega)$ fulfill the boundary condition $u = 0$ on $\partial\Omega$ naturally. The previously considered Sturm–Liouville problem (15) in shorthand notation reads as

$$\nabla \cdot (n\nabla v) = \zeta \tag{19}$$

and defines a bilinear form $Q$ on $H_0^1(\Omega)$ by

$$Q(u, v) = \langle u, -\nabla \cdot (n\nabla v)\rangle_2 = \langle \nabla u, n\nabla v\rangle_2, \tag{20}$$

where we used integration by parts and that functions in $H_0^1(\Omega)$ vanish at the border. We take advantage of the fact that (19) has a (weak) solution $v \in H_0^1(\Omega)$ if, and only if, $Q(u, v) = -\langle u, \zeta\rangle_2$ for all $u \in H_0^1(\Omega)$. This immediately leads us to the necessity $\zeta \in L^2(\Omega)$. Now the answer to the question of solvability is at hand with the theorem of Lax–Milgram [16].

**Theorem 3** (Lax–Milgram)**.** *Let $Q$ be a coercive continuous bilinear form on a Hilbert space $\mathcal{H}$. Then for every continuous linear functional $f$ on $\mathcal{H}$, there exists a unique $u_f \in \mathcal{H}$ such that*

$$Q(u, u_f) = f(u) \tag{21}$$

*holds for all $u \in \mathcal{H}$.*

A bilinear form $Q$ is said to be coercive if there exists a constant $c > 0$ such that $Q(u, u) \geqslant c\|u\|^2$ for all $u \in \mathcal{H}$. In our case, this can be established by means of the Poincaré inequality

$$\|u\|_2 \leqslant \lambda\|\nabla u\|_2, \quad \forall u \in H_0^1(\Omega), \tag{22}$$

where $0 < \lambda = \lambda(\Omega) < \infty$. As an additional assumption, we add that $n$ is bounded by a constant $m > 0$ almost everywhere on $\Omega$ from below. Then

$$Q(u, u) = \langle \nabla u, n\nabla u\rangle_2 \geqslant m\|\nabla u\|_2^2 \tag{23a}$$

$$\lambda^2 Q(u, u) \geqslant \lambda^2 m\|\nabla u\|_2^2 \geqslant m\|u\|_2^2. \tag{23b}$$

Combination of these results yields

$$Q(u, u) \geqslant \frac{m}{1+\lambda^2}\left(\|u\|_2^2 + \|\nabla u\|_2^2\right) = \frac{m}{1+\lambda^2}\|u\|_{1,2}^2. \tag{24}$$

We thus have established the coercivity of $Q$. As for the continuity we add another assumption on $n$ that is boundedness from above by a constant $M > 0$ almost everywhere on $\Omega$:

$$\begin{aligned} |Q(u, v) - Q(u_0, v)| &= |Q(u - u_0, v)| = |\langle \nabla(u - u_0), n\nabla u\rangle_2| \\ &\leqslant M\|\nabla(u - u_0)\|_2 \cdot \|\nabla v\|_2 \leqslant M\|u - u_0\|_{1,2} \cdot \|v\|_{1,2} < \infty. \end{aligned} \tag{25}$$

These restrictions on $n$ also imply that the differential operator defined by the left-hand side of (19) is elliptic. If $n$ can be assumed to be continuous on the closed domain $\bar{\Omega}$, then $n$ also attains its extremal values on $\bar{\Omega}$ and the restrictions reduce to the form

$$0 < n < \infty \quad \text{on } \bar{\Omega}. \tag{26}$$

Now everything left to show for the application of theorem 3 is that the right-hand side $-\langle \cdot, \zeta\rangle_2$ is indeed a continuous linear functional on the real Hilbert space $H_0^1(\Omega)$. This is easily established by considering for arbitrary $u, u_0 \in H_0^1(\Omega)$

$$|\langle u, \zeta\rangle_2 - \langle u_0, \zeta\rangle_2| = |\langle u - u_0, \zeta\rangle_2| \leqslant \|u - u_0\|_2 \cdot \|\zeta\|_2 \leqslant \|u - u_0\|_{1,2} \cdot \|\zeta\|_2 < \infty. \tag{27}$$

We subsume our results for the solvability of the Sturm–Liouville problem (19) in the following corollary.

**Corollary 1.** *Consider the Sturm–Liouville problem* $\nabla \cdot (n\nabla v) = \zeta$ *on the bounded open domain* $\Omega \subset \mathbb{R}^3$ *with piecewise* $\mathcal{C}^1$ *boundary. Let* $\zeta \in L^2(\Omega)$ *and* $n : \Omega \to \mathbb{R}$ *be almost everywhere bounded by* $0 < m \leqslant n \leqslant M < \infty$. *Then there exists a unique solution* $v \in H_0^1(\Omega)$.

Looking back to the original formulation of the Sturm–Liouville problem (15) we note that if $n^{(0)}$ fulfills the prerequisites of $n$ in corollary 1 and the right-hand side is indeed in $L^2(\Omega)$, then all orders of the potential $v_\Delta^{(k)}$ are uniquely defined in $H_0^1(\Omega)$. We now want to examine which constraints on the higher orders $n^{(k)}$ are sufficient for $\zeta^{(k)} \in L^2(\Omega)$. For this we assume $q^{(k)}$ and $q'^{(k)}$ already in $L^2(\Omega)$ for all $k$ and thus we get a unique $v_\Delta^{(0)} \in H_0^1(\Omega)$ trivially by corollary 1. For $k > 1$ we apply inductive reasoning: let us assume $v_\Delta^{(l)} \in H_0^1(\Omega)$ be given uniquely for $l < k$. Then (15) in a shorter notation reads as

$$\nabla \cdot \left(n^{(0)} \nabla v_\Delta^{(k)}\right) = q^{(k)} - q'^{(k)} - \sum_{l=0}^{k-1} \binom{k}{l} \nabla \cdot \left(n^{(k-l)} \nabla v_\Delta^{(l)}\right). \tag{28}$$

The (elliptic) differential operator on the left defines the same coercive continuous bilinear form on $H_0^1(\Omega)$ as before, thus everything to show for an application of theorem 3 is that the right-hand side yields a continuous linear functional. The sum of such functionals is again linear continuous; therefore, we can examine all terms separately. For $q^{(k)}$ and $q'^{(k)}$ the same reasoning as in (27) applies. Finally with $u, u_0 \in H_0^1(\Omega)$ continuity of the separate terms of the sum is established by

$$\begin{aligned} &\left|\left\langle u, \nabla \cdot \left(n^{(k-l)} \nabla v_\Delta^{(l)}\right)\right\rangle_2 - \left\langle u_0, \nabla \cdot \left(n^{(k-l)} \nabla v_\Delta^{(l)}\right)\right\rangle_2\right| \\ &\quad = \left|\left\langle \nabla(u - u_0), n^{(k-l)} \nabla v_\Delta^{(l)}\right\rangle_2\right| \leqslant \|\nabla(u - u_0)\|_2 \cdot \left\|n^{(k-l)} \nabla v_\Delta^{(l)}\right\|_2 \\ &\quad \leqslant \|u - u_0\|_{1,2} \cdot \left\|n^{(k-l)} \nabla v_\Delta^{(l)}\right\|_2 < \infty \quad \text{if} \quad n^{(k-l)} \nabla v_\Delta^{(l)} \in L^2(\Omega). \end{aligned} \tag{29}$$

By induction, we know that $v_\Delta^{(l)} \in H_0^1(\Omega)$ and thus $\nabla v_\Delta^{(l)} \in L^2(\Omega)$ so $n^{(k)}$ bounded almost everywhere by some $M_k > 0$ for all $k$ is a sufficient condition for theorem 3 to be applied. We subsume this in a second corollary.

**Corollary 2.** *Consider the system of Sturm–Liouville problems (15) on the bounded open domain* $\Omega \subset \mathbb{R}^3$ *with piecewise* $\mathcal{C}^1$ *boundary. Let* $q^{(k)}, q'^{(k)} \in L^2(\Omega)$ *and* $n^{(k)} : \Omega \to \mathbb{R}$ *be almost everywhere bounded by* $n^{(k)} \leqslant M_k$ *with* $M_k > 0$ *for all* $k$ *and additionally* $n^{(0)} \geqslant m > 0$ *almost everywhere. Then there exists a unique sequence of solutions* $v_\Delta^{(k)} \in H_0^1(\Omega)$.

If we turn to the problem of a classical solution, we refer the reader to the Weyl lemma as given in [17, 18] in various forms. (In a more general formulation, it can be found as the 'fundamental theorem on weak solutions' in [19].) There the operator $\hat{K} v = \nabla \cdot (n \nabla v)$ has domain

$$\mathrm{dom}(\hat{K}) = \{u | u \in \mathcal{C}^1(\bar{\Omega}), u \in \mathcal{C}^2(\Omega), \hat{K} u \in L^2(\Omega); u = 0 \text{ on } \partial\Omega\}. \tag{30}$$

**Theorem 4.** *For* $n > 0$ *on* $\bar{\Omega}$ *and* $n \in \mathcal{C}^3(\bar{\Omega})$ *and the boundary condition* $v = 0$ *on* $\partial\Omega$, *the equation*

$$\nabla \cdot (n \nabla v) = \zeta \tag{31}$$

*has a classical solution* $v \in \mathrm{dom}(\hat{K})$ *if* $\zeta \in \mathcal{C}^1(\bar{\Omega})$ *or* $\zeta$ *is Hölder continuous, i.e.* $|\zeta(r) - \zeta(r')| \leqslant h|r - r'|^\alpha$ *for all* $r, r' \in \bar{\Omega}$ *with* $h$ *and* $0 < \alpha < 1$ *independent of* $r, r'$.

Whether $\zeta^{(k)}$ in our actual problem (15) fulfills one of the conditions for a weak or classical solution, respectively, depends on the properties of the initial configurations and on the interactions under consideration. The terms $q^{(k)}$ and $q'^{(k)}$ implicate already for $k = 0$ spatial

partial derivatives of order 4 and spatial partial derivatives of the involved interaction potential of order 3. Hence, to have a well-defined Sturm–Liouville problem, the wavefunctions of the initial configurations and the interaction potentials have to fulfill certain restrictions with respect to their spatial behavior.

## 4. Sets of potentials and $v$-representable densities

We will introduce the set of external potentials for the extended Runge–Gross theorem in accordance with the classical Sturm–Liouville theory. Therefore, the defined sets will only be subsets of the actual sets of $v$-representable densities and potentials connected via the extended Runge–Gross theorem. Further, we will restrict our considerations to the above-introduced domain $\Omega \subset \mathbb{R}^3$. We assume the boundary to be far away from the center of the system such that it will not influence the dynamics. One has to be careful at this point as we will assume the initial one-particle density to be nonzero on $\bar{\Omega}$, hence also on the boundary $\partial\Omega$. This is different to the usual notion of a physical system restricted to a finite region, where one assumes an infinite boundary potential to restrict the wavefunction to this domain. In this case, the wavefunction and thus the one-particle potential will be zero at the boundary.

For a free Hamiltonian $\hat{H}_0 = \hat{T} + \hat{V}_{\text{int}}$ assumed self-adjoint and an initial configuration $\hat{\rho}_0$ at time $t = t_0$ we have

$$\begin{aligned}\mathcal{V}(\hat{\rho}_0, \hat{V}_{\text{int}}) := \{v \mid v \text{ analytic about } t = t_0, v(t) \in \text{dom}(\hat{K}),\\ v \text{ real; } n[v] \text{ analytic about } t = t_0 \text{ for } \hat{\rho}_0 \text{ and } \hat{V}_{\text{int}}\}.\end{aligned} \tag{32}$$

Here $n[v]$ is the time-dependent density, defined via the propagation of the initial configuration $\hat{\rho}_0$ with the Hamiltonian $\hat{H}(t) = \hat{T} + \hat{V}_{\text{int}} + \hat{V}([v]; t)$. It is straightforward to proof self-adjointness of this Hamiltonian by application of the Kato perturbation theory [5] as one can use $v(t) \in L^\infty(\Omega)$. Further we define the set of $v$-representable variations by

$$\begin{aligned}\delta\mathcal{N}(\hat{\rho}_0, \hat{V}_{\text{int}}) := \left\{ \delta n \;\middle|\; \delta n(r,t) = \sum_{k=2}^{\infty} \frac{1}{k!} n^{(k)}([v]; r)(t - t_0)^k \text{ for } \hat{\rho}_0 \text{ and } \hat{V}_{\text{int}},\right.\\ \left. v \in \mathcal{V}(\hat{\rho}_0, \hat{V}_{\text{int}}) \right\}.\end{aligned} \tag{33}$$

The set of $v$-representable densities is an affine set

$$\mathcal{N}(\hat{\rho}_0, \hat{V}_{\text{int}}) := n^{(0)}(r) + n^{(1)}(r,t) + \delta\mathcal{N}(\hat{\rho}_0, \hat{V}_{\text{int}}), \tag{34}$$

where $n^{(0)}(r) = n(r, t_0)$ is the initial density and $n^{(1)}(r,t) = \text{tr}(\hat{\rho}_0 \nabla \cdot \hat{j}(r))(t - t_0)$ in accordance with (13*a*) and (13*b*), respectively. For these sets, we then have in accordance with the Runge–Gross theorem an invertible mapping

$$\begin{aligned}v_{\hat{\rho}_0} : \mathcal{N}(\hat{\rho}_0, \hat{V}_{\text{int}}) &\to \mathcal{V}(\hat{\rho}_0, \hat{V}_{\text{int}})\\ n(r,t) &\mapsto v_{\hat{\rho}_0}([n]; r, t)\end{aligned} \tag{35}$$

connecting the $v$-representable one-particle densities with the external potentials. Nevertheless, if we now define a second mapping for a different initial configuration $\hat{\rho}_0'$ subject to the conditions (13*a*) and (13*b*), and a different interaction $\hat{V}_{\text{int}}'$, we do not know if $n$ is simultaneously element in $\mathcal{N}(\hat{\rho}_0, \hat{V}_{\text{int}})$ and $\mathcal{N}(\hat{\rho}_0', \hat{V}_{\text{int}}')$. This, however, is of fundamental importance if we want a rigorous formulation of the time-dependent Kohn–Sham scheme.

To achieve this goal we introduce further restrictions. We will assume smooth interactions and initial configurations in what follows. This excludes the usual Coulombic interaction as

it is not infinitely differentiable at the origin. However, one may regularize the Coulombic interaction by a so-called soft-core interaction, i.e. by replacing $|r| \to \sqrt{r^2 + \epsilon}$ and $\epsilon > 0$.

Due to equation (10) we have a direct connection between $v$-representable densities and potentials. Hence, we can formulate the following lemma.

**Lemma 1.** *Let $\hat{\rho}_0$ be chosen such that all its wavefunctions are in $\mathcal{C}^\infty(\bar{\Omega}^N)$, $v$ Taylor expandable about $t = t_0$, $v^{(k)} \in \mathcal{C}^\infty(\bar{\Omega})\ \forall\ k$, and $v_{\text{int}}(|r - r'|)$ infinitely differentiable. Then $n^{(k)}(r) \in \mathcal{C}^\infty(\bar{\Omega})$ for all $k$.*

**Proof.** We will use equation (10). Obviously we have $n^{(0)}(r)$ and $n^{(1)}(r)$ in $\mathcal{C}^\infty(\bar{\Omega})$. Thus $n^{(2)}(r)$ is in $\mathcal{C}^\infty(\bar{\Omega})$ if $q^{(0)}(r)$ is infinitely differentiable, where

$$q^{(0)}(r) = \text{tr}[\hat{\rho}_0\, \hat{q}(r)]. \tag{36}$$

$\hat{q}(r)$ consists of partial derivatives with respect to $r$ and of derivatives of $v_{\text{int}}(|r - r'|)$. We have assumed $v_{\text{int}}(|r - r'|)$ infinitely differentiable. Hence, we have $q^{(0)}(r) \in \mathcal{C}^\infty(\bar{\Omega})$. For $n^{(3)}(r)$ we need to know $q^{(1)}(r)$. This is the commutator of $\hat{q}(r)$ with $\hat{H}(t)$ at $t = t_0$. All functions in $\hat{H}(t)$ are infinitely differentiable. Again the above reasoning applies, and we find $q^{(1)}(r) \in \mathcal{C}^\infty(\bar{\Omega})$. All higher terms are to be found via successive application of the Heisenberg equation for $\hat{q}(r)$ with $\hat{H}(t)$ at $t = t_0$. The only difference to the above reasoning is the appearance of $v^{(k)}(r)$-terms, which are again infinitely differentiable. Therefore one can successively construct all $n^{(k)}(r) \in \mathcal{C}^\infty(\bar{\Omega})$. □

Now we introduce the restricted set of smooth one-particle potentials

$$\mathcal{V}^*(\hat{\rho}_0, \hat{V}_{\text{int}}) = \{v \mid v \in \mathcal{V}(\hat{\rho}_0, \hat{V}_{\text{int}}),\ v(t) \in \mathcal{C}^\infty(\bar{\Omega})\}, \tag{37}$$

and by lemma 1 the corresponding smooth $v$-representable one-particle densities

$$\mathcal{N}^*(\hat{\rho}_0, \hat{V}_{\text{int}}) = \left\{ n \;\middle|\; n(r,t) = \sum_{k=0}^{\infty} \frac{1}{k!} n^{(k)}([v]; r)(t - t_0)^k \text{ for } \hat{\rho}_0 \text{ and } \hat{V}_{\text{int}}, v \in \mathcal{V}^*(\hat{\rho}_0, \hat{V}_{\text{int}}) \right\}. \tag{38}$$

With this we can reformulate the extended Runge–Gross theorem as follows.

**Theorem 5.** *Let $\hat{\rho}_0$ and $\hat{V}_{\text{int}}$ be infinitely differentiable, $n(r,t) \in \mathcal{N}^*(\hat{\rho}_0, \hat{V}_{\text{int}})$ and $v_{\hat{\rho}_0}([n]; r, t) = v(r,t) \in \mathcal{V}^*(\hat{\rho}_0, \hat{V}_{\text{int}})$ the associated external potential. For a system with infinitely differentiable interaction $\hat{V}'_{\text{int}}$ and the initial configuration $\hat{\rho}'_0$ consisting of infinitely differentiable functions subject to the constraint*

$$n(r, t_0) = n^{(0)}(r) = n'(r, t_0) > 0, \tag{39}$$

$$\text{tr}\,(\hat{\rho}_0\, \nabla \cdot \hat{\jmath}(r)) = \text{tr}(\hat{\rho}'_0\, \nabla \cdot \hat{\jmath}(r)), \tag{40}$$

*there exists a unique effective potential depending on both initial configurations*

$$v_{\hat{\rho}_0, \hat{\rho}'_0}([n]; r, t) = \sum_{k=0}^{\infty} \frac{1}{k!}\ v_\Delta^{(k)}(r)\ \ (t - t_0)^k, \tag{41}$$

*where $v_\Delta^{(k)}(r)$ is defined via*

$$\nabla \cdot \left[ n^{(0)}(r) \nabla v_\Delta^{(k)}(r) \right] = q^{(k)}(r) - q'^{(k)}(r) - \sum_{l=0}^{k-1} \binom{k}{l} \nabla \cdot [n'^{(k-l)}(r) \nabla v'^{(k)}(r)], \tag{42}$$

*with $v' = (v + v_{\hat{\rho}_0, \hat{\rho}'_0}) \in \mathcal{V}^*(\hat{\rho}'_0, \hat{V}'_{\text{int}})$ generating the same density. It holds that*

$$\mathcal{N}^*(\hat{\rho}_0, \hat{V}_{\text{int}}) = \mathcal{N}^*(\hat{\rho}'_0, \hat{V}'_{\text{int}}) = \mathcal{N}^*(n^{(0)}, n^{(1)}). \tag{43}$$

**Proof.** From the proof of lemma 1, we know that all $q^{(k)}$ and $q'^{(k)}$ are infinitely differentiable. As we have assumed $n_0(r) > 0$ we can apply theorem 4 from which it is clear that

$$\nabla \cdot [n^{(0)}(r)\nabla v'^{(k)}(r)] = q^{(k)}(r) - q'^{(k)}(r) - \sum_{l=0}^{k-1} \binom{k}{l} \nabla \cdot [n'^{(k-l)}(r)\nabla v'^{(k)}(r)] \tag{44}$$

has an existing solution for $k = 0$ if the right-hand side is $\mathcal{C}^1(\bar{\Omega})$. Obviously $v'^{(0)}(r)$ exists due to theorem 4 and is infinitely differentiable. In the next step, we can use $v'^{(0)}(r)$ in the Sturm–Liouville equation defining $v'^{(1)}(r)$. Again existence is guaranteed and we have $v'^{(1)}(r) \in \mathcal{C}^\infty(\bar{\Omega})$ [20]. One can now successively construct $v_{\hat{\rho}_0,\hat{\rho}'_0}$. Then $(v + v_{\hat{\rho}_0,\hat{\rho}'_0})$ is given via its Taylor series within its radius of convergence in accordance with the extended Runge–Gross proof [10]. This construction holds for every $n \in \mathcal{N}^*(\hat{\rho}_0, \hat{V}_{\text{int}})$, and we have $n \in \mathcal{N}^*(\hat{\rho}'_0, \hat{V}'_{\text{int}})$ as well. Hence, the set of $v$-representable densities does not depend on the smooth interaction or on the smooth initial configuration. □

Here it became obvious why we restricted our considerations to infinitely differentiable initial configurations and potentials. With these assumptions we can guarantee the existence of all the classical Sturm–Liouville boundary value problems on $\Omega$. For the general, i.e. weak case, we need to make sure that all $q^{(k)}$, $q'^{(k)}$ and hence $\zeta^{(k)}$ are in $L^2(\Omega)$, in order to proof the existence of a solution using corollary 2.

The special case of a rigorous Kohn–Sham theorem is straightforward as $\hat{V}_{\text{int}} \equiv 0$ is of course infinitely differentiable. One finds that for the above restrictions all interacting-$v$-representable densities are noninteracting-$v$-representable because $\mathcal{N}^*(\hat{\rho}_0, \hat{V}_{\text{int}}) = \mathcal{N}^*(\hat{\rho}'_0, 0)$. Only a noninteracting initial configuration is needed. The condition of $n^{(0)}(r) > 0$ for the existence of the effective potential may be relaxed if there exists some time $t_1$ in a sufficiently small neighborhood of $t_0$ for which $n(r, t_1) > 0$. Then we could use $\hat{\rho}(t_1)$ as a new initial state and prove existence at that time provided we also have the corresponding $\hat{\rho}'(t_1)$.

## 5. Conclusion

Under certain assumptions, we can state sufficient constraints on the one-particle density such that the existence of the effective potential, possibly in the weak sense, is guaranteed. However, only for classical solutions of the corresponding Sturm–Liouville boundary value problems we can reformulate the extended Runge–Gross theorem such that existence of the effective potentials is granted. As long as we consider smooth initial states and smooth interactions the Kohn–Sham system exactly reproduces the physical one-particle density. In general, as pointed out in [9], it seems safe to assume the existence of the Kohn–Sham potential for physical systems. Nevertheless, a rigorous proof of principle is of importance for the foundations of the theory.

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## References

[1] Kohn W 1999 *Rev. Mod. Phys.* **71** 1253
[2] Dreizler R M and Gross E K U 1990 *Density Functional Theory—An Approach to the Quantum Many-Body Problem* (Berlin: Springer)
[3] Hohenberg P and Kohn W 1964 *Phys. Rev.* B **136** 864

[4] Lieb E H 1983 *Int. J. Quantum Chem.* **24** 243
[5] Blanchard P and Brüning E 2003 *Mathematical Methods in Physics: Distributions, Hilbert Space Operators and Variational Methods* (Boston, MA: Birkhäuser)
[6] Kohn W and Sham L J 1965 *Phys. Rev.* **140** A1133
[7] Runge E and Gross E K U 1984 *Phys. Rev. Lett.* **52** 997
[8] Gross E K U *et al* 1995 Density-functional theory of time-dependent systems *Density Functional Theory, NATO ASI Series B* ed R Dreizler *et al* (New York: Plenum)
[9] van Leeuwen R 2001 *Int. J. Mod. Phys.* B **15** 1969
[10] van Leeuwen R 1999 *Phys. Rev. Lett.* **82** 3863
[11] Marques M A L *et al* 2006 *Time-Dependent Density Functional Theory (Lect. Notes Phys.* vol 706*)* (Heidelberg: Springer)
[12] Fetter A L and Walecka J D 2003 *Quantum Theory of Many-Particle Systems* (New York: Dover)
[13] Tokatly I V 2005 *Phys. Rev.* B **71** 165104
[14] Maitra N T, van Leeuwen R and Burke K 2008 *Phys. Rev.* A **78** 056501
[15] Schirmer J and Dreuw A 2008 *Phys. Rev.* A **78** 056502
[16] Blanchard P and Brüning E 1992 *Variational Methods in Mathematical Physics* (Berlin: Springer)
[17] Hellwig G 1964 *Differentialoperatoren der Mathematischen Physik* (Berlin: Springer)
[18] Hellwig G 1960 *Partielle Differentialgleichungen* (Stuttgart: Teubner)
[19] Maurin K 1972 *Methods of Hilbert Spaces* (Warszawa: PWN–Polish Scientific Publishers)
[20] Agmon S 1965 Theorem 6.6 *Lectures on Elliptic Boundary Value Problems* (Princeton, NJ: Van Nostrand)