Consistency of the Local Density Approximation for Time Dependent Closed Quantum Systems

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Abstract

Time dependent quantum systems are the subject of intense inquiry, in mathematics, science, and engineering, particularly at the atomic and molecular levels. In 1984, Runge and Gross introduced time dependent density functional theory (TDDFT). We have previously investigated such systems on bounded domains for Kohn-Sham potentials by use of evolution operators and fixed point theorems. In this article, motivated by usage in the physics community, we consider local power function approximations for the exchange-correlation potential component. By smoothing these so-called local density approximations, we are able to demonstrate that the resulting model has a unique weak solution on any given finite time interval, for each value of the smoothing parameter, via the above-cited theory. Compactness arguments allow the extraction of an appropriately convergent subsequence, with a limit defining a weak solution. Uniqueness remains an open question, however. In summary, we are able to demonstrate a weak solution, on an arbitrary time interval, for local charge density approximations typically used in numerical simulations of the model. For these approximations, we permit both positively and negatively signed potentials, with differing assumptions.
1 Introduction

In previous work [1, 2], we analyzed closed quantum systems on bounded domains of $\mathbb{R}^3$ via time-ordered evolution operators. The article [1] demonstrated strong $H^2$ solutions, whereas the article [2] demonstrated weak solutions; [2] also includes the exchange-correlation component of the Hamiltonian potential not included in [1]. The model we study, time dependent density functional theory (TDDFT), is a significant field for applications, including computational nanoelectronics and chemical physics [3]. It was originally introduced by E. Runge and E.K.U. Gross in [4]; an exposition of the subject may be found in [5]. By permitting time dependent potentials, TDDFT extends the nonlinear Schrödinger equation, which has been studied extensively [6, 7].

The existence/uniqueness theory of [2] precludes the direct use of a local density approximation (LDA) to the exchange correlation potential, such as those frequently used in simulations of the model. A physics perspective of this approximation may be found in [5]. In this article, we permit local positively or negatively signed power function representations, though with different hypotheses. As we demonstrate below, smoothing of such local approximations provides a model within the framework of [2]. By using compactness arguments suggested in [7], we are able to obtain a solution of the originally posed LDA model. Uniqueness is not established, however. The article [8] considers a model with Kohn-Sham potentials, with some overlap to that considered here, which is studied in connection with optimal control. A Faedo-Galerkin method is used to establish existence. The use of evolution operators and smoothing as presented here is consistent with techniques in the applied literature [3] and provides direct support for successive approximation and other numerical procedures [9, 10].

In the following subsections of the introduction, we summarize the basic results of [2], as a starting point for the present article. In section two, we formulate the new model, which incorporates the local density approximation, and we prove that its smoothed version lies within the scope of [2]. In section three, we introduce the compactness arguments, and establish existence of a weak solution as the limit of solutions of the smoothed model.

1.1 The model

In its original form, TDDFT includes three components for the potential: an external potential, the Hartree potential, and a general non-local term representing the exchange-correlation potential, which is assumed to include a time history part. If $\hat{H}$ denotes the Hamiltonian operator of the system, then the state $\Psi(t)$ of the system obeys the nonlinear Schrödinger equation,

$$i\hbar \frac{\partial \Psi(t)}{\partial t} = \hat{H}\Psi(t).$$  (1)
The Local Density Approximation

Here, $\Psi = \{\psi_1, \ldots, \psi_N\}$ consists of $N$ orbitals, and the charge density $\rho$ is defined by

$$\rho(x, t) = |\Psi(x, t)|^2 = \sum_{k=1}^N |\psi_k(x, t)|^2.$$  

An initial condition,

$$\Psi(0) = \Psi_0,$$  

and boundary conditions are included. The particles are confined to a bounded region $\Omega \subset \mathbb{R}^3$ and homogeneous Dirichlet boundary conditions hold within a closed system. $\Psi$ denotes a finite vector function of space and time. The effective potential $V_e$ is a real scalar function of the form,

$$V_e(x, t, \rho) = V(x, t) + W * \rho + \Phi(x, t, \rho).$$

Here, $W(x) = 1/|x|$ and the convolution $W * \rho$ denotes the Hartree potential. If $\rho$ is extended as zero outside $\Omega$, then, for $x \in \Omega$,

$$W * \rho(x) = \int_{\mathbb{R}^3} W(x - y) \rho(y) \, dy,$$

which depends only upon values $W(z), \|z\| \leq \text{diam}(\Omega)$. We may redefine $W$ smoothly outside this set, so as to obtain a function of compact support for which Young’s inequality applies. $\Phi$ represents a time history of $\rho$:

$$\Phi(x, t, \rho) = \Phi_0(x, 0, \rho) + \int_0^t \phi(x, s, \rho) \, ds.$$  

The Hamiltonian operator is given by,

$$\hat{H} = -\frac{\hbar^2}{2m} \nabla^2 + V(x, t) + W * \rho + \Phi(x, t, \rho),$$

and $m$ designates the effective mass and $\hbar$ the normalized Planck’s constant.

1.2 Definition of weak solution

The solution $\Psi$ is continuous from the time interval $J$, to be defined shortly, into the finite energy Sobolev space of complex-valued vector functions which vanish in a generalized sense on the boundary, denoted $H^1_0(\Omega)$: $\Psi \in C(J; H^1_0)$. The time derivative is continuous from $J$ into the dual $H^{-1}$ of $H^1_0$: $\Psi \in C^1(J; H^{-1})$. The spatially dependent test functions $\zeta$ are arbitrary in $H^1_0$. The duality bracket is denoted $\langle f, \zeta \rangle$. Norms and inner products are discussed in the appendix.

**Definition 1.1.** For $J = [0, T]$, the vector-valued function $\Psi = \Psi(x, t)$ is a weak solution of (1, 2, 3) if $\Psi \in C(J; H^1_0(\Omega)) \cap C^1(J; H^{-1}(\Omega))$, if $\Psi$ satisfies the initial condition $\Psi_0$ for $\Psi_0 \in H^1_0$, and if $\forall 0 < t \leq T$:

$$i\hbar \frac{\partial \Psi(t)}{\partial t}, \zeta = \int_{\Omega} \frac{\hbar^2}{2m} \nabla \Psi(x, t) \cdot \nabla \zeta(x) + V_e(x, t, \rho) \Psi(x, t) \zeta(x) \, dx.$$  


1.3 Hypotheses for the Hamiltonian and theorem statement

We assume the following.

• \( \Phi \) is continuous in \( t \) on \( H^1 \) and bounded in \( t \) into \( W^{1,3} \). By boundedness, we mean that every fixed ball in has image under \( \Phi(\cdot, t, \rho) \) which lies in a fixed ball in \( W^{1,3} \), independent of \( t \).

• The derivative \( \frac{\partial \Phi}{\partial t} = \phi \) is assumed measurable, and bounded in its arguments.

• Furthermore, the following smoothing condition is assumed, expressed in a (uniform) Lipschitz norm condition: \( \forall t \in [0, T], \) if \( r \) is a bound for \( \| \Psi_j \|_{H^1}, j = 1, 2, \)

\[
\| [\Phi(t, |\Psi_1|^2) - \Phi(t, |\Psi_2|^2)]\psi \|_{H^1} \leq C(r) \| \Psi_1 - \Psi_2 \|_{H^1} \| \psi \|_{H^1_0}. \tag{5}
\]

Here, \( \psi \) is arbitrary in \( H^1_0 \) and \( C(r) \) depends only on \( r \).

• The so-called external potential \( V \) is assumed to be continuously differentiable on the closure of the space-time domain.

The following theorem was proved in [2], based upon the evolution operator as presented in [11], and will provide a solution for the smoothed problem on \( J \) as introduced in the following section.

**Theorem 1.1.** For any interval \( [0, T] \), the system (4) in Definition 1.1, with Hamiltonian defined by (3), has a unique weak solution if the hypotheses of section 1.3 hold.

2 The Local Density Approximation

In this section, we consider the local density approximation to the exchange-correlation potential \( \Phi \).

**Definition 2.1.** We consider the following approximation, where \( \lambda \) is a real constant, positive or negative.

\[
\Phi_{lda}(\rho) = \lambda \rho^{\alpha/2} = \lambda |\Psi|^\alpha. \tag{6}
\]

Additionally,

• If \( \lambda > 0 \), the range of \( \alpha \) is \( 1 \leq \alpha < 4 \).

• If \( \lambda < 0 \), the range of \( \alpha \) is \( 1 \leq \alpha \leq 4/3 \). We require additionally that \( |\lambda| \) is sufficiently small (as specified in Remark 3.1).
We redefine the Hamiltonian as

\[ \hat{H} = -\frac{\hbar^2}{2m} \nabla^2 + V(x, t) + W \ast \rho + \Phi_{\text{lda}}(\rho). \]  

(7)

The following theorem is the goal of our analysis.

**Theorem 2.1.** If the effective potential is redefined by

\[ V_e(x, t, \rho) = V(x, t) + W \ast \rho + \Phi_{\text{lda}}(\rho), \]

(8)

then there is a weak solution of (4). Here, only the final hypothesis of section 1.3 together with those of Definition 2.1 are assumed. The solution is in the regularity class \( C(J; H^1_0) \cap C^1(J; H^{-1}) \) and satisfies the specified initial condition.

**Remark 2.1.** The techniques of this article allow for the immediate extension of the term defined in (6) to the sum of such terms, with coefficients of arbitrary sign, as defined in Definition 2.1.

The proof of Theorem 2.1 is carried out in section 3 (see Theorems 3.1 and 3.2).

### 2.1 The smoothing

We begin by defining a standard convolution [12].

**Definition 2.2.** Suppose that a nonnegative function \( \phi_1 \) is given, \( \phi_1 \in C_0^\infty(\mathbb{R}^3) \), of integral one. Set

\[ \phi_\epsilon(x) = \epsilon^{-3} \phi_1(x/\epsilon), \quad x \in \mathbb{R}^3, \]

and, for \( f \in L^p(\Omega), 1 \leq p < \infty, \)

\[ f_\epsilon = \phi_\epsilon \ast f. \]

We refer to \( \Phi_\epsilon \) as the smoothing of the local density approximation.

We recall [12] that \( \lim_{\epsilon \to 0} f_\epsilon = f \) in \( L^p \) and \( \|f_\epsilon\|_{L^p} \leq \|f\|_{L^p}, \forall \epsilon > 0. \)

### 2.2 Existence and uniqueness for the smoothed system

As mentioned in the introduction, we will show that the smoothed problem has a unique weak solution on \([0, T]\) for each fixed \( \epsilon > 0. \) We first state the result.

**Proposition 2.1.** If \( \Phi_{\text{lda}} \) is replaced by its smoothing \( \Phi_\epsilon \), then the hypotheses of section 1.3 hold, as applied to \( \Phi_\epsilon. \) In particular, Theorem 1.1 is applicable. There exists a unique weak solution \( \Psi_\epsilon \), as specified in Definition 1.1, of the corresponding system:

\[ i\hbar \left( \frac{\partial \Psi_\epsilon(t)}{\partial t}, \zeta \right) = \int_\Omega \frac{\hbar^2}{2m} \nabla \Psi_\epsilon(x, t) \cdot \nabla \zeta(x) + V_e(x, t, \rho_\epsilon) \Psi_\epsilon(x, t) \zeta(x) \, dx, \]

(9)

\[ V_e(x, t, \rho_\epsilon) = V(x, t) + W \ast \rho_\epsilon + \Phi_\epsilon(\rho_\epsilon). \]

(10)
Proof. We state the two properties required to be verified.

1. \( \Phi_\varepsilon \) maps sets bounded in \( H^1_0 \) into sets bounded in \( W^{1,\lambda} \).

2. The Lipschitz property \( \| \Phi_\varepsilon \|_{L^2} \) holds.

Before verifying properties (1) and (2), we note that there is no restriction on the size of \( |\lambda| \), and the range of \( \alpha \) is \( 1 \leq \alpha < 4 \), whatever the sign of \( \lambda \).

Property (1) is immediate from the inequalities,

\[
\| \Phi_\varepsilon (\rho) \|_{L^2} \leq |\lambda| \| \phi_\varepsilon \|_{L^3} \| \Psi_1^\alpha \|_{L^1}, \quad \| \nabla \Phi_\varepsilon (\rho) \|_{L^2} \leq |\lambda| \| \nabla \phi_\varepsilon \|_{L^2} \| \Psi_1^\alpha \|_{L^1},
\]

which follow from Young’s inequality, applied to the convolution which defines \( \Phi_\varepsilon \). Indeed, recall that \( \alpha < 4 \), so that the Sobolev inequality may be applied.

For the verification of property (2), we begin with the gradient term, and specifically with the product rule as applied to the definition of \( \Phi_\varepsilon /|\lambda| \):

\[
\| \nabla [(\phi_\varepsilon * |\Psi_1|^\alpha - \phi_\varepsilon * |\Psi_2|)\psi] \|_{L^2} = \\
\| \nabla \phi_\varepsilon * (|\Psi_1|\alpha - |\Psi_2|\alpha)\psi + \phi_\varepsilon * (|\Psi_1|\alpha - |\Psi_2|\alpha)\nabla \psi \|_{L^2}.
\]

(11)

We have used the differentiation property of the convolution. When the triangle inequality is employed, the second term is the more delicate to estimate since \( \nabla \psi \in L^2 \) (only). Thus, by use of the Schwarz inequality and Young’s inequality, we must estimate \( |||\Psi_1|\alpha - |\Psi_2|\alpha\|_{L^1} \). The case \( \alpha = 1 \) is immediate. We prepare for the cases \( 1 < \alpha < 4 \) by citing the following useful numerical inequality \( 13 \):

\[
\frac{\left( \frac{y^r - z^r s}{y^s - z^s r} \right)^{\frac{1}{\alpha - 1}}} \leq \max(y, z), \quad y \geq 0, z \geq 0, y \neq z, r > 0, s > 0, s \neq r.
\]

(12)

We apply \( 12 \) with the identifications,

\[
r = \alpha, s = 1, y = |\Psi_1|, z = |\Psi_2|,
\]

to obtain the pointwise estimate,

\[
| |\Psi_1|\alpha - |\Psi_2|\alpha| \leq \alpha (\max(|\Psi_1|, |\Psi_2|))^{\alpha - 1} \| (|\Psi_1| - |\Psi_2|). \]

(13)

Although we will require inequality \( 13 \) later in the article, it is more convenient here to use the less sharp inequality, derived from \( 13 \):

\[
| |\Psi_1|\alpha - |\Psi_2|\alpha| \leq \alpha (1 + |\Psi_1| + |\Psi_2|)\alpha \| |\Psi_1| - |\Psi_2|).
\]

We use a technique motivated by \( 17 \). If \( r = \alpha + 2 \), and \( r' \) is conjugate to \( r \), if \( p = r/r' \), and \( p' \) is conjugate to \( p \), then

\[
\alpha p' = r, \quad r' p = r,
\]

and an application of Hölder’s inequality gives

\[
|| |\Psi_1|\alpha - |\Psi_2|\alpha||_{L^r'} \leq \alpha || |\Psi_1| + |\Psi_2| ||_{L^r} \| |\Psi_1| - |\Psi_2| ||_{L'} \leq C || |\Psi_1 - \Psi_2||_{L^r}.
\]
An application of Sobolev’s inequality shows that the rhs of this inequality is dominated by a locally bounded constant times $\|\Psi_1 - \Psi_2\|_{H^1}$. Since the $L^1$ norm is dominated by a constant times the $L^{r'}$ norm, the estimation of the second term arising from (11) is completed. The first term also reduces to the estimation of $\|\Psi_1^\alpha - \Psi_2^\alpha\|_{L^1}$, as does the non-gradient term. Thus, the proof of property (2) is completed. It follows that a unique weak solution $\Psi_\epsilon$ exists for the smoothed system as formulated.

## 3 Existence for the LDA System

The results of this section are derived for an arbitrary time interval $[0, T]$. The compactness techniques are motivated by [7].

### 3.1 ‘A priori’ bounds for the smoothed solutions

We begin by quoting a result proved in [2], now applied to the family of solutions $\Psi_\epsilon$.

**Lemma 3.1.** If the functional $E(t)$ is defined for $0 < t \leq T$ by,

$$E(t) = \int_\Omega \left[ \frac{\hbar^2}{4m} |\nabla \Psi_\epsilon|^2 + \left( \frac{1}{4} (W * |\Psi_\epsilon|^2) + \frac{1}{2} (V + \Phi_\epsilon) \right) |\Psi_\epsilon|^2 \right] dx,$$

then the following identity holds:

$$E(t) = E(0) + \frac{1}{2} \int_0^t \int_\Omega (\partial V / \partial s)(x, s) |\Psi_\epsilon|^2 dxds,$$

where $E(0)$ is given by

$$\int_\Omega \left[ \frac{\hbar^2}{4m} |\nabla \Psi_0|^2 + \left( \frac{1}{4} (W * |\Psi_0|^2) + \frac{1}{2} (V(\cdot, 0) + (\Phi_\epsilon)(|\Psi_0|^2)) \right) |\Psi_0|^2 \right] dx.$$

The following proposition uses the hypothesis of Definition 2.1 for the case when $\lambda < 0$.

**Proposition 3.1.** There is a bound $r$ in the norm of $C(J; H^1)$ for the smoothed solutions. In fact, if $\lambda > 0$, the number $r^2$ can be chosen as any upper bound for

$$\left( \frac{4m}{\hbar^2} \right) \left[ \sup_{0 \leq t \leq T} E(t) + \sup_{x \in \Omega, t \leq T} \frac{|V|}{2} \|\Psi_0\|_{L^2}^2 \right] + \|\Psi_0\|_{L^2}^2,$$

which allows the choice,

$$r^2 = \left( \frac{4m}{\hbar^2} \right) \left[ E_0 + \left( \frac{1}{2} \right) \sup_{x \in \Omega, t \leq T} \left[ T |\partial V / \partial t| + |V| \right] \|\Psi_0\|_{L^2}^2 \right] + \|\Psi_0\|_{L^2}^2,$$

in this case. If $\lambda < 0$, there is a positive constant $C$ which may replace $\left( \frac{4m}{\hbar^2} \right)$ in both (16) and (17).
Proof. We use the lemma and the hypotheses on $V$ and $\Phi_{lda}$. If $\lambda > 0$, only $V$ and its time derivative require separate estimation. If $\lambda < 0$, in addition to these terms, we require the inequality,

$$
|\lambda| \int_{\Omega} \Phi_{e}(\rho_{e}) |\Psi_{e}|^{2} \leq |\lambda| \|\Psi_{e}^{\alpha}\|_{L^{3/2}} \|\Psi_{e}\|_{L^{6}},
$$

which follows from Young’s inequality and Hölder’s inequality. Since $3\alpha/2 \leq 2$, we may use the unitary bound of the initial condition times a power of $|\Omega|$ to bound the factor $\|\Psi_{e}\|_{L^{3/2}}$ (see the following remark for the precise inequality). Sobolev’s inequality is used to bound the term, $\|\Psi_{e}\|_{L^{5}}$, and, since $|\lambda|$ is sufficiently small, this entire term can be absorbed into the leading term of $E(t)$ to obtain, via reciprocals, the constant $C$.

Remark 3.1. In the preceding proof, we have used the inequality,

$$
|\lambda| \Omega_{0} < \left(\frac{\hbar^{2}}{4m}\right),
$$

where

$$
\Omega_{0} = C_{H^{3} \rightarrow L^{6}}^{2} \|\Psi_{0}\|_{L^{2}}^{\alpha} |\Omega|^{2/3 - \alpha/2}.
$$

Proposition 3.2. There is a uniform bound, in $t \in J$ and $\epsilon > 0$, for the norms,

$$
\|(\Psi_{e})_{t}\|_{H^{-1}}.
$$

Proof. One begins by using the weak form of the equation as discussed in Proposition 2.1 and isolating the time derivative acting on an arbitrary test function. The gradient term and the external potential term are bounded directly by the previous proposition. For the Hartree term, we estimate, by Hölder’s inequality and Young’s inequality,

$$
\left|\int_{\Omega} W \ast |\Psi_{e}|^{2} \Psi_{e} \zeta\right| \leq \|W\|_{L^{1}} \|\Psi_{e}\|_{L^{3}} \|\Psi_{e}\|_{L^{6}} \|\zeta\|_{L^{6}}.
$$

Sobolev’s inequality, combined with Proposition 3.1 gives the bound for this term. For the smoothed LDA term, the sign of $\lambda$ is not relevant and we consider $1 \leq \alpha < 4$. We estimate by Hölder’s inequality, for $r = \alpha + 2$ and $r'$ conjugate to $r$,

$$
\left|\int_{\Omega} \phi_{e} \ast |\Psi_{e}|^{\alpha} \Psi_{e} \zeta\right| \leq \|\phi_{e} \ast |\Psi_{e}|^{\alpha}\|_{L^{r'}} \|\zeta\|_{L^{r'}}.
$$

The LDA term requires additional explanation. We have, by another application of Hölder’s inequality, with $p = r/r'$ and $p'$ conjugate to $p$ (note that $r/\alpha = r'/p'$),

$$
\|\phi_{e} \ast |\Psi_{e}|^{\alpha}\|_{L^{r'}} \leq \|\phi_{e}\|_{L^{r}} \|\Psi_{e}\|_{L^{r}} \leq \|\Psi_{e}\|_{L^{r}} \|\Psi_{e}\|_{L^{r}} \leq \|\Psi_{e}\|_{L^{r}} \|\Psi_{e}\|_{L^{r}} \leq \|\Psi_{e}\|_{L^{r}}^{\alpha+1}.
$$

We conclude that the LDA term is bounded, as claimed. This completes the proof.
The following corollary is an immediate consequence of Propositions 3.1 and 3.2.

**Corollary 3.1.** Any sequence taken from the set $\{\Psi_\epsilon\}$ of solutions of the smoothed systems is bounded in the norms of $C(J; H^1_0)$ and $C^1(J; H^{-1})$.

### 3.2 Convergent subsequences

We begin by stating a basic lemma [7, Proposition 1.3.14, (i)].

**Lemma 3.2.** There is an element $\Psi \in L^\infty(J; H^1_0(\Omega)) \cap W^{1,\infty}(J; H^{-1}(\Omega))$, and a sequence $\Psi_\epsilon(n)$ satisfying the weak convergence property,

$$
\Psi_\epsilon(n)(t) \rightharpoonup \Psi(t), \text{ in } H^1_0, \text{ uniformly } \forall t \in J.
$$

**Proof.** The preceding corollary furnishes the necessary hypotheses. \qed

We divide the verification of Theorem 2.1 into two parts.

**Theorem 3.1.** The function $\Psi$ of Lemma 3.2 satisfies the TDDFT system discussed in Theorem 2.1 with the LDA approximation.

**Proof.** We begin with the observation that, by the compactness of the embedding, $H^1_0 \hookrightarrow L^r$, $1 \leq r < 6$, it follows that

$$
\Psi_\epsilon(n)(t) \rightarrow \Psi(t), \text{ in } L^r, \text{ uniformly } \forall t \in J,
$$

for all such $r$. It follows that $\Psi \in C(J; L^r)$. We now examine the equation satisfied by $\Psi$. By weak convergence,

$$
\lim_{n \rightarrow \infty} \int_\Omega \frac{\hbar^2}{2m} \nabla \Psi_\epsilon(n)(x, t) \cdot \nabla \zeta(x) \, dx = \int_\Omega \frac{\hbar^2}{2m} \nabla \Psi(x, t) \cdot \nabla \zeta(x) \, dx.
$$

We now consider each of the three cases required to verify that

$$
\lim_{n \rightarrow \infty} \int_\Omega V_{e}(x, t, \rho_\epsilon(n))\Psi_\epsilon(n)(x, t)\zeta(x) \, dx = \int_\Omega V_{e}(x, t, \rho)\Psi(x, t)\zeta(x) \, dx.
$$

By the boundedness of the external potential, and the strong convergence of the sequence, we conclude immediately that

$$
\lim_{n \rightarrow \infty} \int_\Omega V(x, t)\Psi_\epsilon(n)(x, t)\zeta(x) \, dx = \int_\Omega V(x, t)\Psi(x, t)\zeta(x) \, dx.
$$

For the Hartree potential, we will use the triangle inequality. Thus, we begin by writing,

$$
\int_\Omega W * \rho_\epsilon(n)\Psi_\epsilon(n)(x, t)\zeta(x) \, dx - \int_\Omega W * \rho_\epsilon(n)\Psi_\epsilon(n)(x, t)\zeta(x) \, dx =
\int_\Omega W * \rho_\epsilon(n)[\Psi_\epsilon(n)(x, t) - \Psi(x, t)]\zeta(x) \, dx + \int_\Omega W * [\rho_\epsilon(n) - \rho]\Psi(x, t)\zeta(x) \, dx.
$$
For the LDA potential, we will also use the triangle inequality, and we write,

\[ \|W * \rho_{\epsilon_n}(\cdot,t) - \Psi(\cdot,t)\|_{L^r} \leq \|W\|_{L^2} \|\Psi_{\epsilon_n}(\cdot,t) - \|\Psi\|\|\|\Psi_{\epsilon_n}\|\|L^r. \]

For the first triple product, Young’s inequality is applied to the convolution term, followed by \(L^2\) boundedness; \(L^3\) convergence is applied to the second term of the first product; and Sobolev’s inequality is applied to the third term. For the second triple product, the only term requiring explanation is the convolution term of the product. We estimate as follows.

\[ \|W * [\rho_{\epsilon_n} - \rho]\|_{L^2} \leq \|W\|_{L^2} \|\|\Psi_{\epsilon_n}\|\| - \|\Psi\|\|\|\|\Psi_{\epsilon_n}\|\| + \|\Psi\|\|L^1, \]

which is estimated by the Schwarz inequality. An application of \(L^2\) boundedness and \(L^2\) convergence yields the final result:

\[
\lim_{n \to \infty} \int_\Omega W * \rho_{\epsilon_n}(x) \zeta(x) \, dx = \int_\Omega W * \rho \Psi(x) \zeta(x) \, dx. \tag{23}
\]

For the LDA potential, we will also use the triangle inequality, and we write,

\[
\int_\Omega \Phi_{\epsilon_n}(\rho_{\epsilon_n}) \Psi_{\epsilon_n}(x) \zeta(x) \, dx - \int_\Omega \Phi_{\text{lda}}(\rho) \Psi(x) \zeta(x) \, dx = \int_\Omega \Phi_{\epsilon_n}(\rho_{\epsilon_n}) [\Psi_{\epsilon_n}(x) - \Psi(x)] \zeta(x) \, dx + \int_\Omega [\Phi_{\epsilon_n}(\rho_{\epsilon_n}) - \Phi_{\text{lda}}(\rho)] \Psi(x) \zeta(x) \, dx.
\]

We apply the Hölder inequality to each of the two rhs terms to obtain two products of norms:

\[
\|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) [\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^{r'}} \leq \|\Phi_{\epsilon_n}(\rho_{\epsilon_n})\|_{L^r} \|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^{r'}}, \quad \|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) [\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^{r'}} \leq \|\Phi_{\epsilon_n}(\rho_{\epsilon_n})\|_{L^r} \|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^{r'}},
\]

where \(r = \alpha + 2\) and \(r'\) is conjugate to \(r\). We use the method employed in the proof of Proposition 3.2 in order to estimate the \(L^{r'}\) norms. For convenience, we suppress the scalar \(|\lambda|\); also, \(1 \leq \alpha < 4\). We have, for the first product,

\[
\|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) [\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^{r'}} \leq \|\Phi_{\epsilon_n}(\rho_{\epsilon_n})\|_{L^r} \|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^{r'}} \leq \|\Phi_{\epsilon_n}(\rho_{\epsilon_n})\|_{L^r} \|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^{r'}},
\]

which converges to zero as remarked at the beginning of the proof (see (19)). Thus, the first product of norms is convergent to zero. For the second product, we estimate.

\[
\|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) - \Phi_{\text{lda}}(\rho)\|_{L^{r'}} \leq \|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) - \Phi_{\text{lda}}(\rho)\|_{L^{r'/\alpha}} \|\Psi(t)\|_{L^r}.
\]

To estimate, we apply the triangle inequality to the first factor:

\[
\|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) - \Phi_{\text{lda}}(\rho)\|_{L^{r'/\alpha}} \leq \|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) - \Phi_{\epsilon_n}(\rho)\|_{L^{r'/\alpha}} + \|\Phi_{\epsilon_n}(\rho) - \Phi_{\text{lda}}(\rho)\|_{L^{r'/\alpha}}.
\]

The first term on the rhs is bounded via Young’s inequality, by

\[
\|\Phi_{\epsilon_n}(\rho_{\epsilon_n}) - \Phi_{\epsilon_n}(\rho)\|_{L^{r'/\alpha}} \leq \|\Psi_{\epsilon_n}\|_{L^{r'/\alpha}} - |\Psi_{\epsilon_n}|_{L^{r'/\alpha}}.
\]
The estimation of this expression requires inequality (13) with the identifications \( \Psi_1 \mapsto \Psi_{\epsilon_n}, \Psi_2 \mapsto \Psi \). When the power \( r/\alpha \) is applied to the inequality, and integration over \( \Omega \) is carried out, one can apply Hölder’s inequality with \( p = \alpha \) and \( p' = \alpha/(\alpha - 1) \) to conclude convergence. Convergence for the second term is a consequence of the property of smoothing; since \( |\Psi|^\alpha \in L^{r/\alpha} \), its convolution is convergent in norm. Altogether, we have shown:

\[
\lim_{n \to \infty} \int_{\Omega} \Phi_{\epsilon_n}(\rho_{\epsilon_n}) \Psi_{\epsilon_n}(x,t) \zeta(x) \, dx = \int_{\Omega} \Phi_{\text{lda}}(\rho) \Psi(x,t) \zeta(x) \, dx. \tag{24}
\]

We now use (20) and (21) to conclude that

\[
\lim_{n \to \infty} \langle \partial \Psi_{\epsilon_n}/\partial t, \zeta \rangle = \langle \partial \Psi/\partial t, \zeta \rangle,
\]

so that \( \Psi \) solves the TDDFT system. The initial condition is a consequence of (19) in, say, \( L^2 \) for \( t = 0 \).

It remains to verify the regularity class for \( \Psi \).

**Theorem 3.2.** The function \( \Psi \) of Theorem 3.1 satisfies

\[
\Psi \in C(J; H^1_0(\Omega)) \cap C^1(J; H^{-1}(\Omega)).
\]

**Proof.** We begin with \( \Psi \in C(J; H^1_0(\Omega)) \). We use the representations contained in Lemma 3.1 as applied to \( \Psi \). We rewrite them as follows.

\[
\mathcal{E}(t) = \int_{\Omega} \left[ \frac{\hbar^2}{4m} |\nabla \Psi|^2 + \frac{1}{4}(W * |\Psi|^2) + \frac{1}{2}(V + \Phi_{\text{lda}}) |\Psi|^2 \right] \, dx, \tag{25}
\]

\[
\mathcal{E}(t) = \mathcal{E}(0) + \frac{1}{2} \int_0^t \int_{\Omega} (\partial V/\partial s)(x,s) |\Psi|^2 \, dxds. \tag{26}
\]

Specifically, the expression \( \mathcal{E}(t) \), as defined in (26), is in \( C(J; H^1_0(\Omega)) \), as follows directly from (20) when the boundedness for \( \partial V/\partial t \) is applied. The approach now is to solve for the gradient term in (26) and deduce its continuity from that of each of the other terms. The only terms requiring analysis are the Hartree and LDA terms. The techniques are similar to those used earlier. For the Hartree potential, we have

\[
\int_{\Omega} W * \rho(t) \rho(x,t) \, dx - \int_{\Omega} W * \rho(s) \rho(x,s) \, dx =
\]

\[
\int_{\Omega} W * \rho(t)[\rho(x,t) - \rho(x,s)] \, dx + \int_{\Omega} W * [\rho(t) - \rho(s)] \rho(x,s) \, dx.
\]
Each of the two rhs terms is estimated by the Schwarz inequality, so that we must estimate the following two products of norms:

$$\| W * \rho(t) \|_{L^2} \| \rho(t) - \rho(s) \|_{L^2} \quad \text{and} \quad \| W * [\rho(t) - \rho(s)] \|_{L^2} \| \rho(s) \|_{L^2}.$$ 

For the first product, the first term is estimated by Young’s inequality, to obtain a quantity, bounded on \( J \). We estimate the second factor as

$$\| \rho(t) - \rho(s) \|_{L^2} \leq \| |\Psi(t)| - |\Psi(s)| \|_{L^2} \| |\Psi(t)| + |\Psi(s)| \|_{L^2},$$

which is convergent to zero as \( t \to s \), by the remark following (19). For the second product, the first term is estimated by Young’s inequality:

$$\| W * [\rho(t) - \rho(s)] \|_{L^2} \leq \| W \|_{L^2} \| \rho(t) - \rho(s) \|_{L^1}.$$ 

After factorization, one again concludes convergence to zero as \( t \to s \). Finally, we consider the LDA term.

$$\int_\Omega \Phi_{lda}(\rho(t)) \rho(x,t) \, dx - \int_\Omega \Phi_{lda}(\rho(s)) \rho(x,s) \, dx = \int_\Omega \Phi_{lda}(\rho(t)) [\rho(x,t) - \rho(x,s)] \, dx + \int_\Omega [\Phi_{lda}(\rho(t)) - \Phi_{lda}(\rho(s))] \rho(x,s) \, dx.$$ 

Hölder’s inequality is applied to each of the terms on the rhs, so that we need to estimate the following norm products:

$$\| |\Psi(t)|^\alpha |\Psi(t)| - |\Psi(s)| \|_{L^{r'}} \| |\Psi(t)| + |\Psi(s)| \|_{L^r},$$

$$\| |\Psi(t)|^\alpha - |\Psi(s)|^\alpha |\Psi(s)| \|_{L^{r'}} \| |\Psi(s)| \|_{L^r},$$

where \( r = \alpha + 2 \) and \( r' \) is conjugate to \( r \). As has been demonstrated previously, the first product is estimated by

$$\| |\Psi(t)|^\alpha |\Psi(t)| - |\Psi(s)| \|_{L^{r'}} \| \Psi(t) \|_{L^r} + \| |\Psi(s)| \|_{L^r},$$

which converges to zero as \( t \to s \). The second product is estimated, with the help of (13) and Hölder’s inequality, as

$$\alpha \| (|\Psi(t)| + |\Psi(s)|)^{\alpha-1} (|\Psi(t)| - |\Psi(s)|) \|_{L^{r'/\alpha}} \| \Psi(s) \|_{L^r}^{2}, \quad (27)$$

and another application of Hölder’s inequality, with \( p = \alpha \) and \( p' \) conjugate to \( \alpha \), gives the bound

$$\alpha \| (|\Psi(t)| + |\Psi(s)|) \|_{L^{r'}} \| |\Psi(t)| - |\Psi(s)| \|_{L^r} \| \Psi(s) \|_{L^r}^{2},$$

so that this term also converges to zero. It follows that \( \Psi \in C(J; H^1) \). In order to conclude that \( \Psi \in C^1(J; H^{-1}) \), we subtract two copies of the TDDFT system, one evaluated at \( t \), and the other at \( s \), and we estimate for an arbitrary
test function $\zeta$. Only the Hartree and LDA terms require explanation. For the Hartree potential, we observe that
\[
\int_{\Omega} W \ast \rho(t) \Psi(x, t) \zeta(x) \, dx - \int_{\Omega} W \ast \rho(s) \Psi(x, s) \zeta(x) \, dx
\]
has been estimated earlier. Similarly, the corresponding LDA difference,
\[
\int_{\Omega} \Phi_{\text{lda}}(\rho(t)) \Psi(x, t) \zeta(x) \, dx - \int_{\Omega} \Phi_{\text{lda}}(\rho(s)) \Psi(x, s) \zeta(x) \, dx,
\]
has also been estimated. This completes the proof.

\[\square\]

**Remark 3.2.** The combination of Theorem 3.1 and Theorem 3.2 gives Theorem 2.1 as formulated earlier. This is the central result of the article.

### 4 Summary Remarks

We have formulated a model within the framework of time dependent density functional theory, employing the local density approximation (LDA), which is typically used in simulation. We have obtained existence, but not uniqueness, for this model on a bounded domain in $\mathbb{R}^3$. The structure of the LDA approximation can be generalized, in terms of its form and time dependence. For general structure properties, we refer the reader to [7], where the nonlinear Schrödinger equation is discussed for potentials not directly depending on time. Although these more general potentials could be incorporated here, the algebraic growth, in terms of the exponent $\alpha$, cannot be modified for the methods of this article to apply. We have selected the form here, because of its wide usage in the literature. In regard to time dependence, there is nothing to prevent the use of separate exchange and correlation potentials, one of the LDA format, and the other having a non-local format as described in section 1.3. We will consider these cases in future work.

As noted, we have not established uniqueness in this article. The use of compactness methods, as carried out here and, for example, in [7, Ch. 3], does not readily provide an avenue to establish uniqueness. This contrasts with the use of the contraction mapping theorem in [2], where uniqueness followed from the method itself. On the other hand, the solutions of the smoothed problems are unique, but there seems no obvious way to extend this property to a limit.

The requirement that $\alpha < 4$ in this article is closely related to the failure of the compactness of the (continuous) Sobolev embedding $H^1_0(\Omega) \hookrightarrow L^6(\Omega)$. Note that the proof of Theorem 3.1 begins with the very application of the compactness of the embedding into $L^r$ for $1 \leq r < 6$. One uses the property that weakly convergent sequences are mapped into strongly convergent sequences. Also, the use of the assumption $1 \leq \alpha \leq 4/3$, for $\lambda < 0$, is needed only in the estimates derived in Proposition 3.1 where the size of $|\lambda|$ also enters as discussed in Remark 3.1.
A Notation and Norms

We employ complex Hilbert spaces in this article.

\[ L^2(\Omega) = \{ f = (f_1, \ldots, f_N)^T : |f_j|^2 \text{ is integrable on } \Omega \} \]

\[ (f, g)_{L^2} = \sum_{j=1}^{N} \int_{\Omega} f_j(x)\overline{g_j(x)} \, dx. \]

However, \( \int_{\Omega} fg \) is interpreted as

\[ \sum_{j=1}^{N} \int_{\Omega} f_j g_j \, dx. \]

For \( f \in L^2 \), as just defined, if each component \( f_j \) satisfies \( f_j \in H_0^1(\Omega; \mathbb{C}) \), we write \( f \in H_0^1(\Omega; \mathbb{C}^N) \), or simply, \( f \in H_0^1(\Omega) \). The inner product in \( H_0^1 \) is

\[ (f, g)_{H_0^1} = (f, g)_{L^2} + \sum_{j=1}^{N} \int_{\Omega} \nabla f_j(x) \cdot \overline{\nabla g_j(x)} \, dx. \]

\[ \int_{\Omega} \nabla f \cdot \nabla g \] is interpreted as

\[ \sum_{j=1}^{N} \int_{\Omega} \nabla f_j(x) \cdot \nabla g_j(x) \, dx. \]

Finally, \( H^{-1} \) is defined as the dual of \( H_0^1 \), and its properties are discussed at length in [14]. The Banach space \( C(J; H_0^1) \) is defined in the traditional manner:

\[ C(J; H_0^1) = \{ u : J \mapsto H_0^1 : u(\cdot) \text{ is continuous} \}, \quad \| u \|_{C(J; H_0^1)} = \sup_{t \in J} \| u(t) \|_{H_0^1}. \]
The Local Density Approximation

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