Consistency of the Local Density Approximation and Generalized Quantum Corrections 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), a non-interacting electron model, which predicts charge exactly. An exchange-correlation potential is included in the Hamiltonian to enforce this property. 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 density approximations (LDA) for building the exchange-correlation potential, as part of a set of quantum corrections. Existence and uniqueness of solutions are established separately within a framework for general quantum corrections, including time-history corrections and ionic Coulomb potentials, in addition to LDA potentials. In summary, we are able to demonstrate a unique weak solution, on an arbitrary time interval, for a general class of quantum corrections, including those typically used in numerical simulations of the model.

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1 Introduction

Time dependent density functional theory (TDDFT) was introduced by E. Runge and E.K.U. Gross in [1] as a non-interacting electron model which tracks electron charge exactly. An exposition of the subject may be found in [2]. When Kohn-Sham potentials are used, the electronic Hamiltonian includes any (time dependent) external potentials, ionic potentials, the Hartree potential, and the compensating exchange-correlation potential to ensure the non-interacting and charge exactness features of the model. By permitting time dependent potentials, TDDFT extends the nonlinear Schrödinger equation, which has been studied extensively [3, 4], principally with potentials not directly depending on time. Some progress for time dependent linear Hamiltonians has been made [5]. In previous work [6, 7], we analyzed closed quantum systems on bounded domains of $\mathbb{R}^3$ via time-ordered evolution operators. The article [6] demonstrated strong H^2 solutions, compatible with simulation, whereas the article [7] demonstrated weak solutions; [7] also includes the exchange-correlation component of the Hamiltonian potential, not included in [6], which is a nonlocal time-history term, satisfying certain regularity hypotheses. TDDFT is a significant field for applications, including computational nano-electronics and chemical physics [8].

An important early article in the time dependent case, directed toward Hartree-Fock Hamiltonians, is [9]. This article included nuclear dynamics as a coupled classical dynamical system, and defined an electronic Hamiltonian in terms of a kinetic term, together with a Hartree potential, an ionic potential with mobile point masses, and an external, electric-field-induced potential. The mathematical framework was defined on $\mathbb{R}^3$ in terms of a Cauchy problem with H^2 initial datum. A recent article directed toward TDDFT, in which a quantum correction is of local density type, is [10]; this article couples quantum mechanics and control theory. Neither of these articles allows for a time-history exchange-correlation potential.

In this article, we introduce a class of quantum corrections, including the local density approximation, but also ionic Coulomb potentials and time-history potentials. As we demonstrate below, smoothing of such potentials provides a model within the framework of [7]. By using compactness arguments suggested in [4], we are able to obtain a solution of the originally posed model. Uniqueness is also established. The use of evolution operators and smoothing as presented here is consistent with techniques in the applied literature [8] and provides direct support for successive approximation and other numerical procedures [11, 12]. In this sense, the results of this article are more inclusive than an existence/uniqueness analysis.

In the following subsections of the introduction, we summarize the basic results of [7], as a starting point for the present article. In section two, we formulate the new model, which incorporates the category of quantum corrections, and we prove that its smoothed version lies within the scope of [7]. In section three, we introduce the compactness arguments, and establish existence of a weak solution as the limit of solutions of the smoothed model. Uniqueness is established in section four. We conclude with some summary remarks.

1.1 The model

In its original form, without ionic influence, TDDFT includes three components for the electronic 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,

$$i\hbar\frac{\partial\Psi(t)}{\partial t} = \hat{H}\Psi(t). \tag{1}$$

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

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

An initial condition,

$$\Psi(0) = \Psi_0, \tag{2}$$

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

$$V_{\mathrm{e}}(\mathbf{x}, t, \rho) = V(\mathbf{x}, t) + W * \rho + \Phi(\mathbf{x}, t, \rho).$$

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

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

which depends only upon values $W(\mathbf{z})$, $\|\mathbf{z}\| \leq \mathrm{diam}(\Omega)$. We may redefine W smoothly outside this set, so as to obtain a function of compact support for which Young's inequality applies. The exchange-correlation potential Φ represents a time-history of ρ:

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

The Hamiltonian operator is given by,

$$\hat{H} = -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{x}, t) + W * \rho + \Phi(\mathbf{x}, t, \rho), \tag{3}$$

and m designates the effective mass and $\hbar$ the normalized Planck's constant. If ionic influence is present, then (3) is adjusted, typically by Coulomb potentials.

1.2 Definition of weak solution and function spaces

The solution Ψ 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 $H_0^1(\Omega)$: $\Psi \in C(J; H_0^1)$. The time derivative is continuous from J into the dual H^{-1} of H_0^1: $\Psi \in C^1(J; H^{-1})$. The spatially dependent test functions ζ are arbitrary in H_0^1. The duality bracket is denoted $\langle f, \zeta \rangle$. *Norms and inner products are discussed in Appendix A.* We will make use of the equivalence of the standard H_0^1 norm and the gradient seminorm, due to the Poincaré inequality, which holds for bounded domains Ω [13].

Definition 1.1. *For $J = [0, T]$, the vector-valued function $\Psi = \Psi(\mathbf{x}, t)$ is a weak solution of (1, 2, 3) if $\Psi \in C(J; H_0^1(\Omega)) \cap C^1(J; H^{-1}(\Omega))$, if Ψ satisfies the initial condition (2) for $\Psi_0 \in H_0^1(\Omega)$, and if $\forall\, 0 < t \leq T$:*

$$i\hbar\langle \frac{\partial \Psi(t)}{\partial t}, \zeta \rangle = \int_\Omega \frac{\hbar^2}{2m} \nabla \Psi(\mathbf{x}, t) \cdot \nabla \zeta(\mathbf{x}) + V_{\mathrm{e}}(\mathbf{x}, t, \rho) \Psi(\mathbf{x}, t) \zeta(\mathbf{x}) d\mathbf{x}. \tag{4}$$

1.3 Hypotheses and theorem statement

We provide some discussion, relevant to the physical model, prior to the statement of the hypotheses. Additional discussion will be provided following the hypotheses. It is emphasized that the hypotheses of this subsection are those required for the original theory of [7] to apply; this was accomplished with evolution operators and the Banach fixed point mapping. Subsequent sections of this article consider more general families of correction potentials.

The time-history potential $\Phi(\mathbf{x}, t, \rho)$ above has a structure, including the time-integrated part, which is motivated by [14, Eqs. (15), (17)]. This article characterizes the action functionals A whose variational derivatives with respect to ρ yield appropriate exchange-correlation potentials. The form of Φ selected above represents a general statement of these ideas. It is not unreasonable that the mathematical hypotheses, to be stated shortly, should resemble the known properties of the Hartree potential because of the restorative nature of exchange and correlation. From a mathematical perspective, the model permits multiple 'copies' of Φ, allowing for quantum corrections. These are seen to be important for applications. For example, in the quantum chemistry community [15], it is appropriate to split Φ: the exchange part is represented by a weighted density approximation (WDA), while the correlation part is represented by a local density approximation (LDA). The nonlocal WDA form for Φ is appropriate for nonuniform mixtures [16]. The general form we have allowed for Φ is intended to anticipate applications of this type.

The following hypotheses are those for which the evolution operator theory of [7] applies. The present article builds upon this established theory.

We assume the following hypotheses in order to apply the results of [7].

- 1. The time-history potential Φ is continuous in $t \in J$ into H_0^1.
  2. Φ is bounded, uniformly in $t \in J$, from H_0^1 into $W^{1,3}$. More precisely, by boundedness, we mean that the family $\{\Phi(\cdot\,, t, \cdot)\}$ maps every fixed ball in H_0^1 into a fixed ball in $W^{1,3}$, uniformly in t.
- The derivative $\partial\Phi/\partial t = \phi$ is assumed measurable, and bounded in its arguments.
- Furthermore, the following smoothing condition is assumed, expressed by a (uniform) Lipschitz norm condition:

  $$\forall t \in [0,T], \text{if } \|\Psi_j\|_{H_0^1}, j = 1,2, \text{ are bounded by } r,$$

  then

  $$\|[\Phi(\cdot\,, t, |\Psi_1|^2) - \Phi(\cdot\,, t, |\Psi_2|^2)]\psi\|_{H^1} \leq C(r)\|\Psi_1 - \Psi_2\|_{H_0^1}\|\psi\|_{H_0^1}. \quad (5)$$

  Here, ψ is arbitrary in H_0^1 and $C(r)$ depends only on r.
- If $\Phi(\cdot\,, 0, \rho)$ fails to be a nonnegative functional of $\rho = |\Psi|^2$, we assume that it satisfies, uniformly in t, for $\|\Psi(t)\|_{L^2} = \|\Psi_0\|_{L^2}$, the constraint that

  $$\|\Phi(\cdot\,, 0, |\Psi|^2)|\Psi|^2\|_{L^1} \leq C_1\|\nabla\Psi\|_{L^2}^2 + C_2,\ \Psi(t) \in H_0^1, \quad (6)$$

  for nonnegative constants C_1 and C_2. It is required that C_2 depend only on $\|\Psi_0\|_{L^2}$ and the problem data, and C_1 is sufficiently small:

  $$C_1 < \frac{\hbar^2}{2m}. \quad (7)$$
- The so-called external potential V is assumed to be continuously differentiable on the closure of the space-time domain.

Remark 1.1. *We comment here on the hypotheses.*

1. *The regularity assumed for Φ in the first assumption is consistent with certain requirements of TDDFT. One of these is the Zero Force Theorem [2], which imposes a gradient condition on Φ. We note that the Hartree potential satisfies these conditions. In fact, any convolution of the form $\Phi = F * \rho$, where $F \in W^{1,1}$, satisfies the conditions.*
2. *An inequality of the form (5) is satisfied by the Hartree potential [20, Theorem 3.1], and by any convolution of the form $\Phi = F * \rho$, with $F \in L^2$ and $\nabla F \in L^1$. It was used in [7] to construct the contraction mapping used there for the evolution operator. For quantum corrections not satisfying this condition, the smoothing is utilized in the following section in order to place the smoothed systems within this framework.*

3. *Hypotheses (6, 7) are relevant only when the associated potentials are negative. This is expected to occur for restoring potentials and certain Coulomb potentials. In the following section, it will be necessary to smooth certain components of the quantum correction potential. The smoothed Coulomb potentials satisfy (6, 7) without qualification. However, for smoothed LDA approximations, there is a disparity in exponent bounds for α. A smaller range is necessary for negative potentials (see (20) to follow for verification in this case). Also, unsmoothed convolutions of the form $\Phi = F * \rho$, with $\nabla F \in L^1$, satisfy the conditions if they have sufficiently small L^∞ bounds.*

The following theorem was proved in [7], based upon the evolution operator as presented in [17], 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 Quantum Corrections and the Local Density Approximation

In this section, we define a class of quantum correction potentials, including the local density approximation to the exchange-correlation potential Φ. These correction potentials are of three types.

1. The local density approximation, discussed in Definition 2.1 to follow. This potential is designated as $\Phi_{\rm lda}(\rho)$.

2. A finite number of Coulomb ionic potentials, $c_j W(\cdot - \mathbf{x}_j)$, subject to the Born-Oppenheimer approximation. In particular, the ionic masses are assumed to be point masses, at fixed locations $\mathbf{x}_j \in \Omega$. The function W is introduced in section 1.1. The constants c_j may be positive or negative. The aggregate of these Coulomb potentials is designated $\Phi_{\rm c}(\cdot)$.

3. A time-history potential of the structure of Φ, introduced in section 1.1. The presence of this potential allows for physical modeling flexibility, since the exchange potential and the correlation potential are viewed separately in TDDFT. We permit one of these to be approximated locally and the other by a time-history among the modeling choices. We retain the notation $\Phi(\cdot, t, \rho)$ for this component, assumed to satisfy the hypotheses detailed in section 1.3. Also, it is assumed that $\Phi(\cdot, t, \rho_n(\cdot, t))$ converges in L^2, uniformly in t, if $\rho_n(\cdot, t)$ converges in L^2, uniformly in t.

The consolidated quantum correction potential is then given by

$$\Phi_{\rm qc}(\cdot, t, \rho) = \Phi_{\rm lda}(\rho) + \Phi_{\rm c}(\cdot) + \Phi(\cdot, t, \rho). \tag{8}$$

Definition 2.1. *The local density approximation $\Phi_{\rm lda}$ is now defined. We consider the following approximation, where λ is a real constant, positive or negative.*

$$\Phi_{\rm lda}(\rho) = \lambda \rho^{\alpha/2} = \lambda |\Psi|^{\alpha}. \tag{9}$$

Additionally,

- *If $\lambda > 0$, the range of α is $1 \leq \alpha < 4$.*
- *If $\lambda < 0$, the range of α is $1 \leq \alpha \leq 4/3$. Also, $|\lambda|$ must be sufficiently small, consistent with (6) and (7).*

We redefine the Hamiltonian considered here as

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

$$\Phi_{\rm qc}(\cdot\,, t, \rho) = \underbrace{\lambda|\Psi|^{\alpha}(\cdot\,, t)}_{\Phi_{\rm lda}} + \underbrace{\sum_{j=1}^{M} c_j \frac{1}{|\cdot - \mathbf{x}_j|}}_{\Phi_{\rm c}} + \Phi(\cdot\,, t, \rho). \tag{10}$$

The proofs accommodate a finite number of terms in $\Phi_{\rm lda}$. One term has been chosen for simplicity. The parameters of $\Phi_{\rm lda}$ satisfy the assumptions of Definition 2.1. The numerical constants c_j are of arbitrary sign, and the ionic locations $\mathbf{x}_j$ are fixed interior points in Ω. Φ satisfies the hypotheses specified in (3) above, and is a nonlocal potential such as weighted density approximation. Convolutions, discussed in Remark 1, represent an important class. For simplicity, we assume that the leading part, $\Phi(\cdot\,, 0, \rho)$, is conserved, up to a positive constant multiple. This holds for convolutions and other important examples. The time integrated part of Φ is motivated by [14]. The following theorem is the goal of our analysis.

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

$$V_{\rm e}(\mathbf{x}, t, \rho) = V(\mathbf{x}, t) + W * \rho + \Phi_{\rm qc}(\cdot\,, t, \rho), \tag{11}$$

then there is a weak solution of (4) in the regularity class $C(J; H^1_0) \cap C^1(J; H^{-1})$ which satisfies the specified initial condition. Uniqueness holds except possibly for $1 < \alpha < 2$.

The existence part of the proof of Theorem 2.1 is carried out in section three (see Theorems 3.1 and 3.2). The uniqueness is demonstrated in section four.

2.1 The smoothing

We begin by defining a standard convolution [18].

Definition 2.2. *Suppose that a nonnegative function ϕ_1 is given, $\phi_1 \in C_0^\infty(\mathbb{R}^3)$, of integral one. Set*

$$\phi_\epsilon(\mathbf{x}) = \epsilon^{-3}\phi_1(\mathbf{x}/\epsilon), \ \mathbf{x} \in \mathbb{R}^3,$$

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

$$f_\epsilon = \phi_\epsilon * f.$$

We recall [18] that $\lim_{\epsilon\to 0} f_\epsilon = f$ in L^p and $\|f_\epsilon\|_{L_p} \leq \|f\|_{L_p}$, $\forall \epsilon > 0$.

Definition 2.3. *We denote by Φ_ϵ a smoothed replacement of $\Phi_{\rm qc}$ as follows.*

1. $\Phi_{\rm lda} \mapsto \phi_\epsilon * \Phi_{\rm lda}$.
2. $\Phi_{\rm C} \mapsto \phi_\epsilon * \Phi_{\rm C}$.
3. *Time-history terms are not smoothed.*

The effective potential for the approximate problem is given by:

$$V_{\rm e}(\mathbf{x}, t, \rho_\epsilon) = V(\mathbf{x}, t) + W * \rho_\epsilon + \Phi_\epsilon(\mathbf{x}, t, \rho_\epsilon). \tag{12}$$

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 $\Phi_{\rm qc}$ is replaced by its smoothing Φ_ϵ, as specified in Definition 2.3, then the hypotheses of section 1.3 hold, as applied to Φ_ϵ. In particular, Theorem 1.1 is applicable. With $V_{\rm e}$ defined by (12), there exists a unique weak solution Ψ_ϵ, as specified in Definition 1.1, of the corresponding system:*

$$i\hbar\langle\frac{\partial\Psi_\epsilon(t)}{\partial t}, \zeta\rangle = \int_\Omega \frac{\hbar^2}{2m}\nabla\Psi_\epsilon(\mathbf{x}, t)\cdot\nabla\zeta(\mathbf{x}) + V_{\rm e}(\mathbf{x}, t, \rho_\epsilon)\Psi_\epsilon(\mathbf{x}, t)\zeta(\mathbf{x})\, d\mathbf{x}. \tag{13}$$

Proof. We observe that the time-history term, if present, is assumed to satisfy the assumptions of section 1.3. This includes (6) and (7), which are required to hold in the aggregate, inclusive of all nonpositive terms for the potential Φ_ϵ. The Coulomb potential does not depend on t or ρ; although the unsmoothed potential fails to be in $W^{1,3}$, its smoothing is in this space. Since individual terms of $\phi_\epsilon * \Phi_{\rm c}$ may be negatively signed, we estimate the collective potential. We show that this potential satisfies (6) and (7), with C_1 preselected to be arbitrarily small. Initially, we estimate, for $\eta > 0$ arbitrary,

$$\|(\phi_\epsilon * \Phi_{\rm c})|\Psi|^2\|_{L^1} \leq (1/2)[\eta^2\|(\phi_\epsilon * \Phi_{\rm c})\Psi\|^2_{L^2} + \eta^{-2}\|\Psi\|^2_{L^2}]. \tag{14}$$

By the Hölder inequality, with conjugate indices $p = 3, p' = 3/2$, we have

$$\|(\phi_\epsilon * \Phi_{\rm c})\Psi\|^2_{L^2} \leq [\|\phi_\epsilon * \Phi_{\rm c}\|_{L^3}\|\Psi\|_{L^6}]^2 \leq [\|\phi_1\|_{L^3}\|\Phi_{\rm c}\|_{L^1}\|\Psi\|_{L^6}]^2. \tag{15}$$

By the equivalence of norms on H^1_0, and by Sobolev's inequality, we may select η so that (7) holds for any preselected C_1. This verifies the final requirement for the Coulomb potential.

For the smoothing of $\Phi_{\rm lda}$, we state the three properties required to be verified.

1. Φ_ϵ maps sets bounded in H^1_0 into sets bounded in $W^{1,3}$.
2. The Lipschitz property (5) holds.
3. If $\lambda < 0$, $\|\phi_\epsilon * \Phi_{\rm lda}(\rho)|\Psi|^2\|_{L^1} \leq C_1 \|\nabla\Psi\|^2_{H^1_0}$, where C_1 does not depend on t and satisfies (7). This is a case where $C_2 = 0$.

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

Property (1) is immediate from the inequalities,

$$\|\phi_\epsilon * \Phi_{\rm lda}(\rho)\|_{L^3} \leq |\lambda| \, \|\phi_\epsilon\|_{L^3} \| |\Psi|^\alpha \|_{L^1}, \quad \|\nabla\phi_\epsilon * \Phi_{\rm lda}(\rho)\|_{L^3} \leq |\lambda| \, \|\nabla\phi_\epsilon\|_{L^3} \| |\Psi|^\alpha \|_{L^1},$$

which follow from Young's inequality, applied to the convolution. 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 $\phi_\epsilon * \Phi_{\rm lda}/|\lambda|$:

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

$$\|\nabla\phi_\epsilon * (|\Psi_1|^\alpha - |\Psi_2|^\alpha)\psi \; + \phi_\epsilon * (|\Psi_1|^\alpha - |\Psi_2|^\alpha)\nabla\psi\|_{L^2}. \tag{16}$$

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 [19]:

$$\left(\frac{y^r - z^r}{y^s - z^s} \frac{s}{r} \right)^{\frac{1}{r-s}} \leq \max(y, z), \; y \geq 0, z \geq 0, y \neq z, r > 0, s > 0, s \neq r. \tag{17}$$

We apply (17) with the identifications.

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

to obtain the pointwise estimate, which holds almost everywhere in Ω,

$$| \, |\Psi_1|^\alpha - |\Psi_2|^\alpha | \leq \alpha (\max(|\Psi_1|, |\Psi_2|))^{\alpha - 1} \, | \, |\Psi_1| - |\Psi_2| \, |. \tag{18}$$

Although we will require inequality (18) later in the article, it is more convenient here to use the less sharp inequality, derived from (18):

$$| \, |\Psi_1|^\alpha - |\Psi_2|^\alpha | \leq \alpha (1 + |\Psi_1| + |\Psi_2|)^\alpha \, | \, |\Psi_1| - |\Psi_2| \, |.$$

We use a technique motivated by [4]. If $r = \alpha + 2$, and r' is conjugate to r, if $p = r/r'$, and p' is conjugate to p, then

$$\alpha r' p' = r, \; r' p = r, \tag{19}$$

and an application of Hölder's inequality gives

$$\| \, |\Psi_1|^\alpha - |\Psi_2|^\alpha \|_{L^{r'}} \leq \alpha \|1 + |\Psi_1| + |\Psi_2|\|^\alpha_{L^r} \| \, |\Psi_1| - |\Psi_2| \, \|_{L^r} \leq C \|\Psi_1 - \Psi_2\|_{L^r}.$$

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 (16) 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.

For property (3), which corresponds to $\lambda < 0$ and $1 \leq \alpha \leq 4/3$, we consider the following estimate via two applications of Hölder's inequality:

$$|\lambda| \left| \int_\Omega |\phi_\epsilon * \Psi_\epsilon|^\alpha |\Psi_\epsilon|^2 \, d\mathbf{x} \right| \leq |\lambda| \, |\Omega|^{2/3 - \alpha/2} \|\Psi_\epsilon\|^\alpha_{L^2} \, \|\Psi_\epsilon\|^2_{L^6}. \tag{20}$$

Since the L^2 norm of $\Psi = \Psi_\epsilon$ is specified in (6), λ can be chosen to satisfy (7) by use of the Sobolev embedding theorem. It follows that a unique weak solution Ψ_ϵ exists for the smoothed system as formulated. □

3 Existence

The results of this section are derived for an arbitrary time interval $[0, T]$. They are directed toward the existence statement in Theorem 2.1. The compactness techniques are motivated by [4].

3.1 'A priori' bounds for the smoothed solutions

We begin by quoting a result proved in [7], now applied to the family of solutions Ψ_ϵ. We have absorbed constants into the conserved Hamiltonian quantities associated with Φ_ϵ. Thus, $\lambda \mapsto \frac{2\lambda}{\alpha+2}$ with a similar statement for the leading term of Φ.

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

$$\mathcal{E}(t) = \int_\Omega \left[\frac{\hbar^2}{4m} |\nabla \Psi_\epsilon|^2 + \left(\frac{1}{4}(W * |\Psi_\epsilon|^2) + \frac{1}{2}(V + \Phi_\epsilon(\cdot, t, \rho_\epsilon)) \right) |\Psi_\epsilon|^2 \right] d\mathbf{x}, \tag{21}$$

then the following identity holds:

$$\mathcal{E}(t) = \mathcal{E}(0) + \frac{1}{2} \int_0^t \int_\Omega [(\partial V/\partial s)(\mathbf{x}, s) + \phi(\mathbf{x}, s)] |\Psi_\epsilon|^2 \, d\mathbf{x} ds, \tag{22}$$

where $\mathcal{E}(0)$ *is given by*

$$\int_\Omega \left[\frac{\hbar^2}{4m}|\nabla\Psi_0|^2 + \left(\frac{1}{4}(W * |\Psi_0|^2) + \frac{1}{2}(V(\cdot\,,0) + \Phi_\epsilon(\cdot\,,0,\rho_0)\right)|\Psi_0|^2\right]\,d\mathbf{x}.$$

Proposition 3.1. *The kinetic term is bounded above by a natural splitting. For each fixed* t*:*

$$\frac{\hbar^2}{4m}\int_\Omega |\nabla\Psi_\epsilon|^2\,d\mathbf{x} \le \mathcal{F}_\epsilon(t) + \mathcal{G}_\epsilon(t).$$

Here, $\mathcal{F}_\epsilon(t)$ *is a quantity which can be bounded above, independently of* t *and* ϵ*, in a manner depending only on the data of the problem. It is given explicitly by*

$$\mathcal{F}_\epsilon(t) = \mathcal{E}(0) + \frac{1}{2}\int_0^t\int_\Omega [(\partial V/\partial s)(\mathbf{x},s) + \phi(\mathbf{x},s)]|\Psi_\epsilon|^2 d\mathbf{x}ds - \frac{1}{2}\int_\Omega V(\mathbf{x},t)|\Psi_\epsilon|^2 d\mathbf{x}.$$

Moreover, $\mathcal{G}_\epsilon(t)$ *can be estimated as the sum of two terms: the first can be absorbed into the kinetic term, while the second is independent of* ϵ *and* t*.* $\mathcal{G}_\epsilon(t)$ *is given explicitly by*

$$\mathcal{G}_\epsilon(t) = -\frac{1}{2}\int_\Omega \Phi_\epsilon(\rho_\epsilon)|\Psi_\epsilon|^2 d\mathbf{x}.$$

Proof.

- The estimation of $\mathcal{F}_\epsilon(t)$

We notice that $V, \partial V/\partial t, \phi$ are bounded on the finite measure space-time domain $\Omega \times [0,T]$, so that the estimation of $\mathcal{F}_\epsilon(t)$ reduces to the analysis of the smoothed term in $\mathcal{E}(0)$ given by

$$\int_\Omega \Phi_\epsilon(\cdot\,,0,\rho_0)|\Psi_0|^2\,d\mathbf{x}.$$

Since the time-history, if present, is not smoothed, and acts boundedly, it suffices to examine the Coulomb and LDA potentials.

- The Coulomb term.

By the Schwarz inequality and Young's inequality, we estimate

$$\|(\phi_\epsilon * \Phi_{\rm c})|\Psi_0|^2\|_{L^1} \le \|\phi_1\|_{L^2}\|\Phi_{\rm c}\|_{L^1}\|\Psi_0\|^2_{L^4}.$$

An application of Sobolev's inequality concludes the argument.

- The LDA term.

This is a direct estimate:

$$\|\phi_\epsilon * \Phi_{\rm lda}(\rho_0)|\Psi_0|^2\|_{L^1} \le \|\phi_\epsilon * |\Psi|^\alpha\|_{L^{3/2}}\|\Psi_0\|^2_{L^6} \le \|\phi_1\|_{L^{3/2}}\||\Psi_0|^\alpha\|_{L^1}\|\Psi_0\|^2_{L^6}.$$

Since $\alpha < 4$, the estimate follows as previously from the embedding theorems.

- The estimation of $\mathcal{G}_\epsilon(t)$

This represents the more delicate part of the proof.

- The time-history term.

If the term,

$$\Phi(\mathbf{x}, t, \rho) = \Phi(\mathbf{x}, 0, \rho) + \int_0^t \phi(\mathbf{x}, s, \rho)\, ds,$$

is included, and the leading term fails to be a positive functional, then we have required that (6, 7) hold, here as applied to Ψ_ϵ. This is consistent with the structure of $\mathcal{G}_\epsilon$ as stated. The integral term has been discussed in the previous part and is bounded. Note that (7) is required to hold for the *aggregate* potential, including those components to be discussed now. We shall mention this at the appropriate time.

- The Coulomb term.

We use the core of the argument as developed in the proof of Proposition 2.1. Indeed, for any preselected C_1, inequality (7) can be satisfied. This follows directly from (14) and (15 with a proper choice of η.

- The LDA term.

This pertains to the case $\lambda < 0$ if this term is included. We have already derived the relevant inequality, viz. , (20) near the conclusion of the proof of Proposition 2.1. This inequality is required here also.

In order to satisfy (7) in the aggregate sense, we reason as follows. We accept the time-history term as given, if at all. We choose λ so that the sum of the LDA potential and time-history potential continues to satisfy this inequality. This can be extended to a finite number of such terms. Finally, we have shown that the Coulomb potential can be included so as to maintain this inequality. This concludes the proof. □

The following corollary is immediate from the equivalence of norms on H^1_0.

Corollary 3.1. *There is a bound r_0 in the norm of $C(J; H^1_0)$ for the smoothed solutions.*

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

$$\|(\Psi_\epsilon)_t\|_{H^{-1}}.$$

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 $\zeta, \|\zeta\|_{H^1_0} \leq 1$. The gradient term is bounded by Corollary 3.1, while the bound for the external potential term follows directly from the hypothesis on V. For the Hartree term, we estimate, by Hölder's inequality and Young's inequality, for each $t \in J$,

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

Sobolev's inequality, combined with Proposition 3.1, gives the bound for this term.

We now consider the components of the quantum correction potential.

- The LDA term.

For the smoothed LDA term, the sign of λ 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, for each $t \in J$,

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

The first factor on the rhs requires additional explanation. We have, by another application of Hölder's inequality, with $p = r/r'$ and p' conjugate to p (note that $r/\alpha = r'p'$),

$$\begin{aligned} \|\phi_\epsilon * |\Psi_\epsilon|^\alpha \ \Psi_\epsilon\|_{L^{r'}} &\leq \|\phi_\epsilon * |\Psi_\epsilon|^\alpha\|_{L^{r/\alpha}} \|\Psi_\epsilon\|_{L^r} \leq \||\Psi_\epsilon|^\alpha\|_{L^{r/\alpha}} \|\Psi_\epsilon\|_{L^r} \\ &\leq \|\Psi_\epsilon\|_{L^r}^{\alpha+1}. \end{aligned} \tag{23}$$

We conclude that the LDA term is bounded in the dual norm, as claimed.

- The Coulomb term.

By the Schwarz inequality and Young's inequality, uniformly in t,

$$\left| \int_\Omega \phi_\epsilon * \Phi_{\rm c} \ \Psi_\epsilon \zeta \right| \leq \|\phi_1\|_{L^2} \|\Phi_{\rm c}\|_{L^1} \|\Psi_\epsilon\|_{L^4} \ \|\zeta\|_{L^4},$$

and the estimate is completed by Sobolev's inequality.

- Time-history term.

By Proposition 3.1, the smoothed solutions are bounded in H_0^1, uniformly in t, so that, by the first hypothesis in section 1.3, the functions $\Phi(\cdot\,, t, \Psi_\epsilon)$ have a uniform H_0^1 bound. It follows as in previous estimates that the term,

$$\int_\Omega \Phi(\cdot\,, 0, \Psi_\epsilon) \ \Psi_\epsilon \zeta \ d\mathbf{x},$$

defines a functional which is bounded in the dual norm. □

The following corollary is an immediate consequence of Corollary 3.1 and Proposition 3.2.

Corollary 3.2. *Any sequence taken from the set* $\{\Psi_\epsilon\}$ *of solutions of the smoothed systems is bounded in the norms of* $C(J; H_0^1)$ *and* $C^1(J; H^{-1})$.

3.2 Convergent subsequences

We begin by stating the two basic lemmas derived from the propositions in Appendix B. These are due, in the form stated there, to the authors of [4] and [21], resp.

Lemma 3.2. *There is an element* $\Psi \in L^\infty(J; H_0^1(\Omega)) \cap W^{1,\infty}(J; H^{-1}(\Omega))$*, and a sequence* Ψ_{ϵ_n} *satisfying the weak convergence property,*

$$\Psi_{\epsilon_n}(t) \rightharpoonup \Psi(t), \text{ in } H_0^1, \ \forall t \in J. \tag{24}$$

Proof. The preceding corollary, coupled with Proposition B.1, part (1), furnishes the necessary argument. □

Lemma 3.3. *Suppose* $r < 6$ *is fixed. A subsequence of the sequence in (24) may be assumed to converge in* $C(J; L^r(\Omega))$.

Proof. The equicontinuity of the sequence from J to H_0^1 is derived from the fundamental theorem of calculus applied on an arbitrary subinterval, together with the boundedness estimates in the dual space. The compact embedding of $H_0^1 \mapsto L^r$, coupled with Proposition B.2, furnishes the necessary remaining details. We have identified Y with L^r here. □

We divide the verification of Theorem 2.1 into two parts.

Theorem 3.1. *The function* Ψ *of Lemma 3.2 satisfies the TDDFT system discussed in Theorem 2.1 with the quantum corrections.*

Proof. By Lemma 3.3, by relabelling if necessary, it follows that

$$\Psi_{\epsilon_n}(t) \to \Psi(t), \text{ in } L^r, \text{ uniformly } \forall t \in J, \tag{25}$$

for an arbitrary $r < 6$ selected in advance. It follows that $\Psi \in C(J; L^r)$. We now examine the equation satisfied by Ψ. By weak convergence (Lemma 3.2),

$$\lim_{n\to\infty} \int_\Omega \frac{\hbar^2}{2m} \nabla \Psi_{\epsilon_n}(\mathbf{x}, t) \cdot \nabla \zeta(\mathbf{x}) \, d\mathbf{x} = \int_\Omega \frac{\hbar^2}{2m} \nabla \Psi(\mathbf{x}, t) \cdot \nabla \zeta(\mathbf{x}) \, d\mathbf{x}. \tag{26}$$

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

$$\lim_{n\to\infty} \int_\Omega V_{\mathrm{e}}(\mathbf{x}, t, \rho_{\epsilon_n}) \Psi_{\epsilon_n}(\mathbf{x}, t) \zeta(\mathbf{x}) \, d\mathbf{x} = \int_\Omega V_{\mathrm{e}}(\mathbf{x}, t, \rho) \Psi(\mathbf{x}, t) \zeta(\mathbf{x}) \, d\mathbf{x}. \tag{27}$$

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

$$\lim_{n\to\infty} \int_\Omega V(\mathbf{x}, t) \Psi_{\epsilon_n}(\mathbf{x}, t) \zeta(\mathbf{x}) \, d\mathbf{x} = \int_\Omega V(\mathbf{x}, t) \Psi(\mathbf{x}, t) \zeta(\mathbf{x}) \, d\mathbf{x}. \tag{28}$$

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

$$\int_\Omega W * \rho_{\epsilon_n} \Psi_{\epsilon_n}(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x} \quad - \quad \int_\Omega W * \rho\ \Psi(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x} =$$

$$\int_\Omega W * \rho_{\epsilon_n}[\Psi_{\epsilon_n}(\mathbf{x},t) - \Psi(\mathbf{x},t)]\zeta(\mathbf{x})\, d\mathbf{x} \quad + \quad \int_\Omega W * [\rho_{\epsilon_n} - \rho]\Psi(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x}.$$

Each of the two rhs terms is estimated by the generalized Hölder inequality. This reduces to estimating the following two triple products of norms:

$$\|W * \rho_{\epsilon_n}\|_{L^2}\|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^3}\|\zeta\|_{L^6}, \quad \|W * [\rho_{\epsilon_n} - \rho]\|_{L^2}\|\Psi(t)\|_{L^3}\|\zeta\|_{L^6}.$$

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:

$$\lim_{n\to\infty} \int_\Omega W * \rho_{\epsilon_n} \Psi_{\epsilon_n}(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x} = \int_\Omega W * \rho\ \Psi(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x}. \tag{29}$$

The potential $\Phi_{\rm qc}$ requires the analysis of the three components introduced in section 2. For the smoothed LDA potential $\phi_\epsilon * \Phi_{\rm lda}$, we will use the triangle inequality, and we write,

$$\int_\Omega \phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n})\Psi_{\epsilon_n}(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x} - \int_\Omega \Phi_{\rm lda}(\rho)\Psi(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x} =$$

$$\int_\Omega \phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n})[\Psi_{\epsilon_n}(\mathbf{x},t) - \Psi(\mathbf{x},t)]\zeta(\mathbf{x})\, d\mathbf{x} +$$

$$\int_\Omega [\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n}) - \Phi_{\rm lda}(\rho)]\Psi(\mathbf{x},t)\zeta(\mathbf{x})\, d\mathbf{x}.$$

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

$$\|\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n})[\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^{r'}}\|\zeta\|_{L^r}, \quad \|[\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n}) - \Phi_{\rm lda}(\rho)]\Psi(t)\|_{L^{r'}}\|\zeta\|_{L^r},$$

where $r = \alpha + 2$ and r' is conjugate to r. We use the method employed in the proof of Proposition 3.2 (cf. (23)) 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,

$$\|\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n})[\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^{r'}} \leq \|\phi_{\epsilon_n} * |\Psi_{\epsilon_n}|^\alpha\|_{L^{r/\alpha}}\|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^r} \leq$$

$$\| |\Psi_{\epsilon_n}|^\alpha \|_{L^{r/\alpha}} \|\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^r} \leq \|\Psi_{\epsilon_n}\|^\alpha_{L^r} \|\Psi_{\epsilon_n}(t) - \Psi(t)]\|_{L^r},$$

which converges to zero as remarked at the beginning of the proof (see (25)). Thus, the first product of norms is convergent to zero. For the second product, we begin as before, to obtain,

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

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

$$\|\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n}) - \Phi_{\rm lda}(\rho)\|_{L^{r/\alpha}} \leq \|\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n}) - \phi_{\epsilon_n} * \Phi_{\rm lda}(\rho)\|_{L^{r/\alpha}} + \|\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho) - \Phi_{\rm lda}(\rho)\|_{L^{r/\alpha}}.$$

The first term on the rhs is bounded, via the smoothing property, by

$$\|\phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n}) - \phi_{\epsilon_n} * \Phi_{\rm lda}(\rho)\|_{L^{r/\alpha}} \leq \| |\Psi_{\epsilon_n}|^\alpha - |\Psi|^\alpha \|_{L^{r/\alpha}}.$$

The estimation of this expression requires inequality (18) with the identifications $\Psi_1 \mapsto \Psi_{\epsilon_n}, \Psi_2 \mapsto \Psi$. When the power r/α is applied to the inequality, and integration over Ω 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:

$$\lim_{n\to\infty} \int_\Omega \phi_{\epsilon_n} * \Phi_{\rm lda}(\rho_{\epsilon_n})\Psi_{\epsilon_n}(\mathbf{x}, t)\zeta(\mathbf{x})\, d\mathbf{x} = \int_\Omega \Phi_{\rm lda}(\rho)\Psi(\mathbf{x}, t)\zeta(\mathbf{x})\, d\mathbf{x}. \tag{30}$$

We now consider the Coulomb term. Again, we write

$$\int_\Omega \phi_{\epsilon_n} * \Phi_{\rm c}\Psi_{\epsilon_n}(\mathbf{x}, t)\zeta(\mathbf{x})\, d\mathbf{x} - \int_\Omega \Phi_{\rm c}\Psi(\mathbf{x}, t)\zeta(\mathbf{x})\, d\mathbf{x} =$$
$$\int_\Omega \phi_{\epsilon_n} * \Phi_{\rm c}[\Psi_{\epsilon_n}(\mathbf{x}, t) - \Psi(\mathbf{x}, t)]\zeta(\mathbf{x})\, d\mathbf{x} + \int_\Omega [\phi_{\epsilon_n} * \Phi_{\rm c} - \Phi_{\rm c}]\Psi(\mathbf{x}, t)\zeta(\mathbf{x})\, d\mathbf{x}.$$

The estimation is now straightforward. The Hölder inequality yields the two triple products for the rhs term estimates:

$$\|\phi_{\epsilon_n} * \Phi_{\rm c}\|_{L^2}\ \|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^3}\ \|\zeta\|_{L^6},\ \|\phi_{\epsilon_n} * \Phi_{\rm c} - \Phi_{\rm c}\|_{L^2}\ \|\Psi(t)\|_{L^3}\ \|\zeta\|_{L^6}.$$

The first term is convergent because of strong convergence; the second, because of the convergence of the smoothing in L^2.

The final term to estimate among the quantum correction terms is the time-history term, if present. Recall that this term is not smoothed. The term $\Phi(\cdot\,, t, \rho)$ is analyzed as follows. We have the algebraic representation,

$$\int_\Omega \Phi(\cdot\,, t, \rho_{\epsilon_n})\ \Psi_{\epsilon_n}\zeta\, d\mathbf{x} - \int_\Omega \Phi(\cdot\,, t, \rho)\ \Psi\zeta\, d\mathbf{x} =$$

$$\int_\Omega [\Phi(\cdot\,, t, \rho_{\epsilon_n}) - \Phi(\cdot\,, t, \rho)]\ \Psi_{\epsilon_n}\zeta\, d\mathbf{x} +$$

$$\int_\Omega [\Phi(\cdot\,,t,\rho)[\Psi_{\epsilon_n} - \Psi)]\zeta \; d\mathbf{x}.$$

The first term converges to zero because of the assumed uniform L^2 continuity of Φ in its third argument, while the second term is governed by the uniform convergence in L^r.

We now use (26) and (27) to conclude that

$$\lim_{n\to\infty} \langle \partial\Psi_{\epsilon_n}/\partial t, \zeta\rangle = \int_\Omega \frac{\hbar^2}{2m}\nabla\Psi(\mathbf{x},t)\cdot\nabla\zeta(\mathbf{x}) + V_{\mathrm{e}}(\mathbf{x},t,\rho)\Psi(\mathbf{x},t)\zeta(\mathbf{x})\; d\mathbf{x}.$$

However, we may deduce from Lemma 3.2 that

$$\lim_{n\to\infty} \langle \partial\Psi_{\epsilon_n}/\partial t, \zeta\rangle = \langle \partial\Psi/\partial t, \zeta\rangle, \tag{31}$$

so that Ψ solves the TDDFT system. The initial condition is a consequence of (25) in, say, L^2 for $t = 0$. □

It remains to verify the regularity class for Ψ.

Theorem 3.2. *The function* Ψ *of Theorem 3.1 satisfies*

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

Proof. We begin with the verification that $\Psi \in C(J; H^1_0)$, and make use of Proposition B.1, part (2), of appendix B. In particular, it suffices to show that

$$\int_\Omega \frac{\hbar^2}{4m}|\nabla\Psi_{\epsilon_n}|^2 \; d\mathbf{x} \to \int_\Omega \frac{\hbar^2}{4m}|\nabla\Psi|^2 \; d\mathbf{x},\; n\to\infty, \text{ uniformly in } t.$$

We use the representations contained in Lemma 3.1 as applied to Ψ_{ϵ_n}. We rewrite them as follows.

$$\mathcal{E}_n(t) = \int_\Omega \left[\frac{\hbar^2}{4m}|\nabla\Psi_{\epsilon_n}|^2 + \left(\frac{1}{4}(W * |\Psi_{\epsilon_n}|^2) + \frac{1}{2}(V + \Phi_{\epsilon_n}(\cdot\,,t,\rho_{\epsilon_n}))\right)|\Psi_{\epsilon_n}|^2\right] d\mathbf{x}, \tag{32}$$

$$\mathcal{E}_n(t) = \mathcal{E}(0) + \frac{1}{2}\int_0^t \int_\Omega [(\partial V/\partial s)(\mathbf{x},s) + \phi(\mathbf{x},s)]|\Psi_{\epsilon_n}|^2 \; d\mathbf{x}ds. \tag{33}$$

Note that the expression $\mathcal{E}_n(t)$, as defined in (32), converges uniformly in t to $\mathcal{E}(t)$, when the boundedness for $\partial V/\partial t + \phi$ is applied, due to strong convergence. The approach now is to solve for the gradient term in (32) and deduce its uniform convergence from that of each of the other terms. Because of the hypotheses made on the external potential and the time-history terms, the terms requiring analysis are the Hartree and remaining quantum correction terms. The techniques are similar to those used earlier. For the Hartree potential, we have

$$\int_\Omega W * \rho_{\epsilon_n}(t)\; \rho_{\epsilon_n}(\mathbf{x},t)\; d\mathbf{x} \quad - \quad \int_\Omega W * \rho(t)\; \rho(\mathbf{x},s)\; d\mathbf{x} =$$

$$\int_\Omega W * \rho_{\epsilon_n}(t)[\rho_{\epsilon_n}(\mathbf{x},t) - \rho(\mathbf{x},t)]\; d\mathbf{x} \quad + \quad \int_\Omega W * [\rho_{\epsilon_n}(t) - \rho(t)]\rho(\mathbf{x},t)\; d\mathbf{x}.$$

Each of the two rhs terms is estimated by the Schwarz inequality, so that we must estimate the following two products of norms:

$$\|W * \rho_{\epsilon_n}(t)\|_{L^2} \|\rho_{\epsilon_n}(t) - \rho(t)\|_{L^2}, \; \|W * [\rho_{\epsilon_n}(t) - \rho(t)]\|_{L^2} \|\rho(t)\|_{L^2}.$$

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

$$\|\rho_{\epsilon_n}(t) - \rho(t)\|_{L^2} \leq \||\Psi_{\epsilon_n}(t)| - |\Psi(t)|\|_{L^4} \||\Psi_{\epsilon_n}(t)| + |\Psi(t)|\|_{L^4},$$

which is convergent to zero as $n \to \infty$, by the strong uniform convergence. For the second product, an application of Young's inequality and the strong uniform convergence allows one to conclude that uniform convergence to zero as $n \to \infty$. Next, we consider the LDA term.

$$\int_\Omega \Phi_{\rm lda}(\rho_{\epsilon_n}(t))\rho_{\epsilon_n}(\mathbf{x},t)\, d\mathbf{x} \quad - \quad \int_\Omega \Phi_{\rm lda}(\rho(t))\rho(\mathbf{x},t)\, d\mathbf{x} =$$
$$\int_\Omega \Phi_{\rm lda}(\rho_{\epsilon_n}(t))[\rho_{\epsilon_n}(\mathbf{x},t) - \rho(\mathbf{x},t)]\, d\mathbf{x} \quad + \quad \int_\Omega [\Phi_{\rm lda}(\rho_{\epsilon_n}(t)) - \Phi_{\rm lda}(\rho(t))]\rho(\mathbf{x},t)\, d\mathbf{x}.$$

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

$$\||\Psi_{\epsilon_n}(t)|^\alpha [|\Psi_{\epsilon_n}(t)| - |\Psi(t)|]\|_{L^{r'}} \; \||\Psi_{\epsilon_n}(t)| + |\Psi(t)|\|_{L^r},$$
$$\| \, [|\Psi_{\epsilon_n}(t)|^\alpha - |\Psi(t)|^\alpha]|\Psi(t)| \, \|_{L^{r'}} \|\Psi(t)\|_{L^r},$$

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

$$\|\Psi_{\epsilon_n}(t)\|^\alpha_{L^r} \; \|\Psi_{\epsilon_n}(t) - \Psi(t)\|_{L^r} (\|\Psi_{\epsilon_n}(t)\|_{L^r} + \|\Psi(t)\|_{L^r}),$$

which converges to zero as $n \to \infty$. The second product is estimated, with the help of (18) and Hölder's inequality, as

$$\alpha \|(|\Psi_{\epsilon_n}(t)| + |\Psi(t)|)^{\alpha - 1} (|\Psi_{\epsilon_n}(t)| - |\Psi(t)|)\|_{L^{r/\alpha}} \|\Psi(t)\|^2_{L^r}, \tag{34}$$

and another application of Hölder's inequality, with $p = \alpha$ and p' conjugate to α, gives the bound,

$$\alpha \|(|\Psi_{\epsilon_n}(t)| + |\Psi(t)|)\|^{\alpha-1}_{L^r} \; \||\Psi_{\epsilon_n}(t)| - |\Psi(t)|\|_{L^r} \|\Psi(t)\|^2_{L^r},$$

so that this term also converges to zero. Finally, the Coulomb term is directly estimated via the strong convergence; we omit the details. It follows that $\Psi \in C(J; H^1_0)$.

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 ζ. We need to show that this difference satisfies a zero limit as $t \to s$, uniformly in $\|\zeta\|_{H^1_0} \leq 1$. The property just established,

$\Psi \in C(J; H^1_0)$, implies this for the gradient and external potential terms. The remaining terms can be estimated via a very useful analogy: replace the $n \to \infty$ limit in the estimates for Theorem 3.1 by the $t \to s$ limit, after constructing parallel algebraic representations. The convergence of the corresponding dominating terms holds since $\Psi \in C(J; H^1_0)$. This completes the proof. □

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

4 Uniqueness

This section is a replacement for the original section. The following theorem will be established in this section by the techniques associated with evolution operators. We first state the theorem, and then establish appropriate background, prior to providing the details of the proof. We note that the analysis presented here excludes from uniqueness the case(s) $1 < \alpha < 2$ in the representation of the LDA component of the potential.

Theorem 4.1. *Under the assumptions of this article, there is a unique weak solution of (4), where* $V_{\rm e}$ *is defined in (11). The defining properties of weak solution are described in Definition 1.1. The cases* $1 < \alpha < 2$ *are excluded.*

Some results allow for $1 < \alpha < 2$. We will be specific when these cases are excluded.

4.1 Background

The evolution operator permits the solution of the linear Cauchy problem,

$$\begin{aligned} \frac{du}{dt} + A(t)u(t) &= F(t), \\ u(0) = u_0, & \end{aligned} \tag{35}$$

on an interval $[0, T]$, with values in a Banach space. The solution is given by

$$u(t) = U(t,0)u_0 + \int_0^t U(t,s)\, F(s)\, ds, \tag{36}$$

under (strong) assumptions on u_0, F. In order that (36) hold rigorously, the evolution operators $U(t, s)$ are derived for a pair, $(A(t), X)$ and $(A(t), Y)$, where Y is continuously embedded in X and is a core subspace of the domains of $A(t)$. The operators are typically generated on X, and shown to be invariant on Y by a commutator relation. For this article, $Y = H^1_0$ and $X = H^{-1}$. This theory is due to Kato, and is developed in [17, Chapter 6]. For this article, we may use the results of [7], where the desired properties of the evolution operators were derived for Hamiltonian operators $A(t) = \hat{H}(t)$, including the kinetic term plus the external, Hartree, and time-history potentials. The Coulomb and LDA potentials were not included in that theory. It follows that any application of these results, intended to derive uniqueness, must shift the Coulomb and LDA terms into the action of $F(t)$. The theory asserts [17, Prop. 6.4.1] that there is a one-to-one correspondence between the representation (36) and the unique solution of (35) if $u_0 \in Y$ and $F \in C(J;X) \cap L^1(J;Y)$. When this hypothesis holds, the unique solution is in $C^1(J;X) \cap C(J;Y)$. However, the characterization of F in the current situation does not satisfy the required regularity. This accounts for the following method which we use. Note that we are able to use the linear theory by defining coefficients of $A(t)$ in terms of the solution itself. Further, for $\rho = |\Psi|^2$, define

$$F(\Psi) = -[\Phi_{\rm c} + \Phi_{\rm lda}(\rho)]\Psi, \tag{37}$$

and $U^\rho(t,s)$ to be the evolution operators derived in [7], based on a Hamiltonian including the kinetic term plus the external, Hartree, and time-history potentials. As defined, F fails to be in $C(J;H_0^1)$. We make use of the following smoothing.

Definition 4.1. *Consider the smoothing of section 2.1, and define*

$$F_\epsilon(\Psi) = -[\phi_\epsilon * \Phi_{\rm c} + \phi_\epsilon * \Phi_{\rm lda}(\rho)]\Psi, \ 1 \leq \alpha < 4. \tag{38}$$

This smoothing will be used to prove the following.

Lemma 4.1. *Suppose that $F(\Psi(s))$ is defined by (37). If Ψ satisfies (4), then*

$$\Psi(t) = U^\rho(t,0)\Psi_0 + \int_0^t U^\rho(t,s)\, F(\Psi(s))\, ds, \tag{39}$$

where the integral is interpreted as a member of $C^1(J;H^{-1})$, and Ψ is interpreted as a distribution. Conversely, if $\Psi \in C(J;H_0^1) \cap C^1(J;H^{-1})$ satisfies (39), then Ψ satisfies (4).

Proof. Suppose Ψ is a solution of (4), with the specified regularity. The function $F_\epsilon(\Psi)$ is a member of $C(J;H_0^1)$ and the replacement in (39) of $F(\Psi)$ by $F_\epsilon(\Psi)$ yields a solution Ψ_ϵ,

$$\Psi_\epsilon(t) = U^\rho(t,0)\Psi_0 + \int_0^t U^\rho(t,s)\, F_\epsilon(\Psi(s))\, ds, \tag{40}$$

of the adjusted equation (4). These may be thought of as nearby approximate linear equations, indexed by ϵ. We notice the important fact for the argument that the functions $\Psi_\epsilon(t)$ form a bounded family, independent of ϵ, in $C(J;H_0^1)$. We use the specific properties that the convolution terms in the definition of F_ϵ are L^∞ functions, with bound independent of ϵ, and possess L^3 derivatives, with this norm independent of ϵ.

In the smoothed case, there is a one to one correspondence between the representation and the adjusted system. As $\epsilon \to 0$, the representations Ψ_ϵ converge in $C(J;H^{-1})$ to a representation

$$\Psi_*(t) = U^\rho(t,0)\Psi_0 + \int_0^t U^\rho(t,s)\, F(\Psi(s))\, ds. \tag{41}$$

The convergence follows from [17, Prop. 7.1.1] and the argument presented now. Since the evolution operators are independent of ϵ, it suffices to estimate the norm of

$$\|F(\Psi) - F_\epsilon(\Psi)\|_{L^1(J;H^{-1})}.$$

The LDA component is estimated in $C(J;H^{-1})$ as follows. For $1 \leq \alpha < 4$, we have that $|\Psi|^\alpha \in C(J;L^{3/2})$. It follows that, for $\phi \in H_0^1$,

$$\|[F(\Psi) - F_\epsilon(\Psi)]\phi\|_{L^1} \to 0,$$

uniformly in t. This combines the generalized Hölder inequality and the properties of the smoothing in $L^{3/2}$.

The duality estimate for the Coulomb potential is carried out by a similar estimate, via the generalized Hölder inequality. All that is required is the known convergence of the smoothing in $L^{3/2}$. We conclude that (41) holds.

It remains to equate Ψ_* with Ψ. If we examine the adjusted system (4), corresponding to (40), we conclude that the family $\partial\Psi_\epsilon\partial t$ is bounded in $C(J;H^{-1})$. This is implied by the boundedness, already noted, of the family $F_\epsilon(\Psi)$ in $C(J;H_0^1)$. This in turn yields the equicontinuity required for the application of Proposition B.1 of appendix B. When this convergence result is applied to a subsequence of Ψ_ϵ, we conclude that the difference $\Psi - \Psi_*$ solves a linear initial value problem, upon cancellation of the terms involving F, for which zero is the unique solution.

For the converse, we begin with Ψ satisfying (39), and define F_ϵ as before. We again argue that Ψ satisfies (4) using the same limit analysis. □

The following is immediate from the lower semicontinuity of the norm with respect to weak convergence.

Corollary 4.1. *For the weakly convergent sequence Ψ_{ϵ_n} of the proof, we have*

$$\|\Psi\|_{C(J;H_0^1)} \leq \liminf_{n\to\infty} \|\Psi_{\epsilon_n}\|_{C(J;H_0^1)}.$$

4.2 The approximation arguments: Proof of Theorem 4.1

In order to establish uniqueness, we consider two separate equations, defined by $\hat{H}_{\rho_1}(t), F(\Psi_1(t))$, and $\hat{H}_{\rho_2}(t), F(\Psi_2(t))$, with solutions Ψ_1, Ψ_2, resp. The evolution operators are denoted by $U^{\rho_j}(t,s), j = 1,2$. The representations satisfy the lemma and are understood to be in $C(J;H^{-1})$, since the functions $F(\Psi_j)$ are in this space. Explicitly,

$$F(\Psi_j) = -[\Phi_{\rm c} + \Phi_{\rm lda}(\rho_j)]\Psi_j, \; j = 1,2. \tag{42}$$

Proposition 4.1. *Suppose that Ψ_1 and Ψ_2 are two distinct solutions of (39), where $\rho_j = |\Psi_j|^2, j = 1,2$. Suppose the respective approximations are given by (40), written here as*

$$\Psi_j^\epsilon(t) = U^{\rho_j}(t,0)\Psi_0 + \int_0^t U^{\rho_j}(t,s)\; F_\epsilon(\Psi_j(s))\; ds. \tag{43}$$

For $\alpha = 1$, or $2 \leq \alpha < 4$, there exists a constant C, not depending on ϵ, such that

$$\|\Psi_1^\epsilon(t) - \Psi_2^\epsilon(t)\|_{H_0^1)} \leq C \int_0^t \|\Psi_1(s) - \Psi_2(s)\|_{H_0^1}\, ds, \tag{44}$$

for all $t \in [0, T]$.

Proof. We begin the argument by writing the operator difference,

$$\Psi_1^\epsilon(t) - \Psi_2^\epsilon(t) = [U^{\rho_1}(t,0) - U^{\rho_2}(t,0)]\Psi_0 + \int_0^t [U^{\rho_1}(t,s) - U^{\rho_2}(t,s)]\; F_\epsilon(\Psi_1(s))\, ds \; +$$

$$\int_0^t U^{\rho_2}(t,s)\; [F_\epsilon(\Psi_1(s)) - F_\epsilon(\Psi_2(s))]\, ds. \tag{45}$$

The estimations of the first and second terms depend on the representations [17, (7.1.3)], for $g \in H_0^1$,

$$U^{\rho_1}(t,r)g - U^{\rho_2}(t,r)g = -\int_r^t U^{\rho_1}(t,s)[\hat{H}^{\rho_1}(s) - \hat{H}^{\rho_2}(s)]U^{\rho_2}(s,r)g\, ds. \tag{46}$$

Here,

$$[\hat{H}^{\rho_1}(s) - \hat{H}^{\rho_2}(s)]g = W * [\rho_1 - \rho_2]g + [\Phi(\Psi_1) - \Phi(\Psi_2)]g \tag{47}$$

is a member of $C(J; H_0^1)$ if $g \in C(J; H_0^1)$. We have employed cancellation of the kinetic term and the external potential term in (47). Both terms are readily estimated, uniformly in s, in the H_0^1 norm, the first by [20, Theorem 3.1], for $g = \Psi_0$, and the second by the hypothesis assumed for Φ, for $g = F_\epsilon(\Psi_1(s))$. After the action of the evolution operators, with respect to bounded sets in H_0^1, uniformly in $t \in J$, is taken into account, we obtain an estimate of the form (44) for these terms in (45).

For the estimation of the third term in (45), we may write the preliminary algebraic step as

$$\phi_\epsilon * |\Psi_1|^\alpha \Psi_1 - \phi_\epsilon * |\Psi_2|^\alpha \Psi_2 = \phi_\epsilon * [|\Psi_1|^\alpha - |\Psi_2|^\alpha]\Psi_1 + \phi_\epsilon * |\Psi_2|^\alpha [\Psi_1 - \Psi_2]. \tag{48}$$

The H_0^1 seminorm requires the estimation of four rhs terms for this relation as computed by the product rule for differentiation. We provide a summary analysis of each term involved in the differentiation of (48). We must show that such terms are Lipschitz in L^2, uniformly in $t \in J$. The partial derivative, with respect to x_j, of the first term has a pointwise upper bound given by

$$\alpha\phi_\epsilon * ||\Psi_1|^{\alpha-1}\partial\Psi_1/\partial x_j - |\Psi_2|^{\alpha-1}\partial\Psi_2/\partial x_j|\; (|\Psi_1|) + \phi_\epsilon * ||\Psi_1|^\alpha - |\Psi_2|^\alpha|\; (|\partial\Psi_1/\partial x_j|). \tag{49}$$

The second term of (49) is readily estimated, since the convolution factor is uniformly in L^∞ by Young's convolution inequality. In fact,

$$\|\phi_\epsilon * ||\Psi_1|^\alpha - |\Psi_2|^\alpha|\|_{L^\infty} \leq \|\phi_\epsilon\|_{L^r}\; \||\Psi_1|^\alpha - |\Psi_2|^\alpha|\|_{L^{r'}},$$

where $r = \alpha + 2$ and r' is conjugate to r. In the proof of Proposition 2.1, we showed that

$$\| |\Psi_1|^\alpha - |\Psi_2|^\alpha \|_{L^{r'}} \leq \text{const} \|\Psi_1 - \Psi_2\|_{H_0^1} \tag{50}$$

uniformly in t. The estimation of the first term in (49) requires the pointwise inequality,

$$\begin{aligned} &\alpha\phi_\epsilon * (|\ |\Psi_1|^{\alpha-1}\partial\Psi_1/\partial x_j - |\Psi_2|^{\alpha-1}\partial\Psi_2/\partial x_j\ |)|\Psi_1| \leq \\ &\quad \alpha\phi_\epsilon * (||\Psi_1|^{\alpha-1} - |\Psi_2|^{\alpha-1}||\partial\Psi_1/\partial x_j|)\ |\Psi_1| + \\ &\quad \alpha\phi_\epsilon * (|\partial\Psi_1/\partial x_j - \partial\Psi_2/\partial x_j|\ |\Psi_2|^{\alpha-1}|)\ |\Psi_1|. \end{aligned} \tag{51}$$

For both terms on the rhs of (51), we demonstrate that the terms being smoothed are uniformly in L^1. The resulting convolution yields a function in every L^p space, with bound uniformly in t. The L^2 estimation of the product with $|\Psi_1|$ is then estimated by Hölder's inequality. We now present the details.

The first term on the rhs of (51) requires a case distinction ($\alpha = 1$ is trivial):

$$1 < \alpha < 2;\ 2 \leq \alpha < 4.$$

For $2 \leq \alpha < 4$, we use (18) so that, pointwise, we have

$$||\Psi_1|^{\alpha-1} - |\Psi_2|^{\alpha-1}||\partial\Psi_1/\partial x_j| \leq (\alpha-1)\max(|\Psi_1|, |\Psi_2|)^{\alpha-2}||\Psi_1| - |\Psi_2||\ |\partial\Psi_1/\partial x_j|.$$

The application of Hölder's inequality, with indices, $1/3, 1/6, 1/2$, resp. together with Sobolev's inequality, gives the desired estimate.

For $1 < \alpha < 2$, this upper bound does not hold in general, hence this interval is excluded.

For the estimation of the second term in (51), the L^1 norm of the term being smoothed is estimated by the Cauchy-Schwarz inequality. Thus, the product of the smoothed term and $|\Psi_1|$ can again be estimated in L^2 by the Hölder inequality. The upper bound of the rhs of (50) is obtained.

We now estimate the derivative of the second term in (48). A pointwise upper bound for the partial derivative is given by

$$\alpha\phi_\epsilon * (|\Psi_2|^{\alpha-1}|\partial\Psi_2/\partial x_j|)|\Psi_1 - \Psi_2| + \phi_\epsilon * |\Psi_2|^\alpha(|\partial\Psi_1/\partial x_j - \partial\Psi_2/\partial x_j|).$$

The estimation of the first of these two terms follows the previous pattern: determination of uniform L^1 bounds for the functions being smoothed, followed by the L^2 product estimation via the Hölder inequality. The upper bound of the rhs of (50) is directly obtained. For the second of these two terms, notice that the convolution factor is uniformly L^∞ by Young's convolution inequality. The conclusion is immediate.

A typical *kth* term for the partial derivative with resp. to x_j of the Coulomb potential is bounded above pointwise:

$$|c_k|\ \phi_\epsilon * (|\cdot - x_k|^{-2})|\Psi_1 - \Psi_2| + |c_k|\ \phi_\epsilon * (|\cdot - x_k|^{-1})|\partial\Psi_1/\partial x_j - \partial\Psi_2/\partial x_j|.$$

Hölder's inequality implies the bound for the first term. The second term is directly estimated, since the convolution factor is uniformly L^∞. This completes the proof of the proposition. □

Corollary 4.2. *The uniqueness for the quantum corrected model holds for the potentials introduced in this article, except for the exclusion* $1 < \alpha < 2$ *in the LDA potential. No further boundary regularity is required.*

Proof. Since the norm is weakly lower semicontinuous, the estimate is transferred to $\|\Psi_1 - \Psi_2\|$. Gronwall's inequality implies the result. □

The uniqueness result permits a useful convergence result for the smoothing 'sequence'.

Corollary 4.3. *We assume the conditions of the uniqueness theorem. Suppose that* ϵ_n *is any positive sequence of real numbers convergent to zero. Then the sequence* Ψ_{ϵ_n}*, satisfying Proposition 2.1, converges in the norm of* $C(J; H_0^1(\Omega)) \cap C^1(J; H^{-1}(\Omega))$ *to the unique solution* Ψ *defined in Theorem 2.1.*

Proof. We use the elementary fact that, if every subsequence has a further subsequence converging to a unique limit, then the entire sequence converges to that unique limit. The first part of the proof of Theorem 3.2 demonstrates subsequential convergence in $C(J; H_0^1(\Omega))$. The arguments leading to (31) demonstrate convergence in $C^1(J; H^{-1}(\Omega))$.

□

5 Summary Remarks

We have formulated a model within the framework of time dependent density functional theory. It is a closed system model, posed on a bounded domain in $\mathbb{R}^3$ with homogeneous boundary conditions. The novelty of the article lies in the flexibility of the choice of potentials. In addition to the Hartree potential and a given external potential, we permit Coulomb potentials with fixed ionic point masses, a time-history potential, and the local density approximation (LDA), which is typically used in simulation. We have obtained existence and uniqueness for this model on a bounded domain in $\mathbb{R}^3$ and a given finite time interval. The growth of the LDA term, in terms of the exponent α, 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. Finally, Corollary 4.3 assumes significance because the smoothed solutions can be obtained via the evolution operator, and its approximations (see the cited references).

We note finally, that the case of periodic boundary conditions frequently occurs in applications. It is a topic of future study.

A Notation and Norms

We employ complex Hilbert spaces in this article.

$$L^2(\Omega) = \{f = (f_1, \dots, f_N)^T : |f_j|^2 \text{ is integrable on } \Omega\}. (f,g)_{L^2} = \sum_{j=1}^{N} \int_\Omega f_j(x)\overline{g_j(x)}\, dx.$$

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 $f_j \in H_0^1(\Omega; \mathcal{C})$, we write $f \in H_0^1(\Omega; \mathcal{C}^N)$, 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 [23]. The Banach space $C(J; H_0^1)$ is defined in the traditional manner:

$$C(J; H_0^1) = \{u : J \mapsto H_0^1 : u(\cdot) \text{is continuous}\}, \; \|u\|_{C(J;H_0^1} = \sup_{t \in J} \|u(t)\|_{H_0^1}.$$

- Since Ω is assumed to be a bounded Lipschitz domain, the standard Sobolev embedding theorems for $H_0^1(\Omega)$ hold, relative to $L^p(\Omega)$ [23].

B Subsequential Convergence for Bounded Families

In section 3.2, we applied two basic compactness results, taken from [4] and [21]. Here, we quote the underlying results for the reader's convenience. The first is cited from [4, Proposition 1.3.14(i,iii)].

Proposition B.1 (Cazenave)**.** *Let I be a bounded interval of $\mathbb{R}$, let m be a nonnegative integer, let Ω be an open subset of $\mathbb{R}^N$, and let $(f_n)_{n \in \mathbb{N}}$ be a bounded sequence of $L^\infty(I; H_0^1(\Omega)) \cap W^{1,\infty}(I; H^{-m}(\Omega))$.*

(1) Then there exist $(f_{n_k})_{k \in \mathbb{N}}$ and $f \in L^\infty(I; H_0^1(\Omega)) \cap W^{1,\infty}(I; H^{-m}(\Omega))$ such that

$$\forall t \in \bar{I}, \; f_{n_k}(t) \rightharpoonup f(t), k \to \infty, \; \text{in } H_0^1(\Omega).$$

(2) If $(f_n)_{n \in \mathbb{N}} \subset C(\bar{I}; H_0^1(\Omega))$ and $\|f_{n_k}(t)\|_{H^1} \to \|f(t)\|_{H^1}$ uniformly on I, then $f \in C(\bar{I}; H_0^1(\Omega))$ and

$$f_{n_k} \to f \text{ in } C(\bar{I}; H_0^1(\Omega)).$$

The next result is cited from [21, Theorem 2.3.14]. It is a generalized Arzela-Ascoli theorem.

Proposition B.2 (Simon)**.** *Let X be a separable metric space and Y a complete metric space, with $C \subset Y$ compact. Let $\mathcal{F}$ be a family of uniformly equicontinuous functions from X to Y with Range$(f) \subset C$ for every $f \in \mathcal{F}$. Then any sequence in $\mathcal{F}$ has a subsequence converging at each $x \in X$. If X is compact, then $\mathcal{F}$ is precompact in the uniform topology.*

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