

Operator Newton Iterative Convergence for Time Dependent Density Functional Theory

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Abstract—In a recent publication, the author has established the existence of a unique weak solution of the initial/boundary-value problem for a closed quantum system modeled by time dependent density function theory (TDDFT). We describe a Newton iteration, based upon the technique used to prove (unique) existence for the TDDFT model. We show that successive approximation at the operator level, based upon the evolution operator, is sufficient to obtain a ‘starting iterate’ for Newton’s method. We discuss the so-called quadratic convergence associated with Newton’s method. In the process, we obtain a Kantorovich type theorem for TDDFT.

I. INTRODUCTION

Time dependent density functional theory (TDDFT) dates from the seminal article [1]. A current account of the subject may be found in [2]. For density functional theory (DFT), we refer the reader to [3], [4]. Our concern is with the former only. Closed quantum systems on bounded domains of \mathbf{R}^3 were analyzed in [5], [6], via time-ordered evolution operators. The article [5] includes simulations based on approximations of the evolution operator, employing the FEAST algorithm. The article [6] extended the existence results of [5] to include weak solutions via a strict contraction argument for an operator K . TDDFT is a significant field for applications (see [7]–[11]). By permitting time dependent potentials, TDDFT extends the nonlinear Schrödinger equation ([12], [13]).

In this article, we build upon [6] by introducing a Newton iteration argument for K . In the following sections of the introduction, we familiarize the reader with the model, and summarize the basic results of [6], which serve as the starting point for the present article.

A. The Model

TDDFT includes an external potential, the Hartree potential, and the exchange correlation potential. If \hat{H} denotes the Hamiltonian operator of the system, then the state $\Psi(t)$ of the system obeys the nonlinear Schrödinger equation,

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

Here, $\Psi = \{\psi_1, \dots, \psi_N\}$ and the charge density ρ is defined by

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

For well-posedness, an initial condition,

$$\Psi(0) = \Psi_0, \quad (2)$$

consisting of N orbitals, and boundary conditions must be adjoined. We will assume that the particles are confined to a bounded region $\Omega \subset \mathbf{R}^3$ and that homogeneous Dirichlet boundary conditions hold for the evolving quantum state within a closed system. In general, Ψ denotes a finite vector function of space and time.

B. Specification of the Hamiltonian Operator

We study effective potentials V_e which are of the form,

$$V_e(\mathbf{x}, t, \rho) = V(\mathbf{x}, t) + W * \rho + \Phi(\mathbf{x}, t, \rho),$$

where, for $W(\mathbf{x}) = 1/|\mathbf{x}|$, the convolution $W * \rho$ denotes the Hartree potential, and where Φ represents a time history of ρ :

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

As explained in [2, Sec. 6.5], Φ_0 is determined by the initial state of the Kohn-Sham system and the initial state of the interacting reference system with

the same density and divergence of the charge-current.

The Hamiltonian operator then assumes the standard form,

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

C. Definition of Weak Solution

The solution Ψ is continuous from the time interval J into the finite energy space of functions which are zero on the boundary, denoted H_0^1 . One writes: $\Psi \in C(J; H_0^1)$. The time derivative is continuous from J into the dual H^{-1} . One writes: $\Psi \in C^1(J; H^{-1})$. The spatially dependent test functions ζ are arbitrary in H_0^1 . The duality bracket is denoted $\langle f, \zeta \rangle$.

Definition 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 $\Psi_0 \in H_0^1$, and if $\forall t \in J$:

$$i\hbar \left\langle \frac{\partial \Psi(\mathbf{x}, t)}{\partial t}, \zeta(\mathbf{x}) \right\rangle =$$

$$\int_{\Omega} \frac{\hbar^2}{2m} \nabla \Psi(\mathbf{x}, t) \cdot \nabla \zeta(\mathbf{x}) + V_e(\mathbf{x}, t, \rho) \Psi(\mathbf{x}, t) \zeta(\mathbf{x}) d\mathbf{x}. \quad (4)$$

D. The Associated Linear Problem

The approach to solve the nonlinear problem (4) is to define a fixed point mapping K . For each Ψ^* in the domain of K , we produce the image $K\Psi^* = \Psi$ by the following steps.

- 1) $\Psi^* \mapsto \rho = \rho(\mathbf{x}, t) = |\Psi^*(\mathbf{x}, t)|^2$.
- 2) $\rho \mapsto \Psi$ by the solution of the *associated* linear problem (4) where the potential V_e uses ρ .

In general, $\Psi \neq \Psi^*$ unless Ψ is a fixed point of K .

In order to construct a fixed point, one introduces the linear evolution operator $U(t, s)$: for given Ψ^* , set $U(t, s) = U^\rho(t, s)$ so that

$$\Psi(t) = U^\rho(t, 0)\Psi_0. \quad (5)$$

For each t , we interpret $\Psi(t)$ as a function (of \mathbf{x}, t). Moreover, the effect of the evolution operator is to obtain $\Psi = K\Psi^*$.

E. Discussion of the Evolution Operator

The evolution operator used here and in [6] was introduced in two fundamental articles [14], [15] by Kato in the 1970s. A description of Kato's theory can be found in [16]. For the application to (4), one identifies the frame space with the dual space H^{-1} and the smooth space with the finite energy space H_0^1 . A significant step is to show that the operators $(-i/\hbar)\hat{H}(\rho)$ generate contraction semigroups on the frame space, which remain stable on the smooth space. If one can demonstrate these properties, then the evolution operator exists and can be used as in (5) to retrieve the solution of the initial value problem.

F. Discussion of the Result

The following theorem was proven in [6]. We include a short appendix which describes the hypotheses under which Theorem 1 holds.

Theorem 1: There is a closed ball $\overline{B(0, r)} \subset C(J, H_0^1)$ on which K is invariant. For t sufficiently small, K defines a strict contraction. The contraction constant is independent of the restricted time interval, so that the unique fixed point can be continued globally in time. In particular, for any interval $[0, T]$, the system (4) has a unique solution which coincides with this fixed point.

The proof uses the Banach contraction mapping theorem. This requires:

- 1) The determination of a radius r such that $K\Psi^*$ is in $\overline{B(0, r)}$ when Ψ^* is in $\overline{B(0, r)}$.
- 2) K uniformly contracts the distance between elements in $\overline{B(0, r)}$.

G. A Variant of Conservation of Energy: The Domain of K

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

$$\int_{\Omega} \left[\frac{\hbar^2}{4m} |\nabla \Psi|^2 + \left(\frac{1}{4}(W * |\Psi|^2) + \frac{1}{2}(V + \Phi) \right) |\Psi|^2 \right] d\mathbf{x},$$

then the following identity holds:

$$\mathcal{E}(t) = \mathcal{E}(0) + \frac{1}{2} \int_0^t \int_{\Omega} [(\partial V / \partial s)(\mathbf{x}, s) + \phi(\mathbf{x}, s, \rho)] |\Psi|^2 d\mathbf{x} ds, \quad (6)$$

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

$$\int_{\Omega} \left[\frac{\hbar^2}{4m} |\nabla \Psi_0|^2 + \left(\frac{1}{4}(W * |\Psi_0|^2) + \frac{1}{2}(V + \Phi_0) \right) |\Psi_0|^2 \right] d\mathbf{x}.$$

The identity (6) allows the determination of r and the domain of K . The functional $\mathcal{E}(t)$ is related to the physical energy $E(t)$ of the system, defined by

$$E(t) = \langle \hat{H}(t)\Psi(t), \overline{\Psi(t)} \rangle : \\ \mathcal{E}(t) = \frac{1}{2} \left(E(t) - \frac{1}{2} \langle \Psi(t), (W * |\Psi(t)|^2) \overline{\Psi(t)} \rangle \right).$$

H. The Contractive Property: Role of the Evolution Operator

There is an integral operator identity satisfied by the evolution operator which permits the estimation of the metric distance between $K\Psi_1^*$ and $K\Psi_2^*$. Note that separate evolution operators $U^{\rho_1}(t, s)$ and $U^{\rho_2}(t, s)$ are generated, for $\rho_1 = |\Psi_1^*|^2, \rho_2 = |\Psi_2^*|^2$. One has the following identity (see [16]):

$$U^{\rho_1}\Psi_0(t) - U^{\rho_2}\Psi_0(t) = \\ \frac{i}{\hbar} \int_0^t U^{\rho_1}(t, s) [\hat{H}(s, \rho_1) - \hat{H}(s, \rho_2)] U^{\rho_2}(s, 0) \Psi_0 ds.$$

The H_0^1 -norm of the evolution operators can be bounded by a constant which depends only on the terminal time T and a technical bound related to the evolution operator construction. Thus, the essential term is:

$$\| [V_e(\rho_1) - V_e(\rho_2)] U^{\rho_2} \Psi_0 \|_{C(J; H_0^1)}.$$

This can be estimated, via contraction for sufficiently small t , in terms of the distance between Ψ_1^* and Ψ_2^* . Continuation to $t = T$ occurs in a finite number of steps.

II. PRELIMINARY BACKGROUND FOR NEWTON ITERATION

By appropriate use of successive approximation, we can determine approximations which can be shown to be in the domain of convergence of Newton's method. We discuss the essential details now. We quote a result proven in [17]. Additional references to the earlier literature can be found there.

Lemma 1: Suppose that $B_r := \overline{B(x_0, r)}$ is a closed ball in a Banach space X and S is a continuous mapping from B_r to a Banach space Z . Suppose that $u_0 \in B_{\alpha r}$ where $0 < \alpha < 1$, and $\|S(u_0)\| \leq \sigma^{-1}$. Suppose that a family $\{G_z\}$ of bounded linear operators is given, for each z in the range of S , where $G_z : Z \mapsto X$. Define the iterates,

$$u_k - u_{k-1} = -G_{u_{k-1}} S(u_{k-1}), \quad k = 1, 2, \dots$$

Let h be chosen so that $h \leq 1/2$, and set

$$t^* = (h\sigma)^{-1} (1 - \sqrt{1 - 2h}).$$

The procedure is consistent, i. e., $\{u_k\} \subset B_r$, if the inequalities,

$$\|u_k - u_{k-1}\| \leq \kappa \|S(u_{k-1})\|, \quad k \geq 1, \quad (7)$$

$$\|S(u_k)\| \leq \frac{h\sigma}{2} \|S(u_{k-1})\|^2, \quad k \geq 1, \quad (8)$$

hold for some $\kappa \leq (1 - \alpha)r/t^*$. Moreover, the sequence converges to a root u described by,

$$\|u - u_k\| \leq \frac{\kappa (1 - \sqrt{1 - 2h})^{2^k}}{h\sigma}. \quad (9)$$

Proposition 1: Let T be an operator valued function, defined on an open subset V of a Banach space X , such that T is Lipschitz continuously Fréchet differentiable on V , and such that $S'(u_0) = I - T'(u_0)$ is invertible for some $u_0 \in V$. Then there is a suitable closed ball containing u_0 for which $S'(v)$ is invertible for each v and there is a choice of σ such that the previous lemma holds with $G_v = [S'(v)]^{-1}$.

Proof: According to the Lipschitz property satisfied by T' on V , say Lipschitz constant c , we can find $\delta > 0$ such that, if $\|u_0 - v\| \leq \delta$, then

$$\| [I - T'(u_0)]^{-1} \| \| T'(u_0) - T'(v) \| \leq \tau < 1.$$

By a standard perturbation lemma [18], it follows that $I - T'(v)$ is invertible in the closed ball of radius δ , centered at u_0 . The perturbation lemma gives the uniform bound for the norms of the inverses $[S'(v)]^{-1}$:

$$\kappa := \frac{\| [S'(u_0)]^{-1} \|}{1 - \tau}.$$

This gives (7). To obtain (8), we employ a version of the fundamental theorem of calculus for Fréchet derivatives (valid in Fréchet spaces [19]):

$$S(u_k) = \int_0^1 [S'(u_{k-1} + t(u_k - u_{k-1})) - S'(u_{k-1})] (u_k - u_{k-1}) dt.$$

By estimating this integral, we obtain via (7):

$$\|S(u_k)\| \leq \frac{c}{2} \|u_k - u_{k-1}\|^2 \leq \frac{c\kappa^2}{2} \|S(u_{k-1})\|^2.$$

We may choose σ sufficiently large so that the following two inequalities hold:

$$c\kappa^2 \leq h\sigma, \quad \frac{\kappa}{h\sigma} \leq (1 - \alpha)r.$$

We thus obtain (8) and the previous lemma applies.

III. DIFFERENTIABILITY AND INVERTIBILITY FOR TDDFT

We assume that the operator defined by $\partial\Phi/\partial\rho$ satisfies, at Ψ , linearity, and admits product evaluations, satisfying

$$\|\partial\Phi/\partial\rho[\zeta\omega]\psi\|_{H_0^1} \leq C\|\zeta\|_{C(J;H_0^1)}\|\omega\|_{H_0^1}\|\psi\|_{H_0^1}$$

for arbitrary functions $\zeta, \omega, \psi \in H_0^1$ and some constant C , which depends only on the domain of Ψ .

Lemma 2: The operator K is continuously Gâteaux differentiable on $C(J; H_0^1)$, and thus continuously Fréchet differentiable. The derivative is Lipschitz continuous.

Proof: Let Ψ be a given element of $C(J; H_0^1)$ and set $\rho = |\Psi|^2$. We begin with the formula introduced in section I-H and make the identifications,

$$\rho_2 = \rho, \rho_1 = |\Psi + \epsilon\omega|^2, \text{ for } \omega \in H_0^1, \epsilon > 0.$$

This gives for the Gâteaux derivative of K at Ψ , evaluated at arbitrary ω : $K'(\Psi)[\omega] =$

$$\lim_{\epsilon \rightarrow 0} \frac{U^{\rho_1}\Psi_0(t) - U^{\rho_2}\Psi_0(t)}{\epsilon} = \frac{2i}{\hbar} \int_0^t U^\rho(t, s) \left[\text{Re}(\bar{\Psi}\omega) * W + \int_0^s \frac{\partial\phi}{\partial\rho} \text{Re}(\bar{\Psi}\omega) d\tau \right] U^\rho(s, 0) \Psi_0 ds.$$

Lipschitz continuity of the derivative (in Ψ) follows from the formula.

The following proposition addresses the invertibility at the fixed point of $I - K'(\Psi)$, on the space of functions continuous from the time interval J into H_0^1 .

Proposition 2: Suppose we denote by Ψ the unique fixed point of K . The operator,

$$S'(\Psi) = I - K'(\Psi),$$

is invertible on $C(J; H_0^1)$.

Proof:

Injective Property

We assume that there is $\omega \in C(J; H_0^1)$, such that

$$S'(\Psi)[\omega] = (I - K'(\Psi))\omega = 0.$$

We can apply Gronwall's inequality to the estimate,

$$\|\omega(\cdot, t)\| \leq C \int_0^t \|\omega(\cdot, s)\| ds,$$

where the norm is the H_0^1 norm, and C is a fixed constant. Gronwall's inequality yields immediately that $\omega \equiv 0$.

Surjective Property

We use a fixed point argument (even though the problem is linear). Suppose f is given in $X = C(J; H_0^1)$. We consider the equation,

$$S'(\Psi)[\psi] = (I - K'(\Psi))\psi = f,$$

for $\psi \in X$. This is equivalent to a fixed point for

$$L\psi = K'(\Psi)\psi + f.$$

L is seen to be a strict contraction for t sufficiently small. By continuation, we obtain a fixed point ψ . If we now combine Propositions 1 and 2 with Lemma 2, we obtain the central result of the paper.

Theorem 2: Under the hypotheses reviewed in the appendix, the TDDFT model admits of Picard iteration in order to obtain a starting iterate in the domain of convergence of Newton's method at the operator level. More specifically, the iterate is defined classically as in Proposition 1 and satisfies the convergence estimates of Lemma 1.

IV. CONCLUSION

The current article suggests successive approximation, proven in [6], combined with operator Newton iteration. The operator foundation for this analysis was demonstrated in [17]. This article permits an approximate inverse for the derivative mapping, and future study will consider extensions defined by approximation theory. Suggestions for very effective approximation methods may be found in [20]. Such methods allow for a more computable version of Theorem 2.

APPENDIX

We make the following assumptions for the existence/uniqueness theory described in Theorem 1.

- The integrand ϕ is assumed measurable and bounded in its arguments.
- Φ is assumed continuous in t into H^1 and bounded in t into $W^{1,3}$. The continuity into H^1 is consistent with the zero-force law as defined in [2, Eq. (6.9)]:

$$\int_{\Omega} \rho(\mathbf{x}, t) \nabla \Phi(\mathbf{x}, t, \rho) = 0.$$

- Furthermore, the following smoothing condition is assumed, expressed in a (uniform) Lipschitz norm condition: $\forall t \in [0, T]$,

$$\begin{aligned} & \|[\Phi(t, |\Psi_1|^2) - \Phi(t, |\Psi_2|^2)]\psi\|_{H^1} \leq \\ & C\|\Psi_1 - \Psi_2\|_{H^1}\|\psi\|_{H_0^1}. \end{aligned}$$

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

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