Mathematical Consistency of Time Dependent Density Functional Theory with Kohn-Sham Potentials

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Introduction

We consider the closed quantum systems analyzed in [1]. The TDDFT model dates from the seminal article [2]. If we denote by $\hat{H}$ the Hamiltonian operator of the system, then the state $\Psi(t)$ of the system obeys the nonlinear Schrödinger equation,

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

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

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

For well-posedness, an initial condition,

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

consisting of N ground states, 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.

Specification of the Hamiltonian operator

We study potentials which are of the form,

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

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 [3, 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,

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

Theorem. Under appropriate assumptions, there is a unique weak solution of (1) on any specified time interval [0,T]. The solution may be characterized as a fixed point of a mapping defined by the evolution operator.

Remarks

- The theory developed here does not (yet) extend to 'time dependent current density functional theory' (TDCDFT). This is discussed in [3].
- The evolution operators employed are capable of discretization and simulation [1]. The theory is due to Kato [4, Ch. 6].

References

[1] J.W. Jerome and E. Polizzi, Discretization of time dependent quantum systems: Real time propagation of the evolution operators. Appl. Anal. **93** (2014), 2574–2597.
[2] E. Runge and E.K.U. Gross, Density functional theory for time dependent systems. Physical Review Letters **52** (1984), 997–1000.
[3] C. A. Ullrich, Time-Dependent Density-Functional Theory: Concepts and Applications. Oxford University Press, 2012.
[4] J.W. Jerome, *Approximation of Nonlinear Evolution Equations*, Academic Press, New York, 1983.

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