Stochastic Time-Dependent Current-Density-Functional Theory

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A time-dependent current-density-functional theory for many-particle systems in interaction with arbitrary external baths is developed. We prove that, given the initial quantum state \(|\Psi_0\rangle\) and the particle-bath interaction operator, two external vector potentials \(A(r,t)\) and \(A'(r,t)\) that produce the same ensemble-averaged current density, \(\bar{j}(r,t)\), must necessarily coincide up to a gauge transformation. This result greatly expands the applicability of time-dependent density-functional theory to open quantum systems, and allows for first-principles calculations of many-particle time evolution beyond Hamiltonian dynamics.

Time-dependent density-functional theory (TDDFT) \([1]\) is becoming an important tool in the study of the dynamics of many-particle systems \([2, 3]\). The theory, which extends the applicability of ground-state density-functional theory \([2, 3]\) to non-equilibrium problems, rests solidly on a theorem proved by Runge and Gross (RG) \([1]\). The RG theorem shows that, for a fixed initial quantum state \(|\Psi_0\rangle\), the external time-dependent scalar potential \(V_{ext}(r,t)\) acting on the many-particle system is uniquely determined (up to a constant) by the time-dependent single particle density \(n(r,t)\). It was subsequently realized by Vignale and Kohn \([4]\) that, since the exchange-correlation (xc) scalar potential in TDDFT is a strongly non-local functional of the density, a theory formulated in terms of the current density \(j(r,t)\) allows for a local gradient expansion of the xc vector potential \(A_{xc}(r,t)\). This theory goes under the name of time-dependent current density-functional theory (TDCDFT) \([4, 5]\), and has been applied to a variety of contexts ranging from optical spectra of solids \([6]\) and atoms \([7]\), to dielectric properties of polymers \([8]\), to transport in nanoscale junctions \([9, 10, 11, 12]\), to the study of systems with memory and/or dissipation \([13, 14, 15]\).

Despite the enormous success that the above time-dependent density-functional based methods have been experiencing, one needs to acknowledge that they can strictly deal with particle systems evolving under Hamiltonian dynamics. There is, however, a large class of physical problems where one needs to consider interactions with an external bath. Examples include dephasing of a quantum system coupled to a boson field, spontaneous emission due to coupling with the zero-point energy fluctuations, non-radiative decay, etc. A TDCDFT able to deal with such open-system problems, without reverting to consider the microscopic dynamics of the bath, would be a tremendous asset in the study of many-particle problems beyond Hamiltonian dynamics \([23]\).

In this Letter, we introduce such a theory which we name “Stochastic TDCDFT” and prove a fundamental existence theorem comparable to the theorems of TDDFT and TDCDFT. Our theorem allows us to rigourously map the many-particle dynamics in the presence of an external bath into an effective single-particle dynamics in the presence of the same bath.

In non-equilibrium quantum statistical mechanics, the effect of a bath is effectively described using stochastic processes: the fluctuations introduced by the presence of the bath are accompanied by dissipative effects in the particle dynamics. Our starting point is therefore the stochastic time-dependent Schrödinger equation, which reads (\(\hbar = 1\)) \([16]\)

\[
\partial_t \Psi(r,t) = -i \hat{H}(t) \Psi(r,t) - \frac{1}{2} \hat{V}^\dagger \hat{V} \Psi(r,t) + \hat{\ell}(t) \Psi(r,t)
\]  

where \(\ell\) describes a stochastic process, and \(\hat{V}\) is an operator which describes the bath, and its interaction with the many-particle system \([26]\). In the following we will assume that \(\hat{V}\) is independent of time and uniform in space. These restrictions can be easily removed (provided one adds the requirement that \(\hat{V}\) admits a series expansion in time) and our theorem remains valid, but we maintain them for the sake of simplicity. A few examples of the operator \(\hat{V}\) can be found in \([16]\). The Hamiltonian operator is

\[
\hat{H}(t) = \sum_i \left[ \hat{p}_i + eA(\hat{r}_i,t) \right]^2 / 2m + \frac{1}{2} \sum_{i \neq j} U(\hat{r}_i - \hat{r}_j),
\]  

with \(\hat{A}(\hat{r}_i,t)\) a vector potential and \(U(\hat{r}_i - \hat{r}_j)\) a potential describing two-particle interactions \([27]\). Without loss of generality the stochastic process, \(\ell(t)\), is chosen such that it has both zero ensemble average and \(\delta\)–autocorrelation, i.e.,

\[
\overline{\ell(t)} = 0; \quad \overline{\ell(t)\ell(t')} = \delta(t - t'),
\]  

where the symbol \(\overline{}\) indicates the average over a statistical ensemble of identical systems all prepared in the same initial quantum state \(|\Psi_0\rangle\) \([28]\).

The last term on the rhs of Eq. (1) describes precisely the “fluctuation” induced by the bath; the second term is the compensating “dissipative” part. Equation (1), with the stochastic process defined in \([23]\), is a stochastic differential equation which preserves the ensemble-averaged norm and the orthogonality between any two
states, $\psi_\alpha(r, t)$ and $\psi_\beta(r, t)$, $\int dr \psi_\alpha^*(r, t)\psi_\beta(r, t) = \delta_{\alpha,\beta}$. Ensemble-averaged orthonormality can be easily demonstrated by integrating Eq. (1) in a small interval of time $\Delta t$, taking the scalar product between two states, and ensemble-averaging this product using the properties [5] [10].

Given any operator in the Heisenberg representation, its average over the initial state is obtained via the standard quantum mechanical definition $\langle \hat{O}(t) \rangle \equiv \langle \Psi_0 | \hat{O}(t) | \Psi_0 \rangle$. If $\hat{O}$ is the density operator $\hat{n}$, one can easily show that the dynamics induced by Eq. (1) preserves the ensemble-averaged particle number, i.e., the quantity $\mathcal{N} \equiv \int dr \langle \hat{n}(r, t) \rangle \equiv \int dr \langle \hat{\psi}^\dagger(r, t)\hat{\psi}(r, t) \rangle$ is a constant of motion [10]. Physically, this corresponds to the case in which the bath may exchange particles with the system, while keeping their average number constant. Similarly, one can prove that, given any observable $\hat{O}(t)$, its ensemble average time evolution, $\overline{\hat{O}(t)}$, satisfies [16]

$$\partial_t \overline{\hat{O}} = i \left[ \hat{H}, \overline{\hat{O}} \right] - \frac{1}{2} \hat{V}^\dagger \hat{V} \overline{\hat{O}} - \frac{1}{2} \overline{\hat{O}} \hat{V}^\dagger \hat{V} + \hat{V}^\dagger \hat{O} \hat{V}. \quad (4)$$

In passing, we note that if $\overline{\hat{O}}, \hat{V} \equiv 0$ at any time, then the equation of motion (3) reduces to $\partial_t \overline{\hat{n}} = i \left[ \hat{H}, \overline{\hat{n}} \right]$, i.e., the presence of the bath does not affect, on average, the observable $\overline{\hat{O}}$. Since, in general, $|\hat{H}, \hat{V}| \neq 0$, the total energy is not conserved. Additionally, if $\hat{\rho}$ is the density matrix operator, from (1) we get the well-known (Lindblad) quantum master equation

$$\partial_t \hat{\rho} = -i \left[ \hat{H}, \hat{\rho} \right] - \frac{1}{2} \hat{V}^\dagger \hat{V} \hat{\rho} - \frac{1}{2} \hat{\rho} \hat{V}^\dagger \hat{V} + \hat{V}^\dagger \hat{\rho} \hat{V}. \quad (5)$$

We define the ensemble-average density

$$\overline{n(r, t)} = \langle \hat{n}(r, t) \rangle, \quad (6)$$

and current density

$$\overline{j(r, t)} = \langle \hat{j}(r, t) \rangle, \quad (7)$$

where the current operator is defined as $\hat{j}(r, t) = \frac{1}{2} \sum_i \{ \delta(r - \hat{r}_i), \hat{v}_i \}$ with

$$\hat{v}_i = \frac{\hat{p}_i + eA(\hat{r}_i, t)}{m}, \quad (8)$$

the velocity operator of particle $i$, and the symbol $\{ \hat{A}, \hat{B} \} \equiv (\hat{A}\hat{B} + \hat{B}\hat{A})$ is the anticommutator of any two operators $\hat{A}$ and $\hat{B}$.

We have now set the stage to formulate and demonstrate the following theorem.

**Theorem:** Consider a many-particle system described by the dynamics in Eq. (1) with the many-body Hamiltonian given by Eq. (2). Let $\overline{n}(r, t)$ and $\overline{j}(r, t)$ be the ensemble-averaged single-particle density and current density, respectively, with dynamics determined by the external vector potential $A(r, t)$ and bath $\hat{V}$. Under reasonable physical assumptions, given an initial condition $|\Psi_0\rangle$, and an assigned bath operator $\hat{V}$, another external potential $A'(r, t)$ which gives the same ensemble-averaged current density, must necessarily coincide, up to a gauge transformation, with $A(r, t)$.

**Proof:** To prove this statement we follow a line of reasoning commonly used to establish similar theorems in TDDFT and TDCDFT [17] [18]. Let us assume that the same ensemble-averaged density $n(r, t)$ and current density $\overline{j}(r, t)$ are also obtained from another many-particle system with Hamiltonian

$$\hat{H}'(t) = \sum_i \left[ \hat{p}_i + eA'(\hat{r}_i, t) \right]^2 \frac{1}{2m} + \frac{1}{2} \sum_{i \neq j} U'(\hat{r}_i - \hat{r}_j), \quad (9)$$

evolving from an initial state $|\Psi_0'\rangle$ and following the stochastic Schrödinger equation (1) with the same bath operator $\hat{V}$. $|\Psi_0'\rangle$ gives, in the primed system, the same initial current and particle densities as in the unprimed system.

The core of the demonstration is as follows: By writing the equations of motion for $\overline{j}(r, t)$ determined by both $A(r, t)$ and $A'(r, t)$, we obtain an equation of motion for the potential difference $\Delta A(r, t) = A(r, t) - A'(r, t)$. We then prove that $\Delta A(r, t)$ is completely determined by the initial condition via a series expansion in time about $t = 0$. Finally, if the two systems coincide then the unique solution is $\Delta A \equiv 0$, up to a gauge transformation.

The equation of motion for the ensemble-averaged current density is easily obtained from the equation of motion of the current density operator. From Eq. (4) we get

$$\partial_t \overline{j}(r, t) = \frac{\overline{n}(r, t)}{m} \partial_t A(r, t) - \frac{\overline{j}(r, t)}{m} \times [\nabla \times A(r, t)]$$

$$+ \frac{\langle \hat{F}(r, t) \rangle}{m} + \frac{\langle \hat{G}(r, t) \rangle}{m} \quad (10)$$

where we have defined

$$\hat{G}(r, t) = \hat{V}^\dagger \hat{j}(r, t) \hat{V} - \frac{1}{2} \hat{j}(r, t) \hat{V}^\dagger \hat{V} - \frac{1}{2} \hat{V}^\dagger \hat{V} \hat{j}(r, t),$$

$$\hat{F}(r, t) = -\sum_{i \neq j} \delta(r - \hat{r}_i) \nabla_j U(\hat{r}_i - \hat{r}_j) + m \nabla \cdot \hat{\sigma}(r, t)$$

(11)

with the stress tensor $\hat{\sigma}(r, t)$ given by

$$\hat{\sigma}_{i,j}(r, t) = -\frac{1}{4} \sum_k \{ \hat{v}_i, \{ \hat{v}_j, \delta(r - \hat{r}_k) \} \}. \quad (12)$$

The first two terms on the rhs of Eq. (10) describe the effect of the applied electromagnetic field on the dynamics of the many-particle system; the third is due to particle-particle interactions while the last one is the “force” density exerted by the bath on the system.
Equations similar to Eqs. (8) – (12) can now be written for the system with the vector potential \( \mathbf{A}'(\mathbf{r}, t) \). Similar force terms \( \mathcal{F}' \) and \( \mathcal{G}' \) appear in these new equations. \( \mathcal{F}' \) and \( \mathcal{G}' \) differ from the same forces in the unprimed system, since the initial state, the external vector potentials and the velocity \( \dot{v} \) are different. By assumption, the ensemble-averaged current and particle densities are the same in the two systems, thus

\[
\partial_t j_i(r, t) = \frac{n(r, t)}{m} \partial_t \mathbf{A}'(\mathbf{r}, t) - \frac{j_i(r, t)}{m} \times [\nabla \times \mathbf{A}'(\mathbf{r}, t)] + \frac{\mathcal{F}'(r, t)}{m} + \frac{\mathcal{G}'(r, t)}{m}.
\]

Taking the difference of Eqs. (10) and (13) we arrive at

\[
\frac{n(r, t)}{m} \partial_t \Delta \mathbf{A}(r, t) = \frac{\mathcal{J}(r, t)}{m} \times [\nabla \times \Delta \mathbf{A}(\mathbf{r}, t)] + \Delta Q(r, t)
\]

where \( \Delta \mathbf{A}(\mathbf{r}, t) \equiv \mathbf{A}'(\mathbf{r}, t) - \mathbf{A}(\mathbf{r}, t) \) and \( \Delta Q(r, t) \equiv Q'(r, t) - Q(r, t) \) with \( Q(r, t) = \langle \mathcal{F}(r, t) \rangle + m\langle \mathcal{G}(r, t) \rangle \), and \( Q'(r, t) \) the same quantity but in the primed system.

We now need to prove that Eq. (14) admits only one solution, i.e., \( \Delta \mathbf{A}(\mathbf{r}, t) \) is completely determined by the averaged dynamics of the current and particle densities, once the coupling with the bath, \( \dot{V} \), is assigned. To this end we expand Eq. (14) in series about \( t = 0 \) and obtain an equation for the \( l \)-th derivative of the vector potential \( \Delta \mathbf{A}(\mathbf{r}, t) \). That one can expand this equation in a time series about \( t = 0 \) follows immediately from the analyticity of the vector potential and Eq. (11). We thus arrive at the equation

\[
\Delta \mathbf{A}_{l+1}(\mathbf{r}) = - \sum_{k=0}^{l} (k+1) \Delta \mathbf{A}_{k+1}(\mathbf{r}) + \Delta Q_l(\mathbf{r}) + \sum_{k=0}^{l} j_{-k}(\mathbf{r}) \times [\nabla \times \Delta \mathbf{A}_k(\mathbf{r})]
\]

where, given an arbitrary function of time \( f(t) \), we have defined the series expansion \( J_i(\mathbf{r}) \equiv \frac{\mathcal{J}(r, t)}{m} \times [\nabla \times \Delta \mathbf{A}(\mathbf{r}, t)] \). We are now left to prove the rhs of Eq. (15) does not contain any term \( \Delta \mathbf{A}_{l+1}(\mathbf{r}) \). This follows from the fact that the dynamics of any ensemble-averaged operator is given by Eq. (11). Indeed, this implies that the \( l \)-th time derivative of any operator can be expressed in terms of its derivatives of order \( k < l \), time derivatives of the Hamiltonian of order \( k < l \), and powers of the operators \( \dot{V} \) and \( \dot{V}^\dagger \). The time derivatives of the Hamiltonian contain time derivatives of the vector potential \( \mathbf{A}(\mathbf{r}, t) \), but always of order \( k < l \). Then on the rhs of Eq. (15) no time derivative of order \( l + 1 \) appears. Equation (15) can be thus viewed as a recursive relation for the time derivatives of the vector potential \( \Delta \mathbf{A}(\mathbf{r}, t) \). To complete the recursion procedure we only need to assign the initial value \( \Delta \mathbf{A}_0(\mathbf{r}) = \mathbf{A}(\mathbf{r}, t = 0) - \mathbf{A}'(\mathbf{r}, t = 0) \).

Since in the unprimed and primed systems the densities and current densities are, by hypothesis, equal, the initial condition is simply given by \( \frac{n(r, t = 0)}{m} \Delta \mathbf{A}_0(\mathbf{r}) = \langle \mathcal{F}_0(\mathbf{r}, t = 0) \rangle - \langle \mathcal{F}'_0(\mathbf{r}, t = 0) \rangle \rangle \rangle \).

The same considerations as in Ref. [18] about the finiteness of the convergence radius of the time series (15) apply to our case as well. We rule out the case of a vanishing convergence radius by observing that it seems implausible that the smooth (in the ensemble-averaged sense) dynamics induced by Eq. (1) can introduce a dramatic explosion of the initial derivatives of \( \Delta \mathbf{A}(\mathbf{r}) \). If this holds, the expansion procedure (15) can be iterated from the convergence radius time onward. We have then proved that Eq. (15) completely determines the vector potential \( \Delta \mathbf{A}(\mathbf{r}, t) \) and thus, since \( \mathbf{A}(\mathbf{r}, t) \) is assumed known, it determines \( \mathbf{A}'(\mathbf{r}, t) \) uniquely, up to a gauge transformation.

To finalize our proof, we consider the case in which \( U = U' \) and \( |\Psi_0\rangle = |\Psi'_0\rangle \). If this holds, \( \Delta \mathbf{A}_0(\mathbf{r}) \equiv 0 \). Then the recursion relation admits the unique solution \( \Delta \mathbf{A}(\mathbf{r}, t) \equiv 0 \) for any \( l \), and at any instant of time \( t \) we have \( \mathbf{A}(\mathbf{r}, t) = \mathbf{A}'(\mathbf{r}, t) \) (still up to a gauge transformation).

Discussion: The theorem we proved provides solid grounds for the development of a Kohn-Sham (KS) scheme for the study of the dynamics of open quantum systems, i.e., many-particle systems coupled to an external bath. Indeed, we can choose as the primed system a non-interacting one, i.e., \( U' = 0 \). We thus find that any (ensemble-averaged) current density that is interacting \( \mathbf{A} \)-representable is also non-interacting \( \mathbf{A} \)-representable [18, 19]. (It is worth to point out that, in general, the current density is not \( \mathbf{A} \)-representable, i.e., the mapping between current density and external scalar potential is not invertible [20].) The dynamics of the many-particle system can then be mapped into the dynamics of an ”auxiliary” KS Slater determinant evolving according to

\[
\partial_t \Psi^{KS}(\mathbf{r}, t) = -i \hat{H}^{KS}(\mathbf{r}, t) \Psi^{KS}(\mathbf{r}, t) - \frac{1}{2} \hat{V}^\dagger \hat{V} \Psi^{KS}(\mathbf{r}, t) + \hat{V} \ell(t) \Psi^{KS}(\mathbf{r}, t),
\]

where

\[
\hat{H}^{KS}(t) = \sum_i \left[ \hat{p}_i + e \mathbf{A}(\hat{r}_i, t) + e \mathbf{A}_{xc}(\hat{r}_i, t) \right]^2 \frac{2m}{2m} + V_H(\hat{r}_i, t),
\]

with \( \mathbf{A}_{xc}(\mathbf{r}, t), |\Psi_0\rangle, \hat{V} \) the xc vector potential, and \( V_H(\mathbf{r}, t) \) the Hartree potential. In actual calculations, one can apply, as a starting point, available approximations for \( \mathbf{A}_{xc} \), [4, 21, 22] but more work in identifying the role of the bath on the correlations of the system is necessary.
We conclude by noting that the present theorem shows that given a bath and its interaction with the system, the ensemble-averaged expectation value of any observable can be written in terms of the ensemble-averaged current density. The most simple example is the average total current flowing in the system. Indeed, given an arbitrary surface \( S \), we can easily show that the ensemble-averaged total current \( I_S(t) \) that flows across that surface is

\[
I_S(t) = \int_S \mathbf{j}(\mathbf{r}, t) \cdot d\mathbf{S} = \int_S \mathbf{j}^{KS}(\mathbf{r}, t) \cdot d\mathbf{S} = I_S^{KS}(t),
\]

i.e., it is equal to the ensemble-averaged KS current \( I_S^{KS}(t) \), where \( \mathbf{j}^{KS}(\mathbf{r}, t) \) is the (ensemble-averaged) sum of expectation values of the one-electron current-density operator in the populated KS states \( \psi_{i}^{KS}(\mathbf{r}, t) \).

The result of Eq. (18) extends the one proved in Ref. 24 for finite and isolated systems [3] to the case in which the system is coupled to a bath. However, we note that the theorem we have proved in this paper is not limited to any specific boundary condition on the current density, and is thus valid for infinite systems as well.

In summary, we have introduced a TDCCDFT theory for many-particle systems in interaction with arbitrary external baths. We have named it Stochastic TDCCDFT. We have proved the uniqueness (up to a gauge transformation) of the external vector potential, once the initial condition and the bath operator are assigned. This theory greatly expands the applicability of density-functional-based methods to systems evolving beyond Hamiltonian dynamics. We thus expect it will be of great value in the study of several physical problems, ranging from optics to transport.

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[24] See, e.g., M.A.L. Marques, C.A. Ullrich, F. Nogueira, A. Rubio, K. Burke, and E. K. U. Gross, in Time-Dependent Density Functional Theory, vol 706 of Lecture Notes in Physics (Springer, Berlin, 2006), for an up-to-date state-of-the-art of the theory and its applications.
[25] A first step in this direction was taken in K. Burke, R. Car, and R. Gebauer, Phys. Rev. Lett. 94, 146803 (2005). Here TDCCDFT is used in combination with a master equation to study electronic transport in the presence of dissipation. However, due to the dependence of the Kohn–Sham Hamiltonian \( H_{KS} \) on the density, a Kohn–Sham master equation can not be easily derived, because for any operator \( \hat{O} \) one has in general \( [H_{KS}, \hat{O}] \neq [\hat{H}_{KS}, \hat{O}] \).
[26] The operator \( \hat{V} \) can always be chosen traceless. Moreover, we can generalize the theory to more than one bath, each described by an operator \( \hat{V}_{\alpha} \), by solving \( \partial_t \hat{\Psi}(\mathbf{r}, t) = \left[-i\hat{H} - \frac{1}{2} \sum_{\alpha} \hat{V}_{\alpha} \hat{\Psi}(\mathbf{r}, t) + \sum_{\alpha} \hat{V}_{\alpha} \hat{\ell}_{\alpha}(t) \right] \hat{\Psi}(\mathbf{r}, t) \), with \( \hat{\ell}_{\alpha}(t) = 0 \) and \( \hat{\ell}_{\alpha}(t) \delta_{\alpha\beta} \delta(t - t') \).
[27] We are working in a gauge in which the dynamical external scalar potential is zero at any time.
[28] The theorem may not necessarily hold for stochastic processes with arbitrary higher moments.