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Foundations of stochastic time-dependent current-density functional theory for open quantum systems: Potential pitfalls and rigorous results

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We clarify some misunderstandings on the time-dependent current density functional theory for open quantum systems we have recently introduced [M. Di Ventra and R. D'Agosta, *Phys. Rev. Lett.* **98**, 226403 (2007)]. We also show that some of the recent formulations attempting to improve on this theory suffer from some inconsistencies, especially in establishing the mapping between the external potential and the quantities of interest. We offer a general argument about this mapping, that applies to any density functional theory, showing that it must fulfil certain “dimensionality” requirements.

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I. INTRODUCTION

Experience teaches us that no physical system can be considered as completely closed, in the sense that some degrees of freedom are either too many to consider explicitly, or their microscopic features are of no interest, and only their macroscopic thermodynamic properties are of importance. These degrees of freedom are then treated as baths or reservoirs of the system of interest, making the latter “open”. Nonetheless, the theory of open quantum systems is relatively new, starting with the pioneering works by Zwanzig and Nakajima,^{1,2} who worked out the theory in terms of the statistical operator (density matrix), and more recently with the formulation of the theory of open quantum system in terms of a state vector (“wavefunction”).^{3,4}

In parallel, another theory, Density Functional Theory (DFT), developed since the first papers by Kohn and co-workers,^{5,6} has changed the way we think in terms of many-body quantum mechanical systems. Indeed, the successes of DFT are countless, and the theory has now become a standard for many calculations of electronic structure, electron transport, materials properties, and chemical reactions.^{7–12}

The attempt to bring together the two theories appears therefore completely natural.^{13–17,19,25} In particular, the present authors have proved a theorem that establishes a one-to-one correspondence between a vector potential acting on an open quantum system and its current density,^{14,15} a theorem that was later extended to the correlated motion between electrons and (in principle quantum) ions.^{16,18} This theory, which we named stochastic time-dependent current-DFT (STDCDFT), then allows for the solution of a many-body open quantum system with effective single-particle equations, a significant computational simplification.

Although still in its infancy, STDCDFT for open quan-

tum systems has received a certain amount of attention, especially for its promise to expand the field to systems previously inaccessible within standard DFT. However, there is also some confusion regarding its foundations which is mostly evident in recent developments, for instance a new result that shows how in principle we can mimic the dynamics of an open quantum system by reverting to the analysis of a closed quantum system made of non-interacting particles.^{17,19} This result would further dramatically reduce the computational cost for evaluating the dynamics of the open quantum system.

The reason for this reduction is simple. In general, the open quantum system could be described in two ways. On the one hand, we can use the many-body statistical operator, or the single-particle reduced density matrix. However, for large systems, the number of degrees of freedom, i.e., the dimensionality of the operator we need to consider, scales badly although some schemes can be devised to cure this scaling.²⁰ On the other hand, a formulation in terms of a stochastic state vector is computationally cheaper than a density matrix one, but nonetheless it requires the consideration of a statistical ensemble of replicas of the system. Larger the ensemble, finer will be the physical information we can extract from the dynamics. Instead, if we could describe the dynamics of the open system with a closed non-interacting system, we would have reduced a complex and expensive problem to a size similar to that we usually tackle with our present computational means.

Obviously,—a situation common in DFT—we cannot expect to have access to the complete physical information about the open quantum system. We must accept that our non-interacting *doppelgänger* will be able to describe only some quantity of choice, normally either the single-particle density or the single-particle current density. In principle, any other physical observable can be extracted provided we know its expression in terms of these “fundamental” quantities. However, in practice, we rarely

know how to write such observables in terms of the density or the current density, thus limiting the range of physical quantities that can be actually computed. Our goal for this paper is to both clarify some of the theoretical foundations of STDCDFT that do not seem to have been appreciated in the literature, as well as to show that some of the recent attempts to extend this theory^{17,19} suffer from inconsistencies, in particular those related to the mapping between external potentials and the basic quantities of any density functional theory.

II. ON THE MAPPING BETWEEN VECTOR POTENTIAL AND DENSITIES

Let us then start by noting that the choice about which quantity we want to base our theory on is usually made *a priori*. For this reason we usually talk about DFT or Current-DFT.^{10,21} For closed systems, Current-DFT is regarded as a more complete theory than DFT since, via the continuity equation, one can calculate the single particle density starting from the single-particle current density. The situation is instead more complex in open quantum systems since, as we are going to show in the following, *prima facie* the continuity equation would seem not to uniquely determine the single-particle density, given the single-particle current density.

Let us then begin by recalling that in the theory of open quantum systems, the equation of motion for the ensemble-averaged particle density, $n(r, t)$, is given by (see, e.g., Ref. 22)

$$\partial_t n(r, t) = -\vec{\nabla} \cdot \vec{j}(r, t) + \mathcal{F}_B(r, t) \quad (1)$$

where $\mathcal{F}_B(r, t)$ describes the density modulation induced by the presence of the bath(s) and $\vec{j}(r, t)$ is the ensemble-averaged current density. Notice that in this theory we do not allow for the system to exchange particles with the environment. Eq. (1) can be obtained from the master equation of motion for the density matrix, $\hat{\rho}$ ($\hbar = 1$)

$$\partial_t \hat{\rho}(t) = -i \left[\hat{H}(t), \hat{\rho}(t) \right] + \int_{t_0}^t dt' K(t, t') \hat{\rho}(t'). \quad (2)$$

Here, $K(t, t')$ is a memory kernel that describes the action of the bath(s) on the system—provided it is chosen to preserve the positivity of the density matrix at any given time^{1,2,23–25} and $\hat{H}(t)$ is the Hamiltonian of the system that evolves under the action of an external vector potential (possibly time dependent), $\vec{A}(r, t)$, and in the presence of a particle-particle interaction operator \hat{W} ,

$$\hat{H}(t) = \frac{1}{2m} \sum_i \left(\hat{p}_i + e\vec{A}(\hat{r}_i, t) \right)^2 + \hat{W} \quad (3)$$

We work here in a gauge where the scalar potential has been set to zero at all times.

In general, little is known about the term \mathcal{F}_B appearing in the continuity equation (1). Although not strictly

necessary for the considerations we make later, it would be useful if \mathcal{F}_B could be written as the divergence of some current. This would immediately imply the conservation of the total number of particles. Recently, Gebauer and Car have shown that this is indeed the case in the Markov approximation for certain couplings between the system and the environment.^{26,27} More recently a formal condition on current conservation and its use to test the validity of the approximations made to arrive at the master equation has been proposed.²⁸ If \mathcal{F}_B is the divergence of some current—this extra current being generated by the exchange of momentum and energy with the external environment—then the continuity equation assumes the usual physical meaning and particle number conservation is guaranteed. Moreover, one could assume that

$$\mathcal{F}_B = -\nabla \cdot \left(\frac{e\vec{C}(r, t)}{m} n(r, t) \right), \quad (4)$$

where without loss of generality we have written the extra current as $en(r, t)\vec{C}(r, t)/m$. If this were the case, the total current $\vec{j}_T(r, t) = \vec{j}(r, t) + en(r, t)\vec{C}(r, t)/m$ fulfills the standard continuity equation,

$$\partial_t n(r, t) = -\nabla \cdot \vec{j}_T(r, t). \quad (5)$$

In the total current, $\vec{C}(r, t)$ appears then to play the role of an extra vector potential, effectively inducing a diamagnetic current in the system, $en(r, t)\vec{C}(r, t)/m$. Interestingly, in Ref. 17 $\vec{C}(r, t)$ has been dubbed “leakage potential”, although we would like to point out that its definition from Eq. (4) essentially prevents any leakage, i.e. loss of particles, from the system.

In standard TDCDFT for closed quantum systems, where the extra term in Eq. (1) is not present, i.e., $\mathcal{F}_B(r, t) = 0$, the continuity equation establishes an important link between the single-particle current density $\vec{j}(r, t)$ and the single-particle density $n(r, t)$. Indeed, this equation allows for expressing the latter in terms of the former via a simple time integration. This seems a trivial point, but let us clearly state it: The continuity equation, when $\mathcal{F}_B(r, t) = 0$ can be used together with the initial conditions to *uniquely* determine the single-particle density given the single-particle current density. The basic theorem of TDCDFT therefore establishes a one-to-one mapping between the current density and the vector potential applied to the system, once the initial conditions are provided.^{10,12,29} In fact, whenever one discusses the uniqueness of the mapping between the single-particle density and single-particle current density with the external vector potential, it is implicitly understood that the particle density is obtained from the current density via the continuity equation. If expressed in terms of the set of densities $\{\vec{j}(r, t), n(r, t)\}$, the mapping between this set and the vector potential would be redundant, in the sense that one of the variables—usually $n(r, t)$ —can be determined by $\vec{j}(r, t)$. As an exercise, in the following

subsection we will detail the proof of the standard TDCDFT theorem for closed system, without the requirement that the single particle density is the same in the Kohn-Sham and in the real system.

The situation becomes a bit trickier whenever $\mathcal{F}_B(r, t) \neq 0$. In this case one needs the expressions of both $\vec{j}(r, t)$ and $\mathcal{F}_B(r, t)$ to be able to determine the single-particle density. Indeed, written in this way the problem corresponds to the solution of a linear differential equation with non-constant coefficients. Then, if one knows $\vec{j}(r, t)$ and $\mathcal{F}_B(r, t)$ we just need to perform a simple time integration to obtain $n(r, t)$. From a TDCDFT formulation of the theory then, as it has been shown,^{14,15} one can build a one-to-one mapping between the (average) current density and the external vector potential, provided the initial condition and the bath operators are fixed. In Ref.14 the thesis of the theorem is that there is a one-to-one mapping between the current density and the vector potential, but in the proof we have implicitly assumed that the particle density could be determined from the continuity equation. In Ref. 15 we have clarified the role of the continuity equation and assumed that the single particle density is determined uniquely by the current density – unfortunately for open quantum system we do not have a proof of this statement. After that, we have formulated the theorem by including the single particle density in the mapping. In this respect, our mapping could appear identical to the one presented in Ref. 17, with the distinction however that for us the single particle density is fixed uniquely by the current density.

Here, we stress that the inclusion in the mapping of the density as a variable along with the current density—as for example it has been tried in Ref. 17—is not just redundant, but simply incorrect. Indeed, we will show in the following that the mapping between the set of current and particle densities and the *vector* potential is not unique. Our argument can be extended to other cases. For example, along the same lines we can prove that the mapping between the current density and the *scalar* potential cannot be one-to-one. One of us (RD) has already presented another proof of the same result, in particular showing that the current density is in general non-V-representable.³⁰ This, therefore, precludes the access to all components of the current density by standard TDDFT. Our following argument also sets a necessary condition for building any density functional theory, in terms of establishing a one-to-one correspondence between two physical quantities. This condition states that the two quantities must have the same “dimensions”: a vector with a vector, a scalar with a scalar, and so on. Any other mapping will be either redundant or wrong.

A. A theorem of standard TDCDFT

Before proceeding we would like to extend the Vignale’s proof of the fundamental theorem of TDCDFT²⁹.

In this extension, we explicitly assume that the continuity equation is not valid, and therefore that the single particle densities in the real and Kohn-Sham system do not coincide. In this section, with a little abuse of notation, we will define $n(r, t)$ and $\vec{j}(r, t)$ the single particle density and current density, while elsewhere these define the corresponding ensemble-average quantities. We will prove that: *Theorem (mapping between vector potential and current density)* Consider a many-particle system described by the time-dependent Hamiltonian

$$\hat{H}(t) = \sum_i \left[\frac{1}{2m} \left(\hat{p}_i + \vec{A}(\hat{r}_i, t) \right)^2 \right] + \sum_{i < j} U(\hat{r}_i - \hat{r}_j) \quad (6)$$

where $\vec{A}(\hat{r}, t)$ is a given external vector potential, which is an analytic function of time in a neighborhood of $t = 0$, and $U(\hat{r}_i - \hat{r}_j)$ is a translationally invariant two-particle interaction. Let $\vec{j}(r, t)$ be the current density that evolves under \hat{H} from a given initial state $|\Psi(0)\rangle$. Then, under reasonable assumptions, the same current density can be obtained from another many-particle system, with Hamiltonian

$$\hat{H}'(t) = \sum_i \left[\frac{1}{2m} \left(\hat{p}_i + \vec{A}'(\hat{r}_i, t) \right)^2 \right] + \sum_{i < j} U'(\hat{r}_i - \hat{r}_j), \quad (7)$$

starting from an initial state $|\Psi'(0)\rangle$ which yields the same current density as $|\Psi(0)\rangle$ at time $t = 0$. The potential $\vec{A}'(r, t)$, is uniquely determined by $\vec{A}(r, t)$, $|\Psi(0)\rangle$, and $|\Psi'(0)\rangle$, up to gauge transformations of the form

$$\vec{A}'(r, t) \rightarrow \vec{A}'(r, t) + \nabla \Lambda(r, t), \quad (8)$$

where Λ is an arbitrary regular function of r and t , which satisfies the initial condition $\Lambda(t = 0) = 0$.

Proof: The proof of this theorem follows in the same footsteps of similar proofs already present in the literature for TDDFT and TDCDFT.^{29,31,32} More advanced proofs have been recently suggested especially for the basic theorem of TDDFT³³ and for TDCDFT on a lattice³⁴. One of the advantages of these novel proofs is that they remove the analyticity requirement around the initial time. Although research in this direction is currently under way⁴³, at the moment there is not a similar proof for TDCDFT so we will use the standard way to prove this theorem. We assume that the same current density $\vec{j}(r, t)$ is also obtained from another many-particle system with Hamiltonian

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

evolving from an initial state $|\Psi'_0\rangle$ and following the time-dependent Schrödinger equation. $|\Psi'_0\rangle$ gives, in the primed system, the same initial current density as in the unprimed system.

The core of the demonstration is as follows: We write the equations of motion for $\vec{j}(r, t)$ determined by both

$\vec{A}(r, t)$ and $\vec{A}'(r, t)$, and compare these two equations to obtain an equation for $\vec{A}'(r, t)$ in terms of $\vec{A}(r, t)$. We then prove that $\vec{A}'(r, t)$ 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 $\vec{A}(r, t) = \vec{A}'(r, t)$ up to a gauge transformation.

The equation of motion for the current density is easily obtained from the equation of motion of the current density operator. A standard derivation leads us to

$$\partial_t \vec{j}(r, t) = \frac{n(r, t)}{m} \partial_t \vec{A}(r, t) - \frac{\vec{j}(r, t)}{m} \times [\nabla \times \vec{A}(r, t)] + \frac{\vec{\mathcal{G}}(r, t)}{m} \quad (10)$$

where we have defined the vector of the internal forces

$$\vec{\mathcal{G}}(r, t) = - \left\langle \sum_{i \neq j} \delta(r - \hat{r}_i) \nabla_j U(\hat{r}_i - \hat{r}_j) \right\rangle + m \left\langle \nabla \cdot \hat{\vec{\sigma}}(r, t) \right\rangle \quad (11)$$

with the stress tensor $\hat{\vec{\sigma}}(r, t)$ 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 right hand side of Eq. (10) describe the effect of the applied electromagnetic field on the dynamics of the many-particle system while the third is due to particle-particle interactions.

Equations similar to Eqs. (10) – (12) can now be written for the system with the vector potential $\vec{A}'(r, t)$. A

similar force term $\vec{\mathcal{G}}'$ appears in these new equations and differs from the same forces in the unprimed system, since the initial state, the external vector potentials and the velocity \hat{v} are different. By assumption, the current density is the same in the two systems, thus

$$\partial_t \vec{j}(r, t) = \frac{n'(r, t)}{m} \partial_t \vec{A}'(r, t) - \frac{\vec{j}(r, t)}{m} \times [\nabla \times \vec{A}'(r, t)] + \vec{\mathcal{G}}'(r, t). \quad (13)$$

In Eq. (13) we have explicitly taken into account the different single particle density in the primed system. Taking the difference of Eqs. (10) and (13) we arrive at

$$\begin{aligned} n'(r, t) \partial_t \vec{A}'(r, t) &= n(r, t) \partial_t \vec{A}(r, t) \\ &\quad - \vec{j}(r, t) \times [\nabla \times (\vec{A}(r, t) - \vec{A}'(r, t))] \\ &\quad + m \vec{\mathcal{G}}(r, t) - m \vec{\mathcal{G}}'(r, t). \end{aligned} \quad (14)$$

We now need to prove that Eq. (14) admits only one solution, i.e., $\vec{A}'(r, t)$ is completely determined by the dynamics of the current density. 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 $\vec{A}'(r, t)$. That one can expand this equation in a time series about $t = 0$ follows immediately from the analyticity of the vector potential and the properties of the Schrödinger equation. We thus arrive at the equation

$$\begin{aligned} n'_0(r)(l+1)\vec{A}'_{l+1}(r) &= n_0(r)(l+1)\vec{A}_{l+1}(r) - \sum_{k=0}^{l-1} (k+1) \left(n'_{l-k}(r)\vec{A}'_{k+1}(r) - n_{l-k}(r)\vec{A}_{k+1}(r) \right) \\ &\quad + m \vec{\mathcal{G}}_l(r) - m \vec{\mathcal{G}}'_l(r) + \sum_{k=0}^l j_{l-k}(r) \times [\nabla \times (\vec{A}'_k(r) - \vec{A}_k(r))] \end{aligned} \quad (15)$$

where, given an arbitrary function of time $f(r, t)$, we have defined the series expansion $f_l(r) \equiv \frac{1}{l!} \left. \frac{\partial^l f(r, t)}{\partial t^l} \right|_{t=0}$.

It is important to point out here that the initial density $n'_0(r) = n'(r, t = 0)$ is determined solely by the initial conditions of the primed system. This quantity is therefore accessible to us, since we assume that the initial conditions are assigned both in the primed and unprimed systems. We are now left to prove that the right hand side of Eq. (15) does not contain any term $A'_{l+1}(r)$. This follows directly from the structure of the time-dependent Schrödinger equation. Indeed, this implies that the l -th time derivative of any operator can be expressed in terms of its derivatives of order $k < l$, and time derivatives of

the Hamiltonian of order $k < l$. The time derivatives of the Hamiltonian do contain time derivatives of the vector potential $\vec{A}(r, t)$, but always of order $k < l$. Then on the right hand side 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 $\vec{A}'(r, t)$. To complete the recursion procedure we only need to assign the initial value $\vec{A}'(r, t = 0)$. Since in the unprimed and primed systems the current densities are, by hypothesis, equal, the initial condition is simply given by $n'(r, t = 0)A'_0(r) = n(r, t = 0)A_0(r) +$

$\langle \Psi_0 | \hat{j}_p(r, t=0) | \Psi_0 \rangle - \langle \Psi'_0 | \hat{j}_p(r, t=0) | \Psi'_0 \rangle$, where $\hat{j}_p(r) = (1/2m) \sum_i \{ \hat{p}_i, \delta(r - \hat{r}_i) \}$ is the paramagnetic current density operator.

The same considerations as in Ref. 29 about the finiteness of the convergence radius of the time series (15) apply to our case as well. To finalize our proof, we consider the case in which $U = U'$ and $|\Psi_0\rangle = |\Psi'_0\rangle$. If this holds, $A'_0(r) = A_0(r)$. Then the recursion relation admits the unique solution $\tilde{A}'_l(r) \equiv \tilde{A}_l(r)$ for any l , and at any instant of time t we have $A(r, t) = A'(r, t)$ (still up to a gauge transformation). On the other hand, we can assume that $U' = 0$, so that the primed system is made of non-interacting particles, *i.e.* the Kohn-Sham system.

This theorem shows that the standard TDCDFT can be reformulated in terms of *only the current density*. The presence of the continuity equation makes the proof more easy and natural. We can therefore conclude that *any* theorem of TDCDFT that includes the current and particle density in the map with the vector potential creates a redundant mapping since the continuity equation uniquely determines the single-particle density.

The proof of the above theorem also gives us an important tool. Since proofs of the theorem of STDCDFT and any theorem of a time-dependent current-density functional theory for open systems, are essentially based on this very same proof, we can extend it immediately to the theory of open quantum system. This shows that there is a one-to-one mapping between the ensemble-averaged current density and the external vector potential.

B. A simple counterexample

In fact, we can provide a simple counter argument to the existence of a mapping between the vector potential and the set of functions given by the particle and current densities. For this mapping to be non-redundant we need to assume for example that the particle density is not determined by the sole knowledge of the current density. So in the following we are assuming that it is not possible to establish a connection between the particle density and the current density. This would be the case in the DFT for open quantum system if the function F_B cannot be expressed as a functional of the current and particle densities or, if such a functional expression exists, the now-non-linear continuity equation cannot be solved to give the particle density.

Let us therefore consider the extremely simple case in which the response functions of the system are constant in time and in space, and that the (full) response is linear. We can easily generalize this counterexample to the case of response functions that are local in both space and time. Moreover, since the Fourier transform creates a unique mapping between function in real and momentum space, this counterexample can be generalized to the response to a monochromatic external vector potential, thus making it applicable to the case of a uniform free electron gas subject to an external perturbation.^{10,30} For

this system we can write the following response equations (for simplicity, in the next few equations we drop the spatial and temporal dependence of j and n)

$$\begin{pmatrix} j_x \\ j_y \\ j_z \\ n \end{pmatrix} = \hat{\Gamma} \begin{pmatrix} A_x \\ A_y \\ A_z \end{pmatrix} \quad (16)$$

where Γ is a 4×3 matrix of constant coefficients of the system response. The existence of a one-to-one mapping between the particle and current densities and the vector potential would then suggest that we can write a similar equation, starting from the particle and current densities

$$\begin{pmatrix} A_x \\ A_y \\ A_z \end{pmatrix} = \hat{\Gamma}' \begin{pmatrix} j_x \\ j_y \\ j_z \\ n \end{pmatrix} \quad (17)$$

where now Γ' is a 3×4 matrix. Combining these results it follows that

$$\hat{\Gamma} \hat{\Gamma}' = 1_4; \quad \hat{\Gamma}' \hat{\Gamma} = 1_3, \quad (18)$$

where 1_N is the $N \times N$ identity matrix.

We can now prove that this mapping is not one-to-one. Since Eq. (17) is a system of 3 equations in 4 variables, we can always find two distinct sets of current and particle densities providing the same vector potential. Therefore an infinite number of solutions can be found. To show this we proceed in this way: Assume that \vec{j}_0, n_0 are a set of current and particle densities, and that \vec{A}_0 is the resulting vector potential obtained from (17). Now let us consider the particle density $n = n_0 + \Delta n$. We want to show that we can find another current density $j = j_0 + \Delta j$ which together with n gives, via (17), the same vector potential. Since Δn is arbitrary we can choose as initial condition $\Delta n(r, t=0) = 0$, therefore the two sets of current and particle densities do satisfy the same initial conditions. To prove our point, it is enough to prove that the system of equations

$$\hat{\Gamma}' \begin{pmatrix} j_{0,x} \\ j_{0,y} \\ j_{0,z} \\ n_0 \end{pmatrix} = \hat{\Gamma}' \begin{pmatrix} j_x \\ j_y \\ j_z \\ n \end{pmatrix}, \quad (19)$$

or the equivalent

$$\hat{\Gamma}' \begin{pmatrix} \Delta j_x \\ \Delta j_y \\ \Delta j_z \\ \Delta n \end{pmatrix} = 0 \quad (20)$$

admits at least one solution. This is again trivially true, since this is a system of 3 equations in 4 variables. We can now assume the density Δn assigned and find, if the 3×3 submatrix of Γ' is invertible, the corresponding current density that satisfies Eq. (20). If that submatrix of Γ' is not invertible, then we will be able to find more than one solution for Δj , thus reinforcing our statement. The solution found in this way can also be made to fulfill the initial condition $\Delta \vec{j}(r, t=0) = 0$.

C. The role of the continuity equation

It is also worth pointing out that the standard proof of the theorem for STDCDFT¹⁴, and –as we have also shown before– of the theorem of standard TD-CDFT,²⁹ does *not* require that the particle density in the auxiliary system is equal to the particle density in the original one. Indeed, in the standard TD-CDFT proof one uses the standard continuity equation to *infer* the equality between the two densities. In our proof¹⁴, we postulated it from the uniqueness of the solution of the non-linear continuity Eq. (1), with $\mathcal{F}_B(r, t)$ a functional of the current density. In those proofs one only needs to determine the n -th time derivative of all quantities, and only the $(n + 1)$ -th time derivative of the vector potential. The n -th time derivative of the particle density is obtained from Eq. (1) (or, in a closed system from the same equation with $\mathcal{F}_B = 0$). That equation, however, does not contain any $(n + 1)$ -th time derivative. Therefore, the equation for the vector potential in the auxiliary system we use in our proof¹⁴ is still a recursive relation, with a unique solution provided the initial conditions. (See section II A for more details.) The theorem then guarantees the one-to-one correspondence between external vector potential and ensemble-averaged current density, leaving open the (possibly quite difficult) task of obtaining the ensemble-averaged density from the current density, when the continuity equation is given by Eq. (1).

This one-to-one mapping, implies that \mathcal{F}_B is a functional of the current density $\vec{j}(r, t)$, the initial conditions and the coupling between the system and the environment. This ultimately implies that– by the knowledge of this functional–we could uniquely determine the single-particle density. This is however a common situation with DFT. The theorems of DFT offer a solid foundation for certain calculations by guaranteeing that one quantity can be exactly obtained from a *doppelganger* system. The other physical quantities have to be derived from the first one, usually a difficult–if not hopeless–task.

We now want to critically examine Theorem 3 in Ref. 17, where the main result of that paper is presented: it is possible to construct a closed non-interacting quantum system that mimics the dynamics of $n(r, t)$ and $\vec{j}(r, t)$ of the real open interacting system. In the following, we will assume that $\mathcal{F}_B(r, t)$ could be written as the divergence of $en(r, t)\vec{C}(r, t)/m$ as in Eq. (4). We want to point out that, one can in fact find *many* (possibly infinite) closed non-interacting quantum systems that reproduce the dynamics of the exact current and particle densities. One can show that this is *not* in contrast with the general theory of TDCDFT. Indeed, we can show that the *total* current in the closed KS system is uniquely determined by the total vector potential, given by the sum of the KS vector potential and $\vec{C}(r, t)$. However, the particle and the current density $\vec{j}(r, t)$ do not share this property, and we can find infinitely many closed non-interacting KS systems that reproduce these quantities.

Indeed, in the proof of the Theorem 3 in Ref. 17 it is stated that the vector

$$\vec{C}(r, t) = -\frac{m}{en(r, t)} \int d^3r \left(\frac{\partial n(r, t)}{\partial t} + \vec{\nabla} \cdot \vec{j}(r, t) \right) \quad (21)$$

can be uniquely determined by Eqs. (1) and (4) once a boundary condition in space that fixes an arbitrary function of time is assigned.

However, the above equation is not the unique solution. For instance, easily fulfilling the assigned boundary condition, one can add the curl of an arbitrary vector, $\vec{g}(r, t)$, to the leakage potential $\vec{C}(r, t)$ and still satisfy Eq. (21). Indeed, it is easily proven that $\vec{C}'(r, t) = \vec{C}(r, t) + (\vec{\nabla} \times \vec{g}(r, t))/n(r, t)$ still satisfies Eqs. (1) and (4), if $\vec{C}(r, t)$ satisfies the same equation. If we now continue with the proof and use $\vec{C}'(r, t)$ we arrive at a new vector potential $\vec{A}'_{KS}(r, t)$ that gives the same $n(r, t)$ and $\vec{j}(r, t)$ as the couple $\vec{A}_{KS}(r, t)$ and $\vec{C}(r, t)$. This ambiguity reflects the fact that we are trying to mimic the effect of a scalar function – the term $\mathcal{F}_B(r, t)$ in Eq. (1) – with a vector function, $\vec{C}(r, t)$, without imposing strict boundary conditions (BCs) on $\vec{C}(r, t)$.

A simple solution to the aforementioned problem may appear by setting $\vec{\nabla} \times \vec{C}(r, t) \equiv 0$. However, it is important to realize that the imposition of certain boundary conditions on the dynamics of these quantities has a direct impact on the uniqueness of the results. For example, assuming that $\vec{C}(r, t)$ reaches a certain uniform limit when $|r| \rightarrow \infty$, might be *inconsistent* with the bath operator acting on the true many-body system. Indeed, certain bath operators can be strongly non-local in space, effectively transferring charge from one region of space to another, with the two arbitrarily far from each other. Therefore, the BCs on $\vec{\nabla} \times \vec{C}(r, t)$ have not a clear physical origin or relation to any physical observable. Fixing their value to obtain one solution appears utterly arbitrary.

Notice that a similar problem appears also in the case of the standard theorem of TDDFT (see for example Ref. 32) where the additional boundary condition $n(r)\vec{\nabla}\Delta V(r, t) \rightarrow 0$ when $|r| \rightarrow \infty$ is added to the proof. However, in this case, while in principle this condition is arbitrary and one may choose another condition, this choice is motivated by physical arguments that are valid for a wide range of systems. Further investigation of the role of this boundary condition on the validity of the theorem for standard TDDFT has been carried out in Refs. 35, 33, and 36. The same considerations instead do not apply to all the components of the vector $\vec{C}(r, t)$.

III. A MAPPING THEOREM: FROM OPEN TO CLOSED QUANTUM SYSTEM

These ambiguities derive again from the attempt to find a mapping between the vector potential and the par-

ticle and current densities, when the continuity equation is not valid. In this attempt, one needs to include another scalar quantity to make that mapping meaningful. The choice of a vector quantity like $\vec{C}(r, t)$ in Eq. (21) is ill-posed and leads to another ambiguity. On the other hand, if we are to gain any physical insight on the particle density, we need to obtain reliable approximations of the leakage potential. Instead, if one accepts that the quantity of interest is the current density, the leakage potential must be a functional of the current density and therefore we could solve the continuity equation for the particle density. For this reason, we can prove the following theorem (see below about some caveats regarding the use of a density matrix vs. a state vector formulation)

Theorem A (unique mapping from open to closed system): Consider the dynamics of a many-body system in contact with an external environment. Assume the evolution of the density matrix is given by Eq. (2) with the full many-body Hamiltonian (3). Then under reasonable physical assumptions, there exists a *closed* non-interacting auxiliary many-body Kohn-Sham system which starting from given initial conditions for the state of the many-body open system, evolves according to

$$i\partial_t|\Psi_{KS}(t)\rangle = \hat{H}_{KS}(t)|\Psi_{KS}(t)\rangle \quad (22)$$

with Hamiltonian

$$\hat{H}_{KS}(t) = \sum_i \left(\frac{\hat{p}_i + e\vec{A}_{KS}(\hat{r}_i, t)}{2m} \right)^2 \quad (23)$$

and reproduces the dynamics of the current density of the original many-body *open* system,

$$\vec{j}_{KS}(r, t) = \vec{j}(r, t). \quad (24)$$

The proof of this theorem is identical to those already present in the literature,^{14,15,25} and with little modifications to the one we propose in Sec. II A, we are therefore not reproducing it here. The only difference is that the current density of the original many-body open system follows the dynamics induced by the many-body Hamiltonian and by the external environment.^{14,15,25} The single-particle density can be obtained from Eq. (1), once the leakage potential is written as a functional of $\vec{j}(r, t)$. The uniqueness of the closed quantum system then follows from the fact that two closed systems which share the same single-particle current density and initial conditions do coincide.²⁹

It is also interesting to note that the possibility of studying the dynamics of an open system by using a TD-CDFT closed system, has been presented in the past. For example in Ref. 37 the dynamics of a 2D electron gas coupled with the electrons confined in a 1D quantum well is investigated. The quantum well, via the Coulomb interaction, acts as an external forcing field that provides energy to the 2D gas. The dynamics of the latter is then described by an effective vector potential which contains no reference to the 1D well. One can show that

the 2D electron gas will relax to a steady state regime. A similar analysis has been performed for a 1D electron gas confined in a quantum well by using the Vignale-Kohn functional for TDCDFT.³⁸ In that case, the system relaxes to the ground state.^{39,40} The KS energy is lost to those degrees of freedom that the Vignale-Kohn functional does not take into account in describing the dynamics of the many-body system through some kind of common variable like the single-particle current density. It should not be surprising then that in this case the KS energy could be given a physical interpretation as the maximum work that could be extracted from the system.⁴⁰ More recently, Tokatly⁴¹ has presented a mapping between an open quantum system, a many-body electron system in contact with the radiation of a cavity, with a KS closed system for the investigation of the dynamics of the electrons, together with an alternative proof of the theorem we present in sec. II A based on the use of a non-linear Schrödinger equation (see Ref. 41 and references therein).

Finally, we want to comment on a fundamental but important issue. As we have discussed at length in our previous publications,^{14,15} and at the beginning of this paper, one has two choices to describe an open quantum system: in terms of the density matrix or the state vector. These two formulations are generally equivalent after the observables calculated with the state vector are averaged over the many replicas of the system built to reproduce the mixed state dynamics proper of an open quantum system, being the state vector formulation an unraveling of the density matrix one. However, the density matrix approach suffers from two drawbacks which do not make it a solid starting point for a formulation of DFT for open quantum systems. This is due to both the possible loss of positivity of the density matrix if an equation of motion of such quantity is employed with the Hamiltonian and/or bath operator(s) dependent on time,⁴² and the fact that the KS Hamiltonian does depend on internal degrees of freedom. Starting from the master equation formulation of the same problem, one needs to exclude from the outset the possibility that the Hamiltonian of any auxiliary system with different interaction potential (and hence the KS Hamiltonian) depends on the internal degrees of freedom. Otherwise, for such a system no closed density-matrix equation can be obtained. In other words, one needs to start from an hypothesis that constitutes part of the final thesis. It is only when one starts from a stochastic Schrödinger equation for the *state vector* that one can prove that the *exact* KS Hamiltonian depends only on the average current density^{14,15}. In view of this criticism, we can easily reformulate the theorem A by starting from the stochastic Schrödinger equation as we have done in Ref. 14.

IV. CONCLUSIONS

In conclusion, we hope we have clarified many misunderstandings regarding a DFT for open quantum systems and its theoretical foundations. We have also shown some of the pitfalls of recent theories that have been advanced to improve on such theory. Such pitfalls originate from a simple, but yet not fully appreciated point in the DFT community, namely that one cannot map vector potentials with single-particle densities, a trivial consequence of the fact that there is no one-to-one correspondence between a vector and a scalar.

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