Causality and Symmetry in Time-Dependent Density-Functional Theory

Robert van Leeuwen

Department of Theoretical Physics, University of Lund, Sölvegatan 14A, S-22362 Lund, Sweden

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We resolve an existing paradox regarding the causality and symmetry properties of response functions within time-dependent density-functional theory. We do this by defining a new action functional within the Keldysh formalism. By functional differentiation the new functional leads to response functions which are symmetric in the Keldysh time contour parameter, but which become causal when a transition to physical time is made. The new functional is further used to derive the equations of the time-dependent optimized potential method. [S0031-9007(97)05233-2]

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Time-dependent density-functional theory (TDDFT) [1–7] provides a rigorous and useful method for calculating properties of many-particle systems in time-dependent external fields. TDDFT has been applied to a wide variety of physical problems both within the linear response regime and beyond [4]. The rigorous foundations of TDDFT were first laid down by Runge and Gross [8] who proved a 1-1 correspondence between the time-dependent external field $v(\mathbf{r}t)$ and the time-dependent density $n(\mathbf{r}t)$, for many-body systems evolving from a fixed initial state. Within TDDFT, one further introduces an auxiliary noninteracting system, known as the Kohn-Sham system, with the same density $n(\mathbf{r}t)$ as the fully interacting system. The Runge-Gross theorem applied to a noninteracting system then says that the external potential v_s of the Kohn-Sham system is uniquely determined by the density. If we subtract from v_s both the Hartree potential and the external potential of the interacting system we obtain the exchange-correlation potential v_{xc} which incorporates all the exchange and correlation effects. This quantity can therefore, by the above construction, be defined without invoking a variational or action principle. However, having an action principle is desirable as it provides an elegant derivation of the Kohn-Sham one-electron equations and a systematic way of deriving approximations to v_{xc}. The original work by Runge and Gross [8] already provided a derivation of the Kohn-Sham equations from an action principle using the action

$$A[n] = \int_{t_0}^{t_1} dt \langle \Psi[n] | i\partial_t - \hat{H}(t) | \Psi[n] \rangle . \qquad (1)$$

It was later discovered [3,4] that this definition of the action leads to a paradox when calculating response functions. On the one hand, response functions like $f_{xc}(\mathbf{r}t, \mathbf{r}'t') = \delta v_{xc}(\mathbf{r}t)/\delta n(\mathbf{r}'t')$ must be causal, i.e., be zero for $t < t'$. On the other hand, v_{xc} should be obtained as the functional derivative $v_{xc}(\mathbf{r}t) = \delta A_{xc}/\delta n(\mathbf{r}t)$ of the exchange-correlation part A_{xc} of the action functional. Interchanging the order of differentiation then yields $f_{xc}(\mathbf{r}t, \mathbf{r}'t') = \delta^2 A_{xc}/\delta n(\mathbf{r}t)\delta n(\mathbf{r}'t') = f_{xc}(\mathbf{r}'t', \mathbf{r}t)$. Hence, we find that f_{xc} must be a symmetric function of its space-time arguments. The symmetry and causality requirements clearly contradict each other. This problem is not specific to the action functional given by Eq. (1) but applies to all twice differentiable action functionals defined in physical time. The main purpose of this Letter is to show how this paradox can be resolved by use of the Keldysh formalism.

A further problem with the action functional in Eq. (1) is related to the treatment of its boundary conditions. In order to derive the time-dependent Schrödinger equation (TDSE) from the action functional, one has to enforce the boundary conditions $\delta\Psi(t_0) = \delta\Psi(t_1) = 0$ on the variations of the wave function. Within TDDFT the action functional A is only defined on the set of v-representable wave functions, i.e., wave functions which satisfy a TDSE. On this set variations $\delta\Psi$ are always caused by potential variations δv. Therefore, since the TDSE is first order in time, the variation $\delta\Psi(t)$ at times $t > t_0$ is completely determined by the boundary condition $\delta\Psi(t_0) = 0$. We are thus no longer free to specify a second boundary condition at a later time t_1. This leads to a nonvanishing boundary term when making the variation δA. The problem is usually treated by using a convergence factor $\exp(\epsilon t)$ in the definition of the action functional and moving one boundary to $-\infty$. This procedure, however, introduces a difficult problem associated with the interchange of the functional differentiation and $\epsilon \to 0$ limits.

In this Letter, we will introduce an action functional without the above-mentioned problems. First of all, our new functional does not explicitly contain the time-derivative operator ∂_t, and thus no boundary terms appear when performing variations. Second, we use the time contour method due to Keldysh [9] in which the physical time t is parametrized by an underlying parameter τ, called pseudotime. This procedure was originally introduced by Keldysh in order to obtain an elegant treatment of nonequilibrium systems in terms of many-body Green functions [9–13]. We will use the same procedure in the definition of our action functional. Higher functional derivatives of the new action functional will lead to response functions which are symmetric in the

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Keldysh time contour parameter. Transforming back to physical time t then yields the desired causal, i.e., retarded response functions in terms of t.

The Keldysh contour is defined by parametrizing the physical time $t(\tau)$ in terms of a pseudotime τ in such a way that if τ runs from τ_i to τ_f then t runs from t_0 to $\tilde{t}$ and from $\tilde{t}$ back to t_0. The value of $\tilde{t}$ can be chosen arbitrarily as long as physical quantities are calculated at earlier times. In practice one often takes $\tilde{t} = +\infty$ [9]. The actual form of the parametrization is irrelevant since and the final results are independent of it. The initial state of the system at time t_0 is given by the wave function Ψ_0. The evolution of this state in pseudotime is governed by the Schrödinger equation

$$[it'(\tau)^{-1}\partial_\tau - \hat{H}(\tau)]\,|\Psi(\tau)\rangle = 0\,, \tag{2}$$

where $t'(\tau) = dt/d\tau$. The Hamiltonian $\hat{H}(\tau)$ is given by $\hat{H}(\tau) = \hat{T} + \hat{U}(\tau) + \hat{W}$ where $\hat{T}$ represents the kinetic energy operator, $\hat{U}$ represents the external field explicitly given by $\hat{U}(\tau) = \int d^3r\, \hat{n}(\mathbf{r})u(\mathbf{r}\tau)$, and $\hat{W}$ represents the two-particle interaction. We first define a functional of the external field u by

$$\tilde{A}[u] = i\ln\langle\Psi_0|V(\tau_f,\tau_i)|\Psi_0\rangle\,, \tag{3}$$

where V is the τ or contour ordered evolution operator of the system

$$V(\tau_f,\tau_i) = T_C \exp\Big[-i\int_{\tau_i}^{\tau_f} d\tau\, t'(\tau)\hat{H}(\tau)\Big], \tag{4}$$

where T_C denotes ordering in τ [12]. It is this redefinition of the time-ordering operator in addition to the introduction of the time contour which makes the Keldysh approach applicable in nonequilibrium Green function theory [12]. It is clear that if the external potential is equal on the forward and backward parts of the contour, i.e., of the form $u(\mathbf{r}\tau) = v(\mathbf{r}t(\tau))$, then this evolution operator will become unity and $\tilde{A}$ will become zero. Potentials of this type will be denoted as physical potentials. Functional derivatives, however, can be nonzero for physical potentials. The functional derivative of $\tilde{A}$ with respect to u yields

$$\frac{\delta\tilde{A}}{\delta u(\mathbf{r}\tau)} = \frac{\langle\Psi_0|V(\tau_f,\tau)\hat{n}(\mathbf{r})V(\tau,\tau_i)|\Psi_0\rangle}{\langle\Psi_0|V(\tau_f,\tau_i)|\Psi_0\rangle} = \langle\hat{n}_H(\mathbf{r}\tau)\rangle = n(\mathbf{r}\tau)\,, \tag{5}$$

where we defined the Heisenberg representation of an operator $\hat{O}$ as usual by $\hat{O}_H(\tau) = V(\tau_i,\tau)\hat{O}V(\tau,\tau_i)$ and the expectation value by

$$\langle\hat{O}_H(\tau)\rangle = \frac{\langle\Psi_0|T_C[V(\tau_f,\tau_i)\hat{O}_H(\tau)]\,|\Psi_0\rangle}{\langle\Psi_0|V(\tau_f,\tau_i)|\Psi_0\rangle}\,. \tag{6}$$

Note that we have used the usual convention of Keldysh Green function theory [12] where the functional derivative is defined by $\delta\tilde{A} = \int d^3r\, d\tau\, t'(\tau)\,[\delta\tilde{A}/\delta u(\mathbf{r}\tau)]\delta u(\mathbf{r}\tau)$; i.e., the term $t'(\tau)$ belongs to the integration measure rather than the functional derivative. If we now take the derivative of $\tilde{A}$ at a physical potential $u(\mathbf{r}\tau) = v(\mathbf{r}t(\tau))$ we obtain

$$\frac{\delta\tilde{A}}{\delta u(\mathbf{r}\tau)}\,\Big|_{u=v(\mathbf{r}t)} = \langle\Psi_0|V(t_0,t)\hat{n}(\mathbf{r})V(t,t_0)|\Psi_0\rangle = n(\mathbf{r}t)\,, \tag{7}$$

where the evolution operator V is now defined in physical time. Therefore, the derivative of $\tilde{A}$ at the physical potential v is the density of the system in the external field v. We now want to use $n(\mathbf{r}\tau)$ as our basic variable, and we perform a Legendre transform and define

$$A[n] = -\tilde{A}[u] + \int_C dt\, d^3r\, n(\mathbf{r}\tau)u(\mathbf{r}\tau) \tag{8}$$

so that $\delta A/\delta n(\mathbf{r}\tau) = u(\mathbf{r}\tau)$. For convenience we introduced the short notation $\int_C dt$ for $\int d\tau\, t'(\tau)$. The Legendre transformation assumes that there is a one-to-one relation between $u(\mathbf{r}\tau)$ and $n(\mathbf{r}\tau)$ such that Eq. (5) is invertible. This can, however, be proven by an extension of the Runge-Gross theorem to the case of a pseudotime parametrization [14].

We now define an action functional for a noninteracting system with the Hamiltonian

$$\hat{H}_s(\tau) = \hat{T} + \hat{U}_s(\tau) \tag{9}$$

and the action

$$\tilde{A}_s[u_s] = i\ln\langle\Phi_0|V_s(\tau_f,\tau_i)\,|\Phi_0\rangle\,. \tag{10}$$

The evolution operator $V_s(\tau_f,\tau_i)$ is defined similarly as in Eq. (4) with $\hat{H}$ replaced by $\hat{H}_s$. The initial wave function Φ_0 at $t = t_0$ is a Slater determinant. We can now do a similar Legendre transform and define

$$A_s[n] = -\tilde{A}_s[u_s] + \int_C dt\, d^3r\, n(\mathbf{r}\tau)u_s(\mathbf{r}\tau)\,. \tag{11}$$

The exchange-correlation part A_{xc} of the action functional is then defined by

$$A[n] = A_s[n] - A_{xc}[n] - \frac{1}{2}\int_C dt\, d^3r_1\, d^3r_2\, \frac{n(\mathbf{r}_1\tau)n(\mathbf{r}_2\tau)}{|\mathbf{r}_1 - \mathbf{r}_2|}\,. \tag{12}$$

The above equation implicitly assumes that the functionals A and A_s are defined on the same domain, i.e., that there exists a noninteracting system described by the Hamiltonian $\hat{H}_s$ with the same density as the interacting system described by the Hamiltonian $\hat{H}$. A necessary requirement in order for this to be true is that the initial states Ψ_0 and Φ_0 must yield the same density. For most applications, Ψ_0 will be the ground state of the system before the time-dependent field is switched on and Φ_0 will be the corresponding Kohn-Sham determinant of stationary density-functional theory. Functional differentiation of Eq. (12) with respect to $n(\mathbf{r}\tau)$ yields

$$u(\mathbf{r}\tau) = u_s(\mathbf{r}\tau) - u_{xc}(\mathbf{r}\tau) - u_H(\mathbf{r}\tau)\,, \tag{13}$$

where $u_H(\mathbf{r}\tau) = \int d^3r'\, n(\mathbf{r}'\tau)/|\mathbf{r} - \mathbf{r}'|$ is the Hartree potential and $u_{xc}(\mathbf{r}\tau) = \delta A_{xc}/\delta n(\mathbf{r}\tau)$ is the exchange-correlation potential. By the above construction the potential u_s of the noninteracting system yields the same density as the potential u in the fully interacting system. The noninteracting system is thus to be identified with the time-dependent Kohn-Sham system. If we take the above derivatives at the physical time-dependent density $n(\mathbf{r}t)$ corresponding to the potential $u(\mathbf{r}\tau) = v(\mathbf{r}t(\tau))$ of the interacting system, we can transform to physical time and the Kohn-Sham system is then given by the equations

$$[-\tfrac{1}{2}\nabla^2 + v(\mathbf{r}t) + v_H(\mathbf{r}t) + v_{xc}(\mathbf{r}t)]\phi_i(\mathbf{r}t) = i\partial_t\phi_i(\mathbf{r}t), \qquad v_{xc}(\mathbf{r}t) = \frac{\delta A_{xc}}{\delta n(\mathbf{r}\tau)}\bigg|_{n=n(\mathbf{r}t)}, \tag{14}$$

where the density $n(\mathbf{r}t)$ can be calculated from the sum of the square of the orbitals. We now address the problem of causality versus symmetry associated with the response functions. The second derivative of the functional $\tilde{A}$ yields

$$\chi(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \frac{\delta^2\tilde{A}}{\delta u(\mathbf{r}_1\tau_1)\delta u(\mathbf{r}_2\tau_2)} = -i\langle T_C \Delta\hat{n}_H(\mathbf{r}_1\tau_1)\Delta\hat{n}_H(\mathbf{r}_2\tau_2)\rangle, \tag{15}$$

where the density fluctuation operator $\Delta\hat{n}_H(\mathbf{r}\tau) = \hat{n}_H(\mathbf{r}\tau) - \langle\hat{n}_H(\mathbf{r}\tau)\rangle$ enters rather than the density operator, due to the derivatives of the denominator in Eq. (5). This density response function is symmetric as it should and from the Legendre transform it follows that its inverse is given by

$$\chi^{-1}(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \frac{\delta^2 A}{\delta n(\mathbf{r}_1\tau_1)\delta n(\mathbf{r}_2\tau_2)}. \tag{16}$$

Taking the second functional derivative of Eq. (12) now yields

$$\chi^{-1} = \chi_s^{-1} - \frac{1}{t'(\tau_1)}\frac{\delta(\tau_1 - \tau_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} - f_{xc}, \tag{17}$$

where χ_s^{-1} is the inverse of the Kohn-Sham density response function and $f_{xc}(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \delta v_{xc}(\mathbf{r}_1\tau_1)/\delta n(\mathbf{r}_2\tau_2)$. Since both χ^{-1} and χ_s^{-1} are symmetric also f_{xc} must be symmetric. However, these functions will become causal in physical time. In order to see how they act in physical time we calculate the density response $\delta n(\mathbf{r}t)$ due to a variation $\delta v(\mathbf{r}t)$. The function χ evaluated at a physical density $n(\mathbf{r}t)$ is given by

$$i\chi(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \theta(\tau_1 - \tau_2)\langle\Delta\hat{n}_H(\mathbf{r}_1t_1)\Delta\hat{n}_H(\mathbf{r}_2t_2)\rangle + (1 \leftrightarrow 2). \tag{18}$$

Hence, we have

$$\begin{aligned}\delta n(\mathbf{r}_1t_1) &= \int_C dt_2\, d^3r_2\, \chi(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)\delta v(\mathbf{r}_2t_2)\\ &= -i\int_{\tau_i}^{\tau_1} d\tau_2\, t'(\tau_2) d^3r_2\langle\hat{n}_H(\mathbf{r}_1t_1)\hat{n}_H(\mathbf{r}_2t_2)\rangle\delta v(\mathbf{r}_2t_2) - i\int_{\tau_1}^{\tau_f} d\tau_2\, t'(\tau_2) d^3r_2\langle\hat{n}_H(\mathbf{r}_2t_2)\hat{n}_H(\mathbf{r}_1t_1)\rangle\delta v(\mathbf{r}_2t_2)\\ &= \int_{t_0}^{+\infty} dt_2\, d^3r_2\, \chi_R(\mathbf{r}_1t_1, \mathbf{r}_2t_2)\delta v(\mathbf{r}_2t_2),\end{aligned} \tag{19}$$

where

$$i\chi_R(\mathbf{r}_1t_1, \mathbf{r}_2t_2) = \theta(t_1 - t_2) \times \langle\Psi_0|[\hat{n}_H(\mathbf{r}_1t_1), \hat{n}_H(\mathbf{r}_2t_2)]|\Psi_0\rangle. \tag{20}$$

In the last step we used the fact that the expectation value of the commutator of the density fluctuation operators is equal to the expectation value of the commutator of the density operators themselves. Similarly for χ_s we obtain $\chi_{s,R}$ which is given by Eq. (20) with Ψ_0 replaced by Φ_0. From Eq. (17) we see that f_{xc} has a similar structure as χ and χ_s. Transformation to physical time yields the causal equivalent $f_{xc,R}$. Acting in physical time, Eq. (17) then becomes

$$\chi_R^{-1} = \chi_{s,R}^{-1} - \frac{\delta(t_1 - t_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} - f_{xc,R}. \tag{21}$$

This is the basic equation used to calculate excitation energies within TDDFT [15,16]. We have thus obtained the main result of this paper. All response functions, i.e., higher order derivatives of the action functional are symmetric functions in pseudotime and become causal or retarded functions when transformed back to physical time. This resolves the paradox arising from the previous definition of the action functional.

Finally we will discuss a useful application of the new formalism, namely, a new derivation of the time-dependent optimized potential method (TDOPM) [17].

The exchange-correlation part A_{xc} of the action functional can be expanded in terms of Keldysh Green functions [18] where the perturbing Hamiltonian is given by $\hat{H} - \hat{H}_s$. The expansion of the logarithm of the evolution operator yields the set of closed connected diagrams. Perturbation theory also requires an adiabatic switching-on of $\hat{H} - \hat{H}_s$ in the physical time interval $(-\infty, t_0)$ in order to connect the states Ψ_0 and Φ_0. This is, however, readily achieved by extending the Keldysh contour to $-\infty$ [18]. If we restrict ourselves to the first order terms

we find that the Hartree term and the term with $u = u_s$ cancel and obtain the exchange-only expression

$$A_x[n] = -\frac{1}{2}\sum_{ij}^{N}\int_C dt\, d^3r_1\, d^3r_2 \times \frac{\phi_i^*(\mathbf{r}_1\tau)\phi_i(\mathbf{r}_2\tau)\phi_j(\mathbf{r}_1\tau)\phi_j^*(\mathbf{r}_2\tau)}{|\mathbf{r}_1 - \mathbf{r}_2|}. \quad (22)$$

One sees that this functional is an implicit functional of $n(\mathbf{r}\tau)$ but an explicit functional of the orbitals. Going to higher order in $\hat{H} - \hat{H}_s$, the Keldysh perturbation expansion, in a similar way, leads to orbital dependent expressions for the correlation part A_c of the action. In that case one may obtain u_{xc} from

$$u_{xc}(\mathbf{r}_2\tau_2) = \int_C dt\, d^3r_1 \frac{\delta A_{xc}}{\delta u_s(\mathbf{r}_1\tau_1)} \frac{\delta u_s(\mathbf{r}_1\tau_1)}{\delta n(\mathbf{r}_2\tau_2)}. \quad (23)$$

Matrix multiplication by χ_s and using the chain rule for differential yields

$$\int_C dt_2\, d^3r_2\, \chi_s(\mathbf{r}_1\tau_1,\mathbf{r}_2\tau_2)u_{xc}(\mathbf{r}_2\tau_2) = \sum_{i=1}^{N}\int_C dt_2\, d^3r_2 \frac{\delta A_{xc}}{\delta\phi_i(\mathbf{r}_2\tau_2)}\frac{\delta\phi_i(\mathbf{r}_2\tau_2)}{\delta u_s(\mathbf{r}_1\tau_1)} + \frac{\delta A_{xc}}{\delta\phi_i^*(\mathbf{r}_2\tau_2)}\frac{\delta\phi_i^*(\mathbf{r}_2\tau_2)}{\delta u_s(\mathbf{r}_1\tau_1)}. \quad (24)$$

In the following we will consider only the realistic case where the functional derivative $\delta A_{xc}/\delta\phi_i^*$ at a physical potential is the complex conjugate of $\delta A_{xc}/\delta\phi_i$. Calculating the functional derivatives $\delta\phi_i/\delta u_s$ and $\delta\phi_i^*/\delta u_s$ requires careful consideration of the boundary conditions. From Eq. (10) it follows that the state $|\Phi_0\rangle$ evolves from τ_i forward in pseudotime, and therefore the variations $\delta\phi_i$ have to satisfy the boundary condition $\delta\phi_i(\tau_i) = 0$. However, the complex conjugate state $\langle\Phi_0|$ evolves from τ_f backwards in pseudotime, and thus the variations $\delta\phi_i^*$ have to satisfy the boundary condition $\delta\phi_i^*(\tau_f) = 0$. Carrying out these variations in a similar way as in Ref. [17] we obtain from the pseudotime Kohn-Sham equations

$$\frac{\delta\phi_i(\mathbf{r}_2\tau_2)}{\delta u_s(\mathbf{r}_1\tau_1)} = -i\theta(\tau_2 - \tau_1)\phi_i(\mathbf{r}_1\tau_1) \times \sum_j \phi_j(\mathbf{r}_2\tau_2)\phi_j^*(\mathbf{r}_1\tau_1),$$
$$\frac{\delta\phi_i^*(\mathbf{r}_2\tau_2)}{\delta u_s(\mathbf{r}_1\tau_1)} = -i\theta(\tau_1 - \tau_2)\phi_i^*(\mathbf{r}_1\tau_1) \times \sum_j \phi_j^*(\mathbf{r}_2\tau_2)\phi_j(\mathbf{r}_1\tau_1). \quad (25)$$

Inserting the above expressions and transforming back to physical time yields the integral equation

$$\sum_j^N \int dt_2\, d^3r_2\, G_R(\mathbf{r}_1t_1,\mathbf{r}_2t_2)\phi_j(\mathbf{r}_1t_1)\phi_j^*(\mathbf{r}_2t_2) \times [v_{xc}(\mathbf{r}_2t_2) - w_{xcj}(\mathbf{r}_2t_2)] + \text{c.c.} = 0, \quad (26)$$

where we defined the retarded Green function by

$$iG_R(\mathbf{r}_1t_1,\mathbf{r}_2t_2) = \theta(t_1 - t_2)\sum_j \phi_j^*(\mathbf{r}_1t_1)\phi_j(\mathbf{r}_2t_2) \quad (27)$$

and the quantity w_{xcj} by

$$w_{xcj}(\mathbf{r}t) = \frac{1}{\phi_j^*(\mathbf{r}t)}\frac{\delta A_{xc}}{\delta\phi_j(\mathbf{r}\tau)}\bigg|_{\phi_i=\phi_i(\mathbf{r}t)}. \quad (28)$$

Equation (26) is the well known equation of the TDOPM [17].

Our results can be summarized as follows: We have resolved an existing paradox regarding the causality and symmetry properties of response functions within TDDFT. This is achieved by introducing an action functional defined on a Keldysh contour. From this action we furthermore derived the time-dependent Kohn-Sham equations and, as an example, the TDOPM equations.

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