Time-Dependent Density-Functional Calculation of Nuclear Response Functions

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Abstract

Basic issues of the time-dependent density-functional theory are discussed, especially a real-time calculation of linear response functions. Some remarks on the derivation of the time-dependent Kohn–Sham equations and on the numerical methods are given.

Keywords: *Time-dependent density-functional theory; time-dependent variational principle; strength function*

1 Introduction

The time-dependent density-functional theory (TDDFT) provides us with a practical tool to study quantum dynamics of many-body systems. It is conceptually very similar to the one known as the time-dependent Hartree–Fock (TDHF) theory with a density-dependent effective interaction in nuclear physics [1]. Although it is much more feasible than directly treating many-body wave functions of many-particle systems, the studies of full dynamics taking into account both the mean-field and the pairing correlations are still computationally highly challenging, even at present. In this paper, for simplicity, I will concentrate the discussion on the time-dependent Kohn–Sham (TDKS) equations, without the Bogoliubov-type extension including pair densities.

There is a number of recent developments in the studies of nuclei with the density functional approaches. For these, there are recent review papers [2–5]. Thus I do not intend here to review all these developments. Instead I would like to present some issues which are not well addressed in published articles. The first issue presented in Sec. 2, is a derivation of the time-dependent Kohn–Sham equations based on the time-dependent variational principle. Exactly the same argument is applicable to the variational derivation of the time-dependent Hartree–Fock equations. Especially, I would like to clarify that the gauge degrees of freedom naturally emerge in the proper derivation. This may be trivial to some readers, however, I think it is not so for non-practitioners. It may be also useful for students. Then, in Sec. 3, I will present some practical issues on numerical calculations such as the choice of the gauge functions, some speed-up techniques, etc.

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2 Remarks on the time-dependent Kohn–Sham equations

2.1 Time-dependent variational principle

It is well-known that the time-dependent Kohn–Sham equations can be obtained using the time-dependent variational principle [6]. In literature, we often find the following arguments: Starting from the action integral

$$S \equiv \int_{t_i}^{t_f} \left\{ \langle \Psi_D(t)| i\frac{\partial}{\partial t} |\Psi_D(t)\rangle - E[\rho(t)] \right\} dt \tag{1}$$

$$= \int_{t_i}^{t_f} \left\{ \sum_{i=1}^{N} \langle \psi_i(t)| i\frac{\partial}{\partial t} |\psi_i(t)\rangle - E[\rho(t)] \right\} dt, \tag{2}$$

the stationary action principle, $\delta S = 0$, leads to the time-dependent Kohn–Sham (TDKS) equations

$$i\frac{\partial}{\partial t}|\psi_i(t)\rangle = h[\rho(t)]|\psi_i(t)\rangle, \quad i = 1, \cdots, N. \tag{3}$$

Here, $E[\rho]$ is the energy density functional and $|\Psi_D(t)\rangle$ is the time-dependent Slater determinant,

$$|\Psi_D(t)\rangle = \frac{1}{\sqrt{N!}} \det\{|\psi_i(t)\rangle_j\}_{i,j=1,\cdots,N}. \tag{4}$$

The Kohn–Sham (single-particle) Hamiltonian is formally defined by

$$h[\rho]|\psi_i\rangle = \frac{\delta E}{\delta \langle \psi_i|}. \tag{5}$$

In the case of TDHF with an effective Hamiltonian, the energy density functional is given by the expectation value of the Hamiltonian as $E[\rho(t)] = \langle \Psi_D(t)|H|\Psi_D(t)\rangle$. In general, $E[\rho]$ can be a more general functional of one-body density ρ in the TDDFT.

Since the TDKS equations (3) are so common in literature, I think, many people take them for granted. However, it is somewhat strange that we have reached the equations which can uniquely determine the Kohn–Sham orbitals, because the Slater determinant $|\Psi_D(t)\rangle$ is invariant under a unitary transformation among the occupied orbitals. Namely, the same Slater determinant $|\Psi_D(t)\rangle$ can be expressed by different orbitals, $|\psi_i'(t)\rangle = \sum_{j=1}^{N} U_{ij}(t)|\psi_j(t)\rangle$, as Eq. (4), where $\{U_{ij}(t)\}$ is an arbitrary time-dependent unitary matrix. Thus, the Kohn–Sham orbitals have gauge degrees of freedom associated with the $U(N)$ transformation.

Apparently, the TDKS equation (3) uniquely determines the time evolution of each single-particle orbital $|\psi_i(t)\rangle$. Since we have not imposed any gauge fixing condition when we derived Eq. (3) from the stationary action principle, $\delta S = 0$, for Eq. (2), the Kohn–Sham (single-particle) orbitals should not be unique.

In fact, to my opinion, the derivation above is not satisfactory because we have used the orthonormal condition among the orbitals, $\langle \psi_i(t)|\psi_j(t)\rangle = \delta_{ij}$, to obtain Eq. (2) from Eq. (1). Therefore, the full variation with respect to each $\langle \psi_i(t)|$ should not be taken. I think that the proper derivation is either (i) the orthonormal relations are not assumed in the first place, or (ii) the variational space is restricted by the constraints $\langle \psi_i(t)|\psi_j(t)\rangle = \delta_{ij}$. In the following, I would like to present these proper derivations of the TDKS equations and to show that the gauge degrees of freedom appear naturally.

2.2 Derivation of TDKS equations (1)

To allow us to take full variation with respect to $\psi_i(t)$, we should not assume the orthonormal relation among $\{|\psi_i\rangle\}$. Let us derive the equations, starting from the action

$$S \equiv \int_{t_i}^{t_f} \left\{ \frac{\langle \Psi_D(t)|i\frac{\partial}{\partial t}|\Psi_D(t)\rangle}{\langle \Psi_D(t)|\Psi_D(t)\rangle} - E[\rho(t)] \right\} dt. \tag{6}$$

In order to perform calculation of the functional derivatives, some formulae, which are well-known in the generator coordinate method (GCM) [7], are very helpful. First, it is useful to define the overlap matrix,

$$B_{ij}(t) \equiv \langle \psi_i(t)|\psi_j(t)\rangle, \quad i,j = 1,\cdots,N, \tag{7}$$

which leads to the following expressions for the norm and the time derivative.

$$\langle \Psi_D(t)|\Psi_D(t)\rangle = \det B, \tag{8}$$

$$\langle \Psi_D(t)|i\frac{\partial}{\partial t}|\Psi_D(t)\rangle = \det B \sum_{ij} \langle \psi_i(t)|i\frac{\partial}{\partial t}|\psi_j(t)\rangle \left(B^{-1}\right)_{ji}. \tag{9}$$

Hereafter, $\sum_i$ means the summation with respect to the occupied (hole) orbitals, $i = 1,\cdots,N$, and the time-dependent overlap matrix $B(t)$ is simply denoted as B for simplicity. Using the cofactor expansion of the inverse matrix B^{-1}, we can prove that

$$\frac{\delta \left(B^{-1}\right)_{ij}(t')}{\delta\langle\psi_k(t)|} = -\left(B^{-1}\right)_{ik} \sum_l |\psi_l\rangle \left(B^{-1}\right)_{lj} \delta(t-t'). \tag{10}$$

In the same manner, the one-body density matrix can be written as

$$\rho(t) = \sum_{ij} |\psi_i(t)\rangle \left(B^{-1}\right)_{ij} \langle\psi_j(t)|. \tag{11}$$

Then, the derivative of $E[\rho]$ with respect to the bra state $\langle\psi_k(t)|$ becomes

$$\frac{\delta E[\rho]}{\delta\langle\psi_k(t)|} = h[\rho(t)] \sum_j |\psi_j(t)\rangle \left(B^{-1}\right)_{jk}. \tag{12}$$

Now it is easy to derive the TDKS equations

$$\left(1 - \sum_l |\psi_l(t)\rangle \left(B^{-1}\right)_{lj} \langle\psi_j(t)|\right) \left(i\frac{\partial}{\partial t} - h[\rho(t)]\right) \sum_k |\psi_k(t)\rangle \left(B^{-1}\right)_{ki} = 0. \tag{13}$$

This looks different from the well-known form of Eq. (3).

We may simplify Eq. (13) by assuming that the orbitals are orthonormal at a certain time t, $\langle\psi_i(t)|\psi_j(t)\rangle = \delta_{ij}$. In this case, we have $B_{ij}(t) = (B^{-1})_{ij} = \delta_{ij}$. Then, Eq. (13) can be written as

$$\left(1 - \sum_j |\psi_j(t)\rangle\langle\psi_j(t)|\right) \left(i\frac{\partial}{\partial t} - h[\rho(t)]\right) |\psi_i(t)\rangle = 0 \tag{14}$$

at time t. This means that the states

$$\left(i\frac{\partial}{\partial t} - h[\rho(t)]\right) |\psi_i(t)\rangle \tag{15}$$

do not contain the particle (unoccupied) orbitals at time t. In other words, they can be expanded in terms of the hole (occupied) orbitals only,

$$i\frac{\partial}{\partial t}|\psi_i(t)\rangle = h[\rho(t)]|\psi_i(t)\rangle + \sum_k \lambda_{ij}(t)|\psi_j(t)\rangle. \tag{16}$$

Although $\lambda_{ij}(t)$ are in principle arbitrary, choosing the Hermitian matrix $\lambda_{ik}(t)$ will conserve the orthonormal relation among the orbitals $\langle\psi_i(t+\Delta t)|\psi_j(t+\Delta t)\rangle = \delta_{ij}$. Therefore, provided that $\lambda_{ij}(t)$ are Hermitian, Eq. (16) can be true for any time t. They can be regarded as a general form of the TDKS equations. Here, the time-dependent Hermitian matrix $\lambda_{ij}(t)$ is a kind of gauge function for fixing the Kohn–Sham orbitals.

Equation (16) is also consistent with the well-known form of the equation for the one-body density matrix. Since the orthonormal relation is kept all the time, the density matrix of Eq. (11) can be simplified by assuming $B_{ij}^{-1} = \delta_{ij}$. Then, the time derivative of $\rho(t)$ can be calculated as

$$i\frac{\partial\rho}{\partial t} = \sum_i \left(h|\psi_i\rangle + \sum_j \lambda_{ij}|\psi_j\rangle\right)\langle\psi_i| - \sum_i |\psi_i\rangle\left(\langle\psi_i|h + \sum_j \langle\psi_j|\lambda_{ij}^*\right) \tag{17}$$

$$= \sum_i \left(h|\psi_i\rangle\langle\psi_i| - |\psi_i\rangle\langle\psi_i|h\right) \tag{18}$$

$$= [h[\rho(t)], \rho(t)]. \tag{19}$$

2.3 Derivation of TDKS equations (2)

We saw in the previous section that the calculation of the functional derivative of the action S in Eq. (6) is rather tedious. The use of Lagrange multipliers may greatly facilitate this calculation. One of the great advantages of the Lagrange multipliers is that, when we impose the constraints in terms of the Lagrange multipliers, we may simplify the functionals (functions) by using the constraints *before variations*. Now, we can use the action S in the simple form of Eq. (2) but with the Lagrange multipliers to impose the constraints of the orthonormal relation $\langle\psi_i(t)|\psi_j(t)\rangle = \delta_{ij}$,

$$\delta\left\{S - \sum_{ij}\lambda_{ij}(t)\left(\langle\psi_i(t)|\psi_j(t)\rangle - \delta_{ij}\right)\right\} = 0. \tag{20}$$

The variation immediately leads to Eq. (16). The form of Eq. (16) with the Hermitian matrix $\lambda(t)$ can be regarded as a general form of the TDKS equations.

Before ending this section, let us show that we can use the constraint conditions to simplify the functions before variation when the Lagrange multipliers are utilized. We consider here a problem of finding extrema of a function $F(\vec{x})$ with a constraint $g(\vec{x}) = 0$. Using the Lagrange multiplier λ, it can be given by the following variational form:

$$\delta\left\{F(\vec{x}) - \lambda g(\vec{x})\right\} = 0 \quad \rightarrow \quad \nabla F(\vec{x}) - \lambda\nabla g(\vec{x}) = 0 \quad \text{with } g(\vec{x}) = 0. \tag{21}$$

Namely, $\nabla F(\vec{x})$ is parallel to $\nabla g(\vec{x})$ which is the condition of the extrema under the constraint of $g(\vec{x}) = 0$. Now let us assume that the functional form of $F(\vec{x})$ can be modified (simplified) into $\tilde{F}(\vec{x})$ if we use the constraint $g(\vec{x}) = 0$,

$$\tilde{F}(\vec{x}) = f(\vec{x}; g = 0), \tag{22}$$

where $f(\vec{x}; g(\vec{x}))$ is a function of $\vec{x}$ and $g(\vec{x})$ satisfying $f(\vec{x}; g(\vec{x})) = F(\vec{x})$. From these, we can rewrite Eq. (21) as

$$\nabla\tilde{F}(\vec{x}) - \left(\lambda - \frac{\partial f}{\partial g}\right)\nabla g(\vec{x}) = 0 \quad \text{with } g(\vec{x}) = 0. \tag{23}$$

This means that $\nabla\tilde{F}(\vec{x})$ is also parallel to $\nabla g(\vec{x})$ at the extrema with $g(\vec{x}) = 0$. Therefore it is identical to the following variation for finding the extrema:

$$\delta\left\{\tilde{F}(\vec{x}) - \lambda g(\vec{x})\right\} = 0 \quad \rightarrow \quad \nabla\tilde{F}(\vec{x}) - \lambda\nabla g(\vec{x}) = 0 \quad \text{with } g(\vec{x}) = 0. \tag{24}$$

Thus we can replace $F(\vec{x})$ by $\tilde{F}(\vec{x})$ for the variational calculation with the Lagrange multiplier λ: $\tilde{F}(\vec{x}) = F(\vec{x})$ where $g(\vec{x}) = 0$ is satisfied. An extension of the present argument to the case of multiple constraints is straightforward.

3 Remarks on the numerical calculations

The Kohn–Sham orbitals are evolved in time according to Eq. (16). As is shown in the previous section, there are gauge degrees of freedom ($\lambda_{ij}(t)$) which we can choose arbitrarily. Although the choice of the gauge should not affect the physical quantities, the feasibility of numerical simulations sometimes depends on it.

3.1 Preparation of the initial state

In most applications, the initial state of the time evolution is prepared by solving the static Kohn–Sham equations:

$$h[\rho]|\psi_i\rangle = \epsilon_i|\psi_i\rangle, \quad \text{and} \quad \rho = \sum_i |\psi_i\rangle\langle\psi_i|. \tag{25}$$

Of course, this is not the only way of constructing the ground-state Kohn–Sham orbitals. Again, the $U(N)$ gauge degrees of freedom exist for the ground state. Nevertheless, they are somewhat special in the sense that both the Hamiltonian $h[\rho]$ and the density ρ are diagonal in these orbitals. They are often called "canonical orbitals".

To reach the ground state, the imaginary-time method is one of the most prevalent methods in nuclear physics [8]. We start from given initial wave functions for $|\psi_i^{(0)}\rangle$ which are orthonormalized to each other. Then, at the (n+1)-th iteration, the imaginary-time evolution of a small time step Δt is calculated as

$$|\psi_i^{(n+1)}\rangle = \exp(-\Delta t\ h[\rho^{(n)}])|\psi_i^{(n)}\rangle \approx \left(1 - \Delta t\ h[\rho^{(n)}]\right)|\psi_i^{(n)}\rangle, \tag{26}$$

where the Kohn–Sham Hamiltonian is constructed at the density of $\rho^{(n)}$ which is defined by

$$\rho^{(n)} = \sum_i |\psi_i^{(n)}\rangle\langle\psi_i^{(n)}|. \tag{27}$$

At each iteration, the Gram-Schmidt orthonormalization must be performed. This procedure converges to the solutions of Eq. (25) from the eigenstate of the lowest energy ϵ_1 to that of the N-th eigenvalue ϵ_N. You may also calculate the particle (unoccupied) states ($i > N$) if you want.

An advantage of the imaginary-time method is that it is a very stable iteration procedure to reach the convergence, though it may require a large number of iterations. Diagonalizing $h[\rho^{(n)}]$ in the space spanned by the set of states $\{|\psi_i^{(n)}\rangle\}_{i=1,\cdots,N}$ may speed up the convergence. Sometimes an additional damping factor associated with the kinetic energy terms, $1/p^2$, could help to lower the energy quickly, especially at the beginning stage of the iterations.

3.2 Strength functions in the linear response

The linear response in real time can be numerically realized if we slightly distort the ground-state density and start the time evolution. The distortion is made by a weak

external field, $V_{\rm ext}(t)$. The time profile of the external field determines the frequency range contained in $V_{\rm ext}(t)$,

$$V_{\rm ext}(t) = \frac{1}{2\pi}\int \tilde{V}_{\rm ext}(\omega)e^{-i\omega t}d\omega. \tag{28}$$

One of the popular choices is the instantaneous field, $V_{\rm ext}(t) \propto \delta(t)$, which correspond to the constant field in the frequency domain, $\tilde{V}_{\rm ext}(\omega) \sim V_0$. An advantage of this instantaneous external field is that the calculation of a single time evolution provides information on the whole frequency (energy) range.

The strength functions can be calculated within the real-time method as follows. Suppose we would like to calculate the strength function associated with the one-body Hermitian operator F for a system whose energy eigenstates are denoted by $|\Phi_n\rangle$. The initial state is constructed by applying the instantaneous external field $V_{\rm ext}(t) = -\eta F\delta(t)$ at $t = 0$, which leads to $|\Psi(t = 0+)\rangle = e^{i\eta F}|\Psi_0\rangle$. Here we adopt a small parameter η to perform numerically the linear approximation. The time-dependent state $|\Psi(t)\rangle$ can be decomposed in terms of $|\Phi_n\rangle$ as

$$|\Psi(t)\rangle = e^{-iHt}e^{i\eta F}|\Psi_0\rangle = e^{-iE_0t}|\Phi_0\rangle + i\eta\sum_n e^{-iE_nt}|\Phi_n\rangle\langle\Phi_n|F|\Phi_0\rangle + \mathcal{O}(\eta^2). \tag{29}$$

Therefore, the calculation of the expectation value of F leads to

$$\langle\Psi(t)|F|\Psi(t)\rangle = \langle\Phi_0|F|\Phi_0\rangle + 2\eta\sum_n |\langle\Phi_n|F|\Phi_0\rangle|^2 \sin\{(E_n - E_0)t\}. \tag{30}$$

Then the strength function is obtained by the Fourier transform:

$$\begin{aligned} S(E;F) &\equiv \sum_n |\langle\Phi_n|F|\Phi_0\rangle|^2\delta(E - (E_n - E_0)) \\ &= \frac{1}{\pi\eta}\int_0^\infty \sin(Et)\{\langle\Psi(t)|F|\Psi(t)\rangle - \langle\Phi_0|F|\Phi_0\rangle\}. \end{aligned} \tag{31}$$

In practice it is impossible to perform the time evolution up to $t = \infty$. Usually we introduce an artificial damping (smearing) factor γ to multiply the integrand of Eq. (31) by $e^{-\gamma t/2}$, and stop integration at $t = T$. The magnitude of the damping factor γ is related to the time duration T. To obtain a smooth curve as a function of energy E, we need to have $\gamma \gtrsim 2\pi/T$.

3.3 Choice of the gauge functions

Canonical orbitals of the ground state defined by Eq. (25) should correspond to stationary solutions of the TDKS equations (16). However, apparently, off-diagonal parts of the gauge functions $\lambda_{ij}(t)$ make the solution non-stationary since a mixing among the hole orbitals takes place in time. When we choose the gauge $\lambda_{ij}(t) = -\epsilon_i\delta_{ij}$, the static canonical orbitals of Eq. (25) become stationary, $\partial\psi_i/\partial t = 0$.

In the real-time calculation of the linear response, the state stays very close to the ground state, only a small part of the Kohn–Sham wave functions is fluctuating. Therefore it is convenient to adopt the same gauge as above, $\lambda_{ij}(t) = -\epsilon_i\delta_{ij}$. Of course, the choice of the gauge is completely arbitrary and should not affect the final results. However this choice has some numerical advantage because the time-dependent phase change of each Kohn–Sham orbital is minimized.

For calculation of nuclear dynamics beyond the linear regime such as a simulation of heavy-ion collisions, the choice of $\lambda_{ij}(t) = -\epsilon_i\delta_{ij}$ is no longer advantageous. Instead, we may adopt $\lambda_{ij}(t) = -\delta_{ij}\langle\psi_i(t)|h[\rho(t)]|\psi_i(t)\rangle$, for instance.

3.4 Numerical applications

In this article we do not show results of numerical calculations. I would like readers to refer to our previous papers [3, 9–25].

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