

Generalized Floquet formulation of time-dependent current-density-functional theory

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(Received 6 July 1998; revised manuscript received 27 August 1998)

We present a generalized Floquet formulation of time-dependent current-density-functional theory for non-perturbative treatment of multiphoton processes of many-electron systems in intense monochromatic electric and magnetic fields. It is shown that the time-dependent Kohn-Sham-like equation can be transformed into an equivalent *time-independent* Floquet Hamiltonian matrix eigenvalue problem. A procedure is outlined for the determination of the dynamical properties in bound-bound multiphoton transitions. For the case of bound-free transitions, such as the multiphoton ionization processes, we introduce the notion of complex density and complex current and present a non-Hermitian Floquet formalism for the calculation of the complex quasienergies of individual spin orbitals and total many-electron systems. [S1050-2947(98)04912-9]

PACS number(s): 32.80.Rm, 31.15.Ew, 31.10.+z, 71.15.Mb

I. INTRODUCTION

Density-functional theory (DFT) for stationary properties of many-electron systems, based on the earlier fundamental works of Hohenberg and Kohn [1] and Kohn and Sham [2], is now a well-established and practical tool in various branches of chemistry and physics [3–7]. It is a formalism of many-body theory in terms of the electron density $\rho(\mathbf{r})$. DFT proves to be accurate and computationally much less expensive than the *ab initio* wave functional methods and this accounts for its great success in the time-independent electron-structure calculations of the ground states of many-electron systems.

For problems where the magnetic properties are of interest, the conventional DFT is not sufficient, and it is necessary to use the current-density-functional theory (CDFT) [8,9]. In the CDFT of Vignale, Rasolt, and Geldart [8,9], the electron current density is included as an additional variable and the energy is minimized with respect to variations in the paramagnetic current as well as in the density. The CDFT has been recently implemented by Colwell and Handy [10] and applied to the calculation of magnetizabilities [11] and nuclear shielding tensors [12]. However, the CDFT still lacks reliable expressions for the exchange-correlation (xc) energy functional in the presence of uniform magnetic field, particularly for the case of strong magnetic fields. A new approach for the construction of spin-density xc energy functionals from CDFT [13] may be of interest here.

To study the more interesting dynamical properties, one needs a time-dependent DFT (TDDFT) [14–17]. Runge and Gross [17] have developed a time-dependent Kohn-Sham theory by considering the action to be stationary with respect to the density. Several groups have also considered a time-dependent current density functional (TDCDFT) recently [18–20]. In particular, using a classical, nonrelativistic treatment of the electromagnetic fields, Colwell *et al.* have recently presented a TDCDFT [20], based on the extension of

the CDFT of Vignale and Rasolt [8], where the action is stationary with respect to variations in paramagnetic current density as well as the density itself. The central result of the modern TDDFT and TDCDFT is a set of time-dependent Kohn-Sham (TDKS) equations which are structurally similar to the time-dependent Hartree-Fock (TDHF) equations but include in principle exactly all many-body effects through a local time-dependent xc potential. To date, with the exception of several of the most recent works on TDDFT [21–23] in intense laser fields, all the applications of TDDFT [24–26] and TDCDFT [20] fall in the regime of linear or nonlinear response in *weak* fields for which the *perturbation theory* is applicable.

In this paper we present a Floquet formulation of TD-CDFT for *nonperturbative* treatment of multiphoton processes in intense electric and magnetic fields. This is an extension of our recent works on the Floquet formulation of TDDFT in intense laser fields [23]. The Floquet theory and its various generalized formalisms [27] have been previously developed and applied to the nonperturbative studies of a number of atomic and molecular multiphoton and nonlinear optical processes in intense laser fields. However, previous Floquet applications are largely focused on the studies of one- or two-electron systems in intense one-color or multi-color laser fields [27]. *Ab initio* wave function approaches for the time-dependent Schrödinger equation beyond two-electron systems are computationally formidable within the current supercomputer technology. The motivation of this and previous [23] works is to develop new Floquet formulations in the context of the TDDFT and TDCDFT, allowing the extension to the nonperturbative treatment of multiphoton dynamics and nonlinear optical processes of *many-electron systems*. The Floquet formulation of TDCDFT is particularly relevant to processes where the magnetic field plays a significant role. Examples are multiphoton processes in the presence of both laser and static magnetic fields, processes involving open-shell atoms or molecules, and multiphoton processes in the presence of *superintense* laser fields (where both time-dependent electric and magnetic fields make important contributions), etc., to mention only a few. In this paper we show that the time-dependent problems in TDCDFT can be exactly transformed into equivalent *time-*

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independent Floquet matrix eigenvalue problems. Procedures are outlined for the determination of the dynamical properties in bound-bound multiphoton transitions. For the case of bound-free processes (such as multiphoton ionization), we introduce the notion of *complex density* and *complex current* and present a *non-Hermitian* Floquet formulation of TDCDFT for the calculation of complex quasienergies. Procedures are outlined for the analytical continuation of the density and current density onto the complex energy plane.

II. GENERALIZED FLOQUET FORMULATION OF TIME-DEPENDENT CURRENT-DENSITY-FUNCTIONAL THEORY

The wave function $\Psi(t)$ of a multielectron system subject to external time-dependent fields satisfies the following equation (atomic units are used throughout the paper):

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

Here $\hat{H}(t)$ is the Pauli Hamiltonian taking into account the external field coupling to the electron spins:

$$\begin{aligned} \hat{H} = & \frac{1}{2} \sum_{j=1}^N \left(-i\nabla - \frac{e}{c} \mathbf{A}(\mathbf{r}_j, t) \right)^2 + \sum_{i=1}^N u(\mathbf{r}_i) + e \sum_{i=1}^N \varphi(\mathbf{r}_i, t) \\ & - \frac{e}{c} \sum_{i=1}^N [\mathbf{B}(\mathbf{r}_i, t) \cdot \hat{\mathbf{s}}_i] + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|}. \end{aligned} \quad (2)$$

The notations in Eq. (2) are as follows: c is the velocity of light, and e is the electron charge which is equal to -1 in atomic units; the vector operator $\hat{\mathbf{s}}_i$ is the spin operator of the i th electron; the potential $u(\mathbf{r}_i)$ describes the Coulomb interaction of the i th electron with the nucleus; $\varphi(\mathbf{r}, t)$ and $\mathbf{A}(\mathbf{r}, t)$ are the scalar and vector potentials of the external field related to the electric and magnetic field strengths \mathbf{E} and \mathbf{B} :

$$\mathbf{E}(\mathbf{r}, t) = -\nabla\varphi(\mathbf{r}, t) - \frac{1}{c} \frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t}, \quad \mathbf{B}(\mathbf{r}, t) = \nabla \times \mathbf{A}(\mathbf{r}, t). \quad (3)$$

The last term on the right-hand side of Eq. (2) represents the classical electron-electron repulsion.

If the external fields are periodic in time, then the Hamiltonian $\hat{H}(t)$ is also periodic in time:

$$\hat{H}(t+T) = \hat{H}(t), \quad (4)$$

where T is the period. One can introduce also the fundamental frequency ω defined as

$$\omega = \frac{2\pi}{T}. \quad (5)$$

The Floquet theorem allows the wave function $\Psi(t)$ to be written in the form

$$\Psi(t) = \exp(-i\varepsilon t)\Phi(t), \quad (6)$$

where ε is the so-called *quasienergy* and $\Phi(t) = \Phi(t+T)$ is a periodic function of time. Equation (1) can be recast in the quasienergy eigenvalue equation [27]:

$$\left(\hat{H}(t) - i \frac{\partial}{\partial t} \right) \Phi(t) \equiv \hat{\mathcal{H}}\Phi(t) = \varepsilon \Phi(t), \quad (7)$$

which is the *steady-state* Hamiltonian eigenvalue problem in the *extended* Hilbert space. For the solution of Eq. (7), we can define a *quasienergy functional*

$$F[\Phi] = \langle\langle \Phi | \hat{\mathcal{H}} | \Phi \rangle\rangle, \quad (8)$$

where the inner product in the extended Hilbert space is defined as

$$\langle\langle \Phi | \Xi \rangle\rangle = \frac{1}{T} \int_0^T dt \langle \Phi | \Xi \rangle. \quad (9)$$

Variation of the functional (8) under the normalization condition

$$\langle\langle \Phi | \Phi \rangle\rangle = 1 \quad (10)$$

leads to Eq. (7) as an equation for the function Φ which brings the stationary value to the functional (8). Note that the normalization condition for the function Φ should be satisfied also in the ordinary configuration space:

$$\langle \Phi(t) | \Phi(t) \rangle = 1. \quad (11)$$

Equation (11) holds irrespectively of the time t although the function Φ depends on t . This important relation follows from the properties of the initial equation (1): for the self-adjoint Hamiltonian, the number of the electrons (or the normalization of the total wave function) is conserved.

Since Eq. (7) represents the *steady-state* problem, it can be rigorously justified that $\hat{\mathcal{H}}(t)$, $\hat{H}(t)$, $\Phi(t)$, and the quasienergy ε are all unique functionals of the electron spin densities and current spin densities (see, e.g., [15] with respect to TDDFT; the generalization to TDCDFT is straightforward). Thus the quasienergy functional Eq. (8) can be expressed as a functional of the spin densities and current spin densities. As $\Phi(t)$ in Eq. (7) is periodic in time, one can introduce the time-periodic Kohn-Sham spin orbitals $\phi_k^\sigma(\mathbf{r}, t)\sigma(s)$ (with σ and s being the spin function and the spin coordinate, respectively; the notation σ stands for both possible spin functions; we shall use the notations α and β when we need to distinguish different spin projections). The spatial parts of the spin orbitals are orthonormal to each other according to

$$\langle \phi_k^\sigma(\mathbf{r}, t) | \phi_i^\sigma(\mathbf{r}, t) \rangle = \delta_{ki}. \quad (12)$$

The electron spin densities $\rho^\sigma(\mathbf{r}, t)$ can be written as follows:

$$\rho^\sigma(\mathbf{r}, t) = \sum_k |\phi_k^\sigma(\mathbf{r}, t)|^2, \quad (13)$$

and the total density $\rho(\mathbf{r}, t)$ is a sum of the spin densities $\rho^\sigma(\mathbf{r}, t)$:

$$\rho(\mathbf{r}, t) = \sum_{\sigma} \rho^{\sigma}(\mathbf{r}, t). \quad (14)$$

The *paramagnetic* current spin densities $\mathbf{j}_p^{\sigma}(\mathbf{r}, t)$ are defined according to

$$\mathbf{j}_p^{\sigma} = \frac{1}{2i} \sum_k \{ [\phi_k^{\sigma}(\mathbf{r}, t)]^* \nabla \phi_k^{\sigma}(\mathbf{r}, t) - \phi_k^{\sigma}(\mathbf{r}, t) [\nabla \phi_k^{\sigma}(\mathbf{r}, t)]^* \}. \quad (15)$$

The summation with respect to k in Eqs. (13), (15) is performed over all occupied spatial orbitals; for the closed-shell atoms the number of spatial orbitals is equal to $N/2$, N being the total number of electrons.

The quasienergy functional (8) can be rewritten in the following form:

$$\begin{aligned} F[\Phi[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]] &\equiv F[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}] = T_s[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}] \\ &+ J[\rho] + E_{\text{xc}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}] \\ &+ E_{\text{ext}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]. \end{aligned} \quad (16)$$

Here $T_s[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]$ is the *modified* kinetic energy functional for the noninteracting reference system represented by the determinant of the Kohn-Sham spin orbitals:

$$\begin{aligned} T_s[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}] \\ = \sum_{k, \sigma} \frac{1}{T} \int_0^T dt \left\langle \phi_k^{\sigma}(\mathbf{r}, t) \left| -\frac{1}{2} \nabla^2 - i \frac{\partial}{\partial t} \right| \phi_k^{\sigma}(\mathbf{r}, t) \right\rangle, \end{aligned} \quad (17)$$

and the functional $J[\rho]$ represents the classical electron-electron repulsion:

$$J[\rho] = \frac{1}{2T} \int_0^T dt \int d^3 r_1 \int d^3 r_2 \frac{\rho(\mathbf{r}_1, t) \rho(\mathbf{r}_2, t)}{|\mathbf{r}_1 - \mathbf{r}_2|}. \quad (18)$$

The exchange-correlation functional $E_{\text{xc}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]$ contains the difference between the exact kinetic energy and $T_s[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]$ as well as the nonclassical part of the electron-electron interaction. It should be stressed that within CDFT and TDCDFT, the exchange-correlation energy is a functional of the spin densities as well as the current spin densities (see below). Finally, $E_{\text{ext}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]$ is the part of the quasienergy functional due to the external potentials. The present consideration assumes that the external magnetic field has a constant direction in space (along the quantization axis for the spins). In this case we can account for the spin coupling to the external magnetic field through the spin-dependent *scalar* potential [8]. Let us introduce the spin-dependent scalar potential $v^{\sigma}(\mathbf{r}, t)$ as

$$\frac{1}{2} [v^{\alpha}(\mathbf{r}, t) - v^{\beta}(\mathbf{r}, t)] = -\frac{e}{2c} B(\mathbf{r}, t), \quad (19)$$

$$\frac{1}{2} [v^{\alpha}(\mathbf{r}, t) + v^{\beta}(\mathbf{r}, t)] = e\varphi(\mathbf{r}, t).$$

Then $E_{\text{ext}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]$ can be expressed as follows:

$$\begin{aligned} E_{\text{ext}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}] \\ = \sum_{\sigma} \frac{1}{T} \int_0^T dt \int d^3 r \\ \times \left(u(\mathbf{r}) + v^{\sigma}(\mathbf{r}, t) + \frac{e^2}{2c^2} A^2(\mathbf{r}, t) \right) \rho^{\sigma}(\mathbf{r}, t) \\ + i \frac{e}{2c} \sum_{k, \sigma} \frac{1}{T} \int_0^T dt \langle \phi_k^{\sigma}(\mathbf{r}, t) | (\mathbf{A} \cdot \nabla) \\ + (\nabla \cdot \mathbf{A}) | \phi_k^{\sigma}(\mathbf{r}, t) \rangle. \end{aligned} \quad (20)$$

A more general theory which allows magnetic fields with variable directions is required to employ spin-density and spin-current matrices [8].

The Kohn-Sham equations for the time-periodic orbitals $\phi_k^{\sigma}(\mathbf{r}, t)$ are obtained from the stationary principle for the quasienergy functional (16) under the constraints (12). Using the functional differentiation of Eqs. (16) and (12) with respect to the orbitals $\phi_k^{\sigma}(\mathbf{r}, t)$ one arrives at the following set of time-dependent Kohn-Sham equations:

$$\begin{aligned} i \frac{\partial}{\partial t} \phi_k^{\sigma}(\mathbf{r}, t) &= \left[-\frac{1}{2} \nabla^2 + u(\mathbf{r}) + v^{\sigma}(\mathbf{r}, t) + v_s^{\sigma}(\mathbf{r}, t) \right. \\ &+ \frac{e^2}{2c^2} A^2(\mathbf{r}, t) + i \frac{e}{2c} [(\mathbf{A} \cdot \nabla) + (\nabla \cdot \mathbf{A})] \\ &\left. + i \frac{e}{2c} [(\mathbf{A}_{\text{xc}}^{\sigma} \cdot \nabla) + (\nabla \cdot \mathbf{A}_{\text{xc}}^{\sigma})] - \epsilon_k^{\sigma} \right] \phi_k^{\sigma}(\mathbf{r}, t), \end{aligned} \quad (21)$$

where the exchange-correlation vector potential $\mathbf{A}_{\text{xc}}^{\sigma}(\mathbf{r}, t)$ is defined as a functional derivative of E_{xc} with respect to the paramagnetic current \mathbf{j}_p^{σ} [that means, the projections of $\mathbf{A}_{\text{xc}}^{\sigma}(\mathbf{r}, t)$ are the functional derivatives of E_{xc} with respect to the corresponding projections of \mathbf{j}_p^{σ}]:

$$\frac{e}{c} \mathbf{A}_{\text{xc}}^{\sigma}(\mathbf{r}, t) = -\frac{\delta E_{\text{xc}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]}{\delta \mathbf{j}_p^{\sigma}}. \quad (22)$$

The single-particle potential $v_s^{\sigma}(\mathbf{r}, t)$ includes the classical electron-electron repulsion as well as the exchange-correlation scalar potential $v_{\text{xc}}^{\sigma}(\mathbf{r}, t)$:

$$v_s^{\sigma}(\mathbf{r}, t) = \int d^3 r' \frac{\rho(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} + v_{\text{xc}}^{\sigma}(\mathbf{r}, t), \quad (23)$$

$$v_{\text{xc}}^{\sigma}(\mathbf{r}, t) = \frac{\delta E_{\text{xc}}[\rho^{\alpha}, \rho^{\beta}, \mathbf{j}_p^{\alpha}, \mathbf{j}_p^{\beta}]}{\delta \rho^{\sigma}}. \quad (24)$$

The Lagrange multipliers ϵ_k^{σ} play the role of *orbital* quasienergies. The set of equations (21) is solved self-consistently producing the Kohn-Sham orbitals $\phi_k^{\sigma}(\mathbf{r}, t)$ and orbital quasienergies ϵ_k^{σ} . Then the total quasienergy ε of the N -

electron system can be determined according to Eq. (16) as a stationary point of the quasienergy functional:

$$\begin{aligned} \varepsilon = & \sum_{k,\sigma} \epsilon_k^\sigma - J[\rho] + E_{\text{xc}}[\rho^\alpha, \rho^\beta, \mathbf{j}_p^\alpha, \mathbf{j}_p^\beta] \\ & - \sum_{\sigma} \frac{1}{T} \int_0^T dt \int d^3r v_{\text{xc}}^\sigma(\mathbf{r}, t) \rho^\sigma(\mathbf{r}, t) \\ & - i \frac{e}{2c} \sum_{k,\sigma} \frac{1}{T} \int_0^T dt \langle \phi_k^\sigma(\mathbf{r}, t) | (\mathbf{A}_{\text{xc}}^\sigma \cdot \nabla) \\ & + (\nabla \cdot \mathbf{A}_{\text{xc}}^\sigma) | \phi_k^\sigma(\mathbf{r}, t) \rangle. \end{aligned} \quad (25)$$

The expressions (21) and (25) are the main equations of the present Floquet formulation of TDCDFT.

Since the paramagnetic currents \mathbf{j}_p^σ are gauge dependent [the physical current $\mathbf{j}^\sigma = \mathbf{j}_p^\sigma - (e/c)\rho^\sigma \mathbf{A}$ is gauge invariant], it may seem that the total quasienergy functional (16) also depends on the gauge. However, it can be shown [8] that the exchange-correlation functional actually depends on the vorticity

$$\mathbf{v}^\sigma = \nabla \times \frac{\mathbf{j}_p^\sigma}{\rho^\sigma}, \quad (26)$$

which is gauge invariant. Specific forms of this functional are available within the adiabatic local density approximation (ALDA) [9]. For example, the following simple form is applied in the case of sufficiently weak magnetic fields:

$$\begin{aligned} E_{\text{xc}}[\rho^\alpha, \rho^\beta, \mathbf{j}_p^\alpha, \mathbf{j}_p^\beta] = & E_{\text{xc}}[\rho^\alpha, \rho^\beta, 0, 0] \\ & + \sum_{\sigma} \frac{1}{T} \int_0^T dt \int d^3r g^\sigma(\rho^\sigma(\mathbf{r}, t)) |\mathbf{v}^\sigma|^2, \end{aligned} \quad (27)$$

where g^σ are some known functions depending locally on the corresponding spin densities [8,9,20]. Other approximate forms of the exchange-correlation functionals within TDCDFT and ALDA can be obtained by the approach of Ref. [13]. A recent treatment of TDCDFT beyond ALDA leads to the appearance of viscoelastic stresses in the electron fluid [28].

If we redefine the exchange-correlation functional as a functional of ρ^σ and \mathbf{v}^σ :

$$E_{\text{xc}}[\rho^\alpha, \rho^\beta, \mathbf{j}_p^\alpha, \mathbf{j}_p^\beta] \equiv \bar{E}_{\text{xc}}[\rho^\alpha, \rho^\beta, \mathbf{v}^\alpha, \mathbf{v}^\beta], \quad (28)$$

then the expressions for the exchange-correlation scalar and vector potentials are written as [8,20]

$$v_{\text{xc}}^\sigma(\mathbf{r}, t) = \frac{\delta \bar{E}_{\text{xc}}[\rho^\alpha, \rho^\beta, \mathbf{v}^\alpha, \mathbf{v}^\beta]}{\delta \rho^\sigma} - \frac{e}{c} \frac{1}{\rho^\sigma} (\mathbf{A}_{\text{xc}}^\sigma \cdot \mathbf{j}_p^\sigma), \quad (29)$$

$$\frac{e}{c} \mathbf{A}_{\text{xc}}^\sigma = - \frac{1}{\rho^\sigma} \nabla \times \frac{\delta \bar{E}_{\text{xc}}[\rho^\alpha, \rho^\beta, \mathbf{v}^\alpha, \mathbf{v}^\beta]}{\delta \mathbf{v}^\sigma}. \quad (30)$$

The solution of the set of TDKS equations (21) can be greatly facilitated by recasting it into a set of *time-*

independent matrix equations by means of the Fourier expansion of the periodic functions $\phi_k^\sigma(\mathbf{r}, t)$, $\rho^\sigma(\mathbf{r}, t)$, $v^\sigma(\mathbf{r}, t)$, $\mathbf{A}(\mathbf{r}, t)$, $\mathbf{A}_{\text{xc}}^\sigma(\mathbf{r}, t)$, and $v_s^\sigma(\mathbf{r}, t)$. For example, one can write the following expansion for the Kohn-Sham orbital $\phi_k^\sigma(\mathbf{r}, t)$:

$$\phi_k^\sigma(\mathbf{r}, t) = \sum_{m=-\infty}^{\infty} \exp(-im\omega t) \phi_{km}^\sigma(\mathbf{r}). \quad (31)$$

Equation (31) defines the Fourier components $\phi_{km}^\sigma(\mathbf{r})$ of the orbital $\phi_k^\sigma(\mathbf{r}, t)$. Similarly we can define the Fourier components of the other quantities mentioned above, namely, $\rho^\sigma(\mathbf{r}, t)$, $v^\sigma(\mathbf{r}, t)$, $\mathbf{A}(\mathbf{r}, t)$, $\mathbf{A}_{\text{xc}}^\sigma(\mathbf{r}, t)$, and $v_s^\sigma(\mathbf{r}, t)$, and denote their m th Fourier components with a subscript m . In this fashion, one can recast Eq. (21) in the following set of infinite-dimensional *time-independent* coupled differential equations:

$$\begin{aligned} \left(-\frac{1}{2} \nabla^2 + u(\mathbf{r}) - m\omega \right) \phi_{km}^\sigma(\mathbf{r}) + \sum_{n=-\infty}^{\infty} v_{m-n}^\sigma(\mathbf{r}) \\ + (v_s^\sigma)_{m-n}(\mathbf{r}) + \frac{e^2}{2c^2} \sum_{n'=-\infty}^{\infty} A_{m-n-n'} A_{n'} \\ + i \frac{e}{2c} \{ (\mathbf{A}_{m-n} \cdot \nabla) + (\nabla \cdot \mathbf{A}_{m-n}) + [(\mathbf{A}_{\text{xc}}^\sigma)_{m-n} \cdot \nabla] \\ + [\nabla \cdot (\mathbf{A}_{\text{xc}}^\sigma)_{m-n}] \} \phi_{kn}^\sigma(\mathbf{r}) = \epsilon_k^\sigma \phi_{km}^\sigma(\mathbf{r}). \end{aligned} \quad (32)$$

Equations (32) are the working equations of the present Floquet formulation of TDCDFT. They can be rewritten in the familiar form of the time-independent Floquet matrix eigenvalue problem [27]:

$$\hat{H}_F^\sigma(\mathbf{r}) \vec{\phi}_k^\sigma = \epsilon_k^\sigma \vec{\phi}_k^\sigma, \quad (33)$$

where \hat{H}_F^σ is the Floquet Hamiltonian defined by Eq. (32) and $\vec{\phi}_k^\sigma$ is the vector consisting of the components $\phi_{km}^\sigma(\mathbf{r})$. The orbital quasienergy eigenvalues ϵ_k^σ and eigenfunctions $\vec{\phi}_k^\sigma$ are to be solved self-consistently.

To calculate bound-bound transition probabilities under the influence of the external fields, let us consider a complete orthonormal set $\{\psi_k^\sigma\}$ of orbital eigenfunctions for the unperturbed field-free Kohn-Sham Hamiltonian. Then the basis in the extended Hilbert space is composed of the vectors $|k\sigma n\rangle \equiv |k\sigma\rangle \otimes |n\rangle = \psi_k^\sigma \exp(-in\omega t)$, where k and σ are, respectively, the electron orbital and spin index, and n is the Fourier index of the classical field. The transition probability from the initial state ψ_i^σ to the final state ψ_f^σ for the time interval $t-t_0$ averaged over the initial times t_0 is written as [27]

$$P_{i \rightarrow f}^\sigma(t-t_0) = \sum_n | \langle f\sigma n | \exp[-i\hat{H}_F^\sigma(t-t_0)] | i\sigma 0 \rangle |^2. \quad (34)$$

Given the time-dependent total electron density $\rho(\mathbf{r}, t)$, one can determine *nonperturbatively* various nonlinear optical properties of many-electron systems in strong laser fields, such as nonlinear optical susceptibilities, high-order harmonic generation rates, etc. For example, for the n th har-

monic of the fundamental frequency ω , the photon emission rate Γ_n^h (the number of photons of frequency $n\omega$ emitted per unit time) can be written as

$$\Gamma_n^h = \frac{4n^3\omega^3|\mathbf{d}_n|^2}{3c^3}, \quad (35)$$

where $|\mathbf{d}_n|^2$ is the squared absolute value of the dipole moment Fourier component calculated as follows:

$$\begin{aligned} |\mathbf{d}_n|^2 &= \left| \frac{1}{T} \int_0^T dt \int d^3\mathbf{r} \mathbf{r} \exp(in\omega t) \rho(\mathbf{r}, t) \right|^2 \\ &= \left| \int d^3\mathbf{r} \mathbf{r} \rho_n(\mathbf{r}) \right|^2. \end{aligned} \quad (36)$$

To conclude this section, we note that the Floquet formulation of TDCDFT presented here is applicable in principle to any bound-bound transitions of multielectron atomic or molecular systems in weak or strong fields. Since the Hamiltonian $\hat{H}(t)$ and the Floquet Hamiltonian \hat{H}_F^σ are Hermitian operators, the orbital and total quasienergies are real numbers. In Sec. III we describe the extension to include bound-free transitions, namely, ionization processes.

III. NON-HERMITIAN FLOQUET FORMULATION OF TIME-DEPENDENT CURRENT-DENSITY-FUNCTIONAL THEORY IN PERIODIC FIELDS

In the presence of intense external electromagnetic fields, atoms (molecules) can be ionized by the absorption of multiple photons, and all the bound states become *shifted* and *broadened* metastable resonance states possessing *complex quasienergies* $\varepsilon = \varepsilon_r - i\Gamma/2$. The real parts of the complex quasienergies, ε_r , provide the ac Stark shifted energy levels, while Γ are equal to the total ionization (dissociation) rates of the corresponding atomic (molecular) states. To determine these complex quasienergy states, the *non-Hermitian Floquet Hamiltonian formalisms* previously developed for atoms [29,30] and molecules [31,32], which employ the use of the complex-scaling transformation methods [33,34], can be extended to TDCDFT as described below. The use of the complex-scaling transformation, $\mathbf{r} \rightarrow \mathbf{r}e^{i\alpha}$, allows the analytical continuation of the Hermitian Floquet Hamiltonian $\hat{H}_F^\sigma(\mathbf{r})$, Eq. (33), into a *non-Hermitian* Floquet Hamiltonian $\hat{H}_F^\sigma(\mathbf{r}e^{i\alpha})$, reducing the problem of the determination of the complex quasienergy eigenvalues ε_k^σ and eigenvectors $\vec{\phi}_k^\sigma(\mathbf{r}e^{i\alpha})$ to the solution of a non-Hermitian eigenvalue problem. (Note that in the spherical coordinates, only the radial distance r is complex rotated.) In the non-Hermitian Floquet formulation of TDCDFT, all the quantities given in the quasienergy functional, Eq. (8), as well as the spin densities and current spin densities themselves become *complex* quantities.

A delicate task is to perform the analytical continuation of the exchange-correlation scalar and vector potentials in Eqs. (29) and (30), which depend on the spin densities ρ^σ and paramagnetic current spin densities \mathbf{j}_p^σ , to the complex plane. In the previous non-Hermitian Floquet formulation of

TDDFT [23], we used the well-known complex-scaling transformation (CST) for the analytical continuation of the Floquet Hamiltonian into the complex plane [27]. Under CST and the associated biorthogonality relationship, only the angular part of the wave function needs to be complex conjugated (for the bra vector) in the inner product or matrix element calculations. The radial part of the wave function needs *not* be complex conjugated in the corresponding bra vector. The implementation of CST procedure leads to the notion of *complex density* which is complex valued on the real axis of the radial coordinate. Physically the nonvanishing imaginary part of the complex density on the real r axis reflects the probability of decay of the quantum system, and a procedure for extracting the orbital (single photon or multiphoton) ionization rates was proposed [23]. The test case results of the photoionization of He atoms are in good agreement with the experimental data [23]. However, the analytical continuation procedure proposed in [23] for the spin density cannot be directly applied to the case of current density, since the latter requires the use of the complex-conjugated radial wave functions (as well as the angular wave functions). This requires further generalization of the analytical continuation procedure for both spin density and current density. In the following we introduce an alternative definition of the complex spin density:

$$\rho^\sigma(\mathbf{r}, t) = \sum_k [\phi_k^\sigma(\mathbf{r}^*, t)]^* \phi_k^\sigma(\mathbf{r}, t). \quad (37)$$

Equation (37) represents an explicitly analytically continuable quantity (the notation \mathbf{r}^* stands for the vector with the complex-conjugated radial coordinate r), and for real r reduces *exactly* to the conventional definition (13). That means the spin density defined by Eq. (37) is always real and non-negative on the real r axis, in contrast with the complex spin density defined in Ref. [23]. In the same manner we can define the complex current spin density:

$$\begin{aligned} \mathbf{j}_p^\sigma(\mathbf{r}, t) &= \frac{1}{2i} \sum_k \{ [\phi_k^\sigma(\mathbf{r}^*, t)]^* \nabla \phi_k^\sigma(\mathbf{r}, t) \\ &\quad - \phi_k^\sigma(\mathbf{r}, t) \nabla [\phi_k^\sigma(\mathbf{r}^*, t)]^* \}. \end{aligned} \quad (38)$$

For real r , Eq. (38) reduces to the conventional form (15). Using Eqs. (37) and (38) for analytical continuation of the spin densities and current spin densities, one can also analytically continue the vorticities $\boldsymbol{\nu}^\sigma$, Eq. (26), in the complex plane of the radial coordinate. The analytical continuation of the potentials in Eq. (21) is as follows. The potentials $u(\mathbf{r})$, $v^\sigma(\mathbf{r}, t)$, and $\mathbf{A}(\mathbf{r}, t)$ are explicit functions of r ; their calculation for complex r is straightforward. For the potentials $v_s^\sigma(\mathbf{r}, t)$ and $\mathbf{A}_{xc}^\sigma(\mathbf{r}, t)$, which are functionals of ρ^σ and $\boldsymbol{\nu}^\sigma$, Eqs. (37), (38), and (26) can be applied to obtain those quantities for complex r .

The actual computational procedure can be summarized as follows. First, the non-Hermitian Floquet eigenvalue problem (33) is solved in the complex r plane. We use the complex-scaling generalized pseudospectral technique [35,36] to discretize the Floquet Hamiltonian $\hat{H}_F^\sigma(\mathbf{r}e^{i\alpha})$ and solve for the complex-rotated Fourier components $\phi_{km}^\sigma(\mathbf{r}e^{i\alpha})$ of the Kohn-Sham orbitals. Second, the Fourier components

$\phi_{km}^\sigma(\mathbf{r}e^{i\alpha})$ are rotated back to the real axis with the help of the back rotation procedure developed in [37] yielding $\phi_{km}^\sigma(\mathbf{r})$. Third, the spin densities ρ^σ and current spin densities \mathbf{j}_p^σ are calculated according to the conventional definitions (13) and (15) for the real radial coordinate r . Then the potentials $v_s^\sigma(\mathbf{r},t)$ in Eq. (23) and $\mathbf{A}_{xc}^\sigma(\mathbf{r},t)$ in Eq. (22) and their time Fourier components are calculated. Fourth, the Fourier components of the potentials are complex rotated to the original position in the complex r plane ($r \rightarrow re^{i\alpha}$) and substituted into Eq. (32). [This approach gives exactly the same result as the direct calculation of the complex-scaled potentials with the help of Eqs. (37) and (38).] Together with the Fourier components of the external fields $v^\sigma(\mathbf{r}e^{i\alpha},t)$ and $\mathbf{A}(\mathbf{r}e^{i\alpha},t)$ and electron-nucleus interaction $u(\mathbf{r}e^{i\alpha})$, they determine the non-Hermitian Floquet eigenvalue problem in Eq. (33). Since Eq. (33) involves a self-consistent field, an iterative process is required to solve it. The procedure described above should be applied on each iteration step until convergence is achieved. On the first iteration step, the required quantities ρ^σ and \mathbf{j}_p^σ can be easily calculated for the initial guess orbitals are defined in the analytical form.

Now let us discuss the calculation of the ionization or multiphoton ionization rates. There is a well-known relation between the total ionization rate Γ and the imaginary part of the total quasienergy ε :

$$\Gamma = -2 \operatorname{Im} \varepsilon. \quad (39)$$

The total quasienergy can be calculated according to the functional form (25). Using the Fourier expansions of the orbitals, densities, and potentials, it can be expressed as follows:

$$\begin{aligned} \varepsilon = & \sum_{k,\sigma} \epsilon_k^\sigma + J[\rho] + E_{xc}[\rho^\alpha, \rho^\beta, \mathbf{j}_p^\alpha, \mathbf{j}_p^\beta] \\ & - \sum_\sigma \sum_m \int d^3r (v_s^\sigma)_m(\mathbf{r}) \rho_{-m}^\sigma(\mathbf{r}) \\ & - i \frac{e}{2c} \sum_{k,\sigma} \sum_{m,n} \langle \phi_{km}^\sigma(\mathbf{r}) | [(\mathbf{A}_{xc}^\sigma)_{m-n} \cdot \nabla] \\ & + [\nabla \cdot (\mathbf{A}_{xc}^\sigma)_{m-n}] | \phi_{kn}^\sigma(\mathbf{r}) \rangle. \end{aligned} \quad (40)$$

All the terms in the right-hand side of Eq. (40) are real except the sum of the orbital quasienergies ϵ_k^σ . One can see it from the equivalent equation (25) if the spatial integrations are performed on the real axis of the radial coordinate r . Thus one arrives at the following result:

$$\operatorname{Im} \varepsilon = \sum_{k,\sigma} \operatorname{Im} \epsilon_k^\sigma. \quad (41)$$

In addition to the total (multiphoton) ionization rates of atoms in external electromagnetic fields, it is also important to determine the partial ionization rate from each individual electronic orbital. In the Appendix we show by means of the equation of continuity that the imaginary parts of the spin-orbital quasienergies ϵ_k^σ have the usual physical meaning, namely,

TABLE I. Total ionization rates Γ for the ground state of the He atom as calculated using the present procedure and that of Ref. [23]. Numbers in brackets indicate the power of 10. ω and F are, respectively, the field frequency and amplitude, and n is the number of photons absorbed to ionize the atom.

n	ω (a.u.)	F (a.u.)	Γ (a.u.)	
			Present work	Ref. [23]
1	1.40	1.0[-3]	5.298[-7]	5.298[-7]
	1.40	1.0[-2]	5.300[-5]	5.298[-5]
	1.00	1.0[-3]	1.293[-6]	1.293[-6]
	1.00	1.0[-2]	1.299[-4]	1.293[-4]
2	0.60	2.0[-2]	5.972[-7]	5.972[-7]
	0.60	5.0[-2]	2.320[-5]	2.320[-5]
	0.50	2.0[-2]	1.156[-6]	1.156[-6]
	0.50	5.0[-2]	4.444[-5]	4.452[-5]
3	0.40	2.0[-2]	6.126[-9]	6.124[-9]
	0.40	5.0[-2]	2.588[-6]	2.578[-6]
	0.35	2.0[-2]	1.022[-8]	1.022[-8]
	0.35	5.0[-2]	2.234[-6]	2.236[-6]

$$\Gamma_k^\sigma = -2 \operatorname{Im} \epsilon_k^\sigma, \quad (42)$$

where Γ_k^σ is the ionization rate from the particular Kohn-Sham spin orbital with the indexes k and σ . Summing Eq. (42) over all occupied spin orbitals and taking into account Eqs. (39) and (41) one obtains

$$\Gamma = \sum_{k,\sigma} \Gamma_k^\sigma. \quad (43)$$

Thus the *total* ionization rate can be expressed as a sum of *spin orbital* ionization rates.

We note that the expression Eq. (42) is considerably simpler than the one we derived in the previous work on Floquet formulation of TDDFT [23]. In the latter work, the current spin densities were not involved, and we used a different procedure to analytically continue the spin densities $\rho^\sigma(\mathbf{r},t)$ to the complex plane. For the special case of the helium atom, it can be shown that there is no contribution from the current density to the exchange-correlation part of the total quasienergy functional. This provides an opportunity to test the analytical continuation procedure introduced here against the previous work. The calculations were performed for multiphoton ionization of He at several laser frequencies (ω) and laser (electric) field amplitudes (F), using the same quasienergy functional as in Ref. [23]. The field parameters used correspond to the two- and three-photon ionization regimes. The computed total ionization rates of the ground state of the helium atom are presented in Table I along with the results of Ref. [23]. One can see that while the analytical continuation procedures are different, the final results for the ionization rates are nearly identical.

The primary purpose of this paper is to present the theoretical formulation of TDCDFT in the Floquet framework. Extension of the Floquet-TDCDFT procedure to the study of

multiphoton and nonlinear optical processes of more complex atoms, in which the current density plays a significant role, is in progress.

ACKNOWLEDGMENTS

D.A.T. acknowledges the support of the Kansas Center for Advanced Scientific Computing sponsored by the K*STAR program.

APPENDIX: EQUATION OF CONTINUITY AND PARTIAL RATES

To calculate the partial ionization rate from each spin orbital, we need to study the equation of continuity on the *real* axis of the radial coordinate r . The spin density $\rho^\sigma(\mathbf{r}, t)$, as defined by Eq. (37), as well as the vector $\mathbf{A}_{xc}^\sigma(\mathbf{r}, t)$ and scalar $v_s^\sigma(\mathbf{r}, t)$ potentials are *real* quantities on the real axis of r . Starting from the TDKS equation (21) and the analogous equation for the complex-conjugated orbital, one readily arrives at the *equation of continuity* for the Kohn-Sham orbital $\phi_k^\sigma(\mathbf{r}, t)$:

$$\begin{aligned} \frac{\partial}{\partial t} |\phi_k^\sigma(\mathbf{r}, t)|^2 = & -[\nabla \cdot (\mathbf{j}_p)^\sigma] + \frac{e}{c} [\nabla \cdot (|\phi_k^\sigma|^2 \mathbf{A})] \\ & + \frac{e}{c} [\nabla \cdot (|\phi_k^\sigma|^2 \mathbf{A}_{xc}^\sigma)] - 2(\text{Im } \epsilon_k^\sigma) |\phi_k^\sigma|^2 \end{aligned} \quad (\text{A1})$$

$$= -(\nabla \cdot \mathbf{j}_k^\sigma) - 2(\text{Im } \epsilon_k^\sigma) |\phi_k^\sigma|^2, \quad (\text{A2})$$

where we have used the notations $(\mathbf{j}_p)^\sigma$ for the *paramagnetic* orbital current spin density:

$$\begin{aligned} (\mathbf{j}_p)^\sigma(\mathbf{r}, t) = & \frac{1}{2i} \{ [\phi_k^\sigma(\mathbf{r}, t)]^* \nabla \phi_k^\sigma(\mathbf{r}, t) \\ & - \phi_k^\sigma(\mathbf{r}, t) [\nabla \phi_k^\sigma(\mathbf{r}, t)]^* \}, \end{aligned} \quad (\text{A3})$$

and \mathbf{j}_k^σ for the *physical* orbital current spin density:

$$\mathbf{j}_k^\sigma(\mathbf{r}, t) = (\mathbf{j}_p)^\sigma(\mathbf{r}, t) - \frac{e}{c} (|\phi_k^\sigma|^2 \mathbf{A}) - \frac{e}{c} (|\phi_k^\sigma|^2 \mathbf{A}_{xc}^\sigma). \quad (\text{A4})$$

Note that the summation over all occupied orbitals leads to the following relation between the total physical and paramagnetic current spin densities:

$$\mathbf{j}^\sigma(\mathbf{r}, t) = \mathbf{j}_p^\sigma(\mathbf{r}, t) - \frac{e}{c} [\rho^\sigma(\mathbf{r}, t) \mathbf{A}(\mathbf{r}, t)], \quad (\text{A5})$$

since the exchange-correlation vector potential is subject to the constraint [8] [see Eq. (30)]

$$[\nabla \cdot (\rho^\sigma \mathbf{A}_{xc}^\sigma)] = 0. \quad (\text{A6})$$

The time-independent form of Eq. (A2) can be obtained by integration over the time period T . Since the function

$|\phi_k^\sigma(\mathbf{r}, t)|^2$ is time periodic, the left-hand side of Eq. (A2) vanishes after the time integration. This results in the following *differential* form of the equation of continuity:

$$\begin{aligned} & -2(\text{Im } \epsilon_k^\sigma) \frac{1}{T} \int_0^T dt |\phi_k^\sigma|^2 \\ & = [\nabla \cdot (\bar{\mathbf{j}}_p)^\sigma] - \frac{e}{c} \left[\frac{1}{T} \int_0^T dt [\nabla \cdot (|\phi_k^\sigma|^2 \mathbf{A})] \right. \\ & \quad \left. + \frac{1}{T} \int_0^T dt [\nabla \cdot (|\phi_k^\sigma|^2 \mathbf{A}_{xc}^\sigma)] \right] = (\nabla \cdot \bar{\mathbf{j}}_k)^\sigma, \end{aligned} \quad (\text{A7})$$

where the time-averaged orbital paramagnetic and physical current spin densities $(\bar{\mathbf{j}}_p)^\sigma(\mathbf{r})$ and $\bar{\mathbf{j}}_k^\sigma(\mathbf{r})$ are defined, respectively, as follows:

$$(\bar{\mathbf{j}}_p)^\sigma(\mathbf{r}) = \frac{1}{T} \int_0^T dt (\mathbf{j}_p)^\sigma(\mathbf{r}, t), \quad (\text{A8})$$

$$\bar{\mathbf{j}}_k^\sigma(\mathbf{r}) = \frac{1}{T} \int_0^T dt \mathbf{j}_k^\sigma(\mathbf{r}, t). \quad (\text{A9})$$

To obtain the *integral* form of the equation of continuity, one needs to perform the spatial integration of Eq. (A7). However, there is a problem regarding the calculation of integrals which involve the resonance wave functions. The complex-scaled ($r \rightarrow r e^{i\alpha}$) resonance wave functions are localized in the coordinate space, but they become delocalized upon the back rotation to the real axis of the radial coordinate. On the real r axis such functions diverge exponentially as $r \rightarrow \infty$. The present formulation of TDCDFT makes use of analytically continuable expressions for the density (37) and current density (38) which for real r reduce to the conventional definitions of those quantities (13) and (15), respectively. Thus the normalization integral for the density contains the squared *absolute values* of the Kohn-Sham orbitals. Such an integral diverges and cannot be regularized if the integration is performed in the infinite coordinate space. This problem does not arise if the Kohn-Sham equations are solved within a finite interval of r , $0 < r < r_{\max}$, with sufficiently large r_{\max} . For a wide range of r_{\max} , the eigenfunctions representing the true resonances are stable against variation of r_{\max} . Such eigenfunctions possess complex quasienergies and describe decay processes. On the other hand, the spin densities calculated with such Kohn-Sham orbitals can be normalized for the integration is performed within the finite interval of the radial coordinate. Note that a similar approach was used recently for construction of the Siegert pseudostates [38].

Performing the integration of Eq. (A7) within the range $0 < r < r_{\max}$ and taking into account the normalization condition (12), one obtains the (time-independent) integral form of the continuity equation:

$$-2(\text{Im } \epsilon_k^\sigma) = \Gamma_k^\sigma. \quad (\text{A10})$$

Here Γ_k^σ is the electron current through the sphere with the radius $r = r_{\max}$:

$$\Gamma_k^\sigma = \int d\hat{\mathbf{r}} [\hat{\mathbf{r}} \cdot \vec{\mathbf{J}}_k^\sigma(\mathbf{r})] |_{r=r_{\max}} \quad (\text{A11})$$

($\hat{\mathbf{r}}$ is the unit vector in the \mathbf{r} direction). As one can see, the current Γ_k^σ corresponding to the orbital $\phi_k^\sigma(\mathbf{r}, t)$ is equal to twice the absolute value of the imaginary part of the orbital quasienergy ϵ_k^σ .

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