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Excited-state populations of He exposed to intense laser fields by the multiconfiguration time-dependent Hartree-Fock method

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[Abstract] We investigate theoretically time-dependent populations of excited states of atoms exposed to intense laser light by the multiconfiguration time-dependent Hartree-Fock method. The excited states are obtained by a newly developed method through the imaginary-time propagation. Numerical simulations on a helium atom exposed to intense VUV light show that the populations in the excited states after the interaction with the light field are properly obtained as long as the number of spatial orbitals involved in the calculations is sufficiently large.

[Introduction] The multiconfiguration time-dependent Hartree-Fock (MCTDHF) method [1, 2] is a versatile method developed for solving the time-dependent Schrödinger equation (TDSE) for many-electron systems exposed to an intense laser field. For example, the time-dependent wave function of a two-electron system is expressed as

$$\Psi(t) = \sum_{i,j=1}^{M} C_{ij}(t)\, |\phi_i(t)\overline{\phi}_j(t)|, \tag{1}$$

where $C_{ij}(t)$ is a time-dependent configuration-interaction (CI) coefficient, and $|\phi_i(t)\overline{\phi}_j(t)|$ is a Slater determinant constructed from time-dependent spatial orbitals $\phi_j(t)$ $(j = 1, \dots, M)$. The key feature of the MCTDHF method is the use of time-dependent orbitals, which enables an efficient description of both electronic excitation and ionization. In recent years, the MCTDHF method has been successfully applied to the simulation of time-dependent dynamics of many-electron systems such as Ne [3].

[Methods] In this contribution, we investigate time-dependent populations of excited states in He by the MCTDHF method. We first introduce a method to calculate stationary states. The stationary states Φ_n are written in the same form as Eq. (1),

$$\Phi_n = \sum_{i,j=1}^{M} C_{ij}^n\, |\varphi_i^n \overline{\varphi}_j^n|. \tag{2}$$

Each state is constructed from a separate set of CI coefficients C_{ij}^n and orthonormal time-independent orbitals φ_j^n. The same number M of orbitals is used for each stationary state. The CI coefficients and orbitals are obtained by solving the MCTDHF equations of motion in imaginary time. The wave function Φ_k of the state k being propagated in imaginary time is

kept orthogonal to the other stationary states $\Phi_{n<k}$ by introducing suitable Lagrange multipliers. In this way we obtain a set of mutually orthogonal states Φ_n $(n = 0, \ldots, n_{\max})$, variationally optimized with respect to the energies ε_n of the respective states.

The population $p_n(t)$ in the n-th state is defined as the square of the overlap of the MCTDHF wave function $\Psi(t)$ and the stationary state Φ_n,

$$p_n(t) = |\langle \Phi_n | \Psi(t) \rangle|^2. \quad (3)$$

We note that a consistent comparison between populations obtained using different values of M is possible because the excited states Φ_n and the time-dependent wave function $\Psi(t)$ are calculated using the same number of orbitals.

[Results and Discussion] In Fig. 1, we show the time-dependent populations of the first two singlet S states ($1s2s$ and $1s3s$) of He exposed to a 108 nm, 5-cycle laser pulse with a peak intensity of 10^{15} W/cm^2. The central wavelength (108 nm) of the laser pulse is in two-photon resonance with the $^1S\ 1s3s \leftarrow {}^1S\ 1s^2$ transition. As seen in Fig. 1, when the number M of orbitals included in the wave function expansion is smaller than 10, the populations exhibit an oscillatory behavior even after the laser pulse has vanished, indicating that the MCTDHF wave function is not converged. At $M = 12$ orbitals, the $1s2s$ and $1s3s$ populations become stationary after the laser-atom interaction.

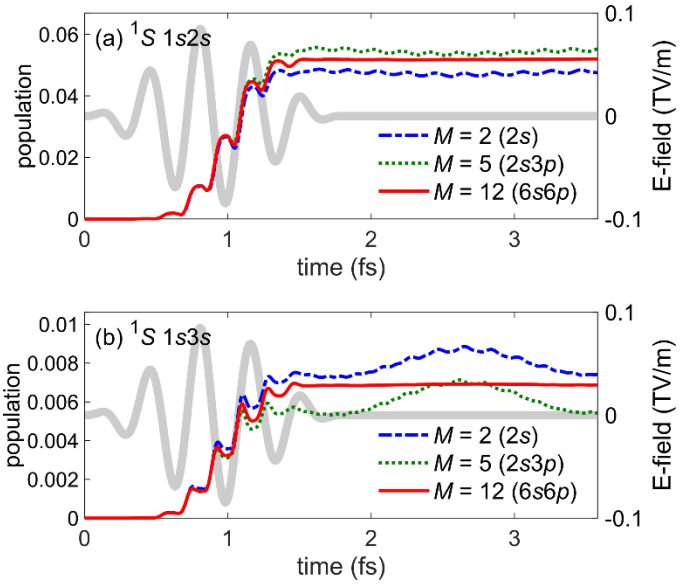

Fig. 1. Time-dependent populations of (a) the ^1S $1s2s$ state and (b) the ^1S $1s3s$ state, for different values of the number of orbitals M. The orbital symmetries at $t = 0$ are indicated in brackets, i.e., $M = 12$ $(6s6p)$ indicates that the initial wave function was constructed from six s-type and six p-type orbitals. The laser field (108 nm, 10^{15} W/cm^2) is indicated by a thick, gray line and the numerical values are shown on the right-hand vertical axis.

We can always expand a wave function $\Psi(t_0)$ at time t_0 in terms of the stationary states as $\Psi(t_0) = \sum_n c_n \Phi_n$. In the absence of a laser field, the time evolution of the wave function according to the TDSE is $\Psi(t) = \sum_n c_n e^{-i\varepsilon_n(t-t_0)/\hbar}\Phi_n$, from which follows that $p_n = |\langle \Phi_n | \Psi(t) \rangle|^2 = |c_n|^2$ is time-independent. The field-free populations of an MCTDHF wave function lack this stationary property in general because of the nonlinear character of the MCTDHF equations of motion. However, as shown in Fig. 1, we recover the correct stationary behavior of the excited-state populations when the number of orbitals M exceeds 10. We note that converged results for other observables such as high-harmonic spectra and ionization probabilities can be obtained at smaller values of $M \leq 5$ [3]. This suggests that the stationarity of excited-state populations can be a sensitive measure of the quality of an MCTDHF wave function.

[References]

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