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Probing two-electron dynamics of helium in time domain via fluorescence channel

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ABSTRACT

We theoretically study two-electron dynamics in a wave packet consisting of low-lying doubly excited states of helium, using an attosecond single-color XUV pump–XUV probe scheme. By solving the time-dependent Schrödinger equation in the hyperspherical coordinates, we analyze an ionization plus excitation process followed by a fluorescence emission. Pump–probe delay time dependence of the transition probabilities, and hence the fluorescence signal, is proved to unravel the bending vibrational motion of the two electrons with the nucleus at the center. Such a motion having an analog to that of a floppy triatomic molecule is the true feature of the electron correlation during temporal evolution of an intrashell two-electron wave packet.

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1. Introduction

Real-time monitoring of ultrafast electron dynamics in atomic and molecular systems can be carried out by pump–probe experiments in which the dynamics are triggered by a pump pulse and subsequently probed by another pulse. Extending the well-established femtosecond scheme [1], an actual attosecond experiment would require both pump and probe pulses in attosecond durations for capturing electron dynamics. The widespread application of such methodology is, however, precluded due to the low focal intensities of existing attosecond sources. Therefore, many pump–probe setups to date have employed a weak extreme-ultraviolet (XUV) attosecond pulse combined with an intense near-infrared (IR) or a visible femtosecond pulse. Strictly speaking, the IR pulse is not only used to probe the wave packet generated by the XUV pulse in the usual manner, it also takes on a role in modulating the electron wave packet, when the two pulses temporally overlap. Moreover, ultrafast modulations could be smeared out by the IR-probe pulse with long duration.

Recent achievement, stimulating this research, has demonstrated a tabletop light source, which generates XUV pulses with duration as short as 500 attosecond and power as high as 1.3 μJ [2].

Such an intense attosecond light source would advance our ability to induce nonlinear phenomena and follow electron dynamics in time domain. Regarding attosecond XUV pump–XUV probe scenarios, there are few proposals for exploring ultrafast electron motion in He atom. The *ab initio* calculations by Hu and Collins [3] showed that a 2-fs oscillation period of a wave packet consisting of singly excited states can be traced by measuring triple-differential cross sections in double ionization. In this scheme, one of the two electrons remains tightly bounded in the ground state, while the other performs an active motion only in the radial direction. As a result, the two-electron dynamics is significantly obscured. In addition, different colors of pump and probe pulses are needed, making experimental situation difficult. Later, a single-color XUV–XUV interferometric pump–probe protocol was put forward [4], managing to follow the two-electron motion in doubly excited resonance states. The temporal evolution is extracted by integrating the double-ionization probability with an appropriate spectral window for one-electron energies. This spectral specification is not always straightforward, and one-photon single ionization component would considerably contribute, especially when the XUV pulses are far from Fourier limit or have significant tail at low-energy region. Moreover, this protocol extracts mainly the radial motion of the outer electron similar to the first scheme. In order to understand the true nature of correlated two-electron motions, another study proposed to extract the molecule-like motions of two excited electrons by analyzing double ionization using appropri-

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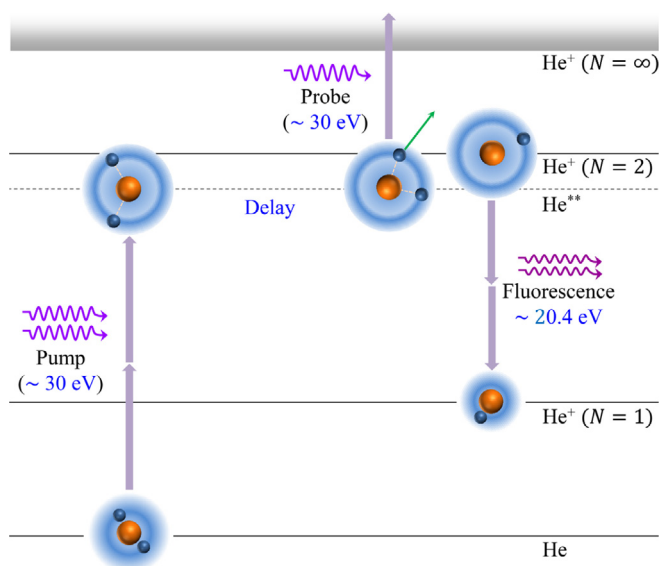


Fig. 1. Schematic diagram shows energy levels and possible XUV pump–XUV probe sequence with single color. The molecule-like motions of two-electron wave packet are observed via fluorescence emission. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

ate coordinates [5]. Such analysis requires a complete experiment, where the full dimensional momentum distribution of the ionized electrons is kinematically resolved. In the view of experimental difficulties, it is desired to realize a feasible setup giving direct access to the molecule-like motions.

In this report, we present a new XUV pump–XUV probe scheme with two identical time-delayed attosecond pulses, which allows to explore the bending vibrational motion of two excited electrons in real time. In our all-optical scheme, we measure only one accessible observable of photon emission. The new setup would therefore rescue us from any difficulty concerning photoelectron detection or coincident measurement. As illustrated in Fig. 1, an XUV-pump pulse interacting with He(1s²) initiates a wave packet of several doubly excited resonance states converging to He+(N=2) + e⁻ threshold. Only the ¹S^e and ¹D^e symmetries are presented due to the dipole selection rule for two-photon absorption. After some delay time for free evolution, another identical XUV pulse singly ionizes the target, leaving the excited states of the residual ion, i.e., He+(2s, 2p) + e⁻. The latter subsequently decays and emits a 40.8 eV photon as He+(2p) → He+(1s) + γ or two photons of 20.4 eV as He+(2s) → He+(1s) + 2γ. They are well spectroscopically separated from the incident XUV photons near 30 eV. Since the two electrons are strongly correlated in doubly excited states, the creation of excited He+ by the probe pulse potentially imprints their behavior at the onset of ionization. Consequently, the fluorescence signal versus pump–probe delay would unravel the electron correlation during the dynamical evolution. We also mention that single ionization to He+(2s, 2p) + e⁻ is the main channel for one photon absorption of the He(2lnl) doubly excited states and others including double ionization are negligibly small. It is because the former is nearly one-electron process, while the latter is true two-electron transition.

2. Method and results

We have accurately calculated the ionization probabilities by solving the time-dependent Schrödinger equation (TDSE) for He atom interacting with XUV pulses,

$$i \frac{\partial}{\partial t} \Psi(t) = [H_0 + V(t)] \Psi(t). \quad (1)$$

Here H_0 is the two-electron Hamiltonian of He, and

$$V(t) = (z_1 + z_2)[f(t) + f(t - \tau)] \quad (2)$$

defines the interaction in the dipole approximation and the length form. Identical pump and probe pulses are described in the following form,

$$f(t) = f_0 \cos^2(\pi t/T) \cos \omega t \quad (3)$$

for $-T/2 \leq t \leq T/2$, and $f(t) = 0$ otherwise. In this work, we set $T = 67.6$ a.u., $f_0 = 0.151$ a.u., and $\omega = 1.11$ a.u., corresponding to the full width at half maximum (FWHM) of 600 as, peak intensity $I = 800$ TW/cm², and photon energy of 30.2 eV. The spectral width $\Delta\omega$ for such cosine-squared pulse shape, defined as the FWHM of the squared Fourier transform of $f(t)$, is $\Delta\omega = 3.65$ eV. These parameters are chosen to dominantly excite three lowest intrashell doubly excited states in the ¹S^e and ¹D^e symmetries via two-photon transition from the ground state. The time propagation is performed using the wave functions described in the hyperspherical coordinates, similar to the method used for ion-He collisions [7]. A detailed description of the numerical method can be found in Ref. [6]. This method involves three steps: (i) Setting up the box-normalized eigenfunctions of H_0 in the hyperspherical coordinates as a basis set by solving the eigenvalue problem, $(H_0 - \epsilon_j)\varphi_j = 0$. (ii) Finding coefficients $C_j(t)$ in the expansion of $\Psi(t) = \sum_j C_j(t)\varphi_j$ by solving the time-dependent equation with the split-operator method. (iii) Extracting the transition amplitudes by projecting the time propagated wavefunction $\Psi(t = \tau + T/2)$ onto the energy-normalized scattering wavefunctions for H_0 with the correct boundary condition. In this way, we are able to calculate the ionization probability in any manifold of He+(nl) + e⁻, and so the fluorescence yield. In order to analyze underlying physics in the two-electron dynamics, we also solve Eq. (1) within the time-dependent perturbation theory (TPTB) in step (ii), using the same basis set in step (i) and the projection manner in step (iii) as described in Ref. [6]. We have checked the convergence of the calculations in terms of all the parameters including the number of basis functions and the box size.

First, we present in the top panel of Fig. 2 the photoelectron spectra for the main resonances triggered by the XUV-pump pulse. The agreement between the TDSE and the second-order TPTB results confirms the dominant contribution of the two-photon process in creating the wave packet, as sketched in Fig. 1. There are three peaks associated with the doubly excited states of 2s²1S^e, 2p²1D^e, and 2p²1S^e. Due to the broad bandwidth of the short XUV pulse, a bunch of resonances belonging to the ¹S^e and ¹D^e symmetries are also populated in the photoelectron energy region higher than 38 eV (not shown). The doubly excited states of He exhibit large configuration mixing based on the mean-field theory. They are, therefore, well characterized by the so-called correlation quantum numbers, $(K, T)^A$ [8] as indicated in the top panel of Fig. 2. However, we will use the convention designated by the dominant configurations. It is well established that the two-electron motions in these doubly excited states are most adequately described by analogy to the rotation and vibration of a floppy triatomic molecule. In order to visualize such motions, we define the vibrational and rotational density distributions by

$$\rho_j^{vib}(\Omega_v) = \int |\varphi_j|^2 d\Omega_r dR, \quad (4a)$$

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