Control of recollision wave packets for asymmetric molecular imaging using an 800 nm+400 nm multicycle bichromatic laser field

Lingling Ji *, Yuhua Wang

College of Sciences, Wuhan University of Science and Technology, Wuhan 430081, People's Republic of China

ABSTRACT

Control of recollision wave packets for asymmetric molecular imaging using an 800 nm+400 nm multicycle bichromatic laser field is explored. With a semi-classical model, it is shown that the one-directional recollision of the electron with the core, which is required for asymmetric molecular imaging based on high-order harmonic generation, can be achieved by adjusting the relative phase between the two components of the bichromatic field. Following the tomographic procedure, the asymmetric orbital is satisfactorily reconstructed with a well tailored 800 nm+400 nm bichromatic field.

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1. Introduction

The main impetus to the development of attosecond sources of coherent X-rays is measuring and understanding the electronic structure and correlated dynamics of matter on its natural timescale [1–16]. Recently, the molecular orbital tomography (MOT) based on high-order harmonic generation (HHG) has attracted a great deal of attention [17–24], due to its potential applications in probing the electron dynamics in attosecond scales and in following the chemical reactions. This MOT scheme was first proposed by Itatani et al. [17] to reconstruct the highest occupied molecular orbital (HOMO) of N_2 molecule. The main idea behind the MOT is that the recolliding electron wave packet, which leads to high harmonic emissions and takes the role of a probe for the molecular structure at the recollision instant, can be approximately regarded as a plane wave. The transition matrix element describing the recombination of the electron with the core is then related to a Fourier transform of the bound-state orbital. Thus, according to the Fourier slice theorem, the target molecular orbital can be reconstructed by performing the inverse Fourier transform of the recombination matrix elements, which can be extracted from the high-order harmonics. In the case of symmetric molecules, the extraction of the recombination matrix elements can be directly performed by following the MOT procedure. As for asymmetric molecules, however, the one-directional recollision of the electron with the parent ion is required [19]. In Ref. [22], it is shown that the unidirectional

recollisions can be achieved using an extremely short tailored laser pulse. However, a requirement of a single-cycle pulse with a stabilized and controllable CEP in this one-color scheme is rather stringent for many laboratories. In Ref. [23], a scheme with less stringent experimental conditions is demonstrated based on the efficient control of the electron dynamics by the bichromatic laser field [25–34]. In their scheme, the authors choose a mid-infrared (1600 nm) and a weak 800 nm laser pulses as the fundamental and controlling fields respectively to synthesize a multicycle bichromaticfield. When using a long-wavelength (such as mid-infrared) laser, although the harmonic spectrum can be significantly extended, the intensities of the high-order harmonics decrease due to the $\lambda^{-(5\sim6)}$ dependence of the harmonic yield on the laser wavelength at constant laser intensity [35]. Additionally, multiple recollision of the electron wave packets also become important in HHG from a long-wavelength laser pulse, which may lead to the control of electron wave packets for asymmetric molecular imaging complicated and difficult.

In this paper, we adopt the two-color scheme to control the recollision wave packets for asymmetric molecular imaging. But an 800 nm laser pulse is chosen as the fundamental field with its second harmonic field as the controlling field. Using a semi-classic model, we calculate the returning probability of the electron as a function of the recombination momentum for various bichromatic fields. It is found that the one-directional recollision of the electron can be achieved by adjusting the relative phase between the 800 nm and 400 nm laser fields. Following the tomographic procedure, the recombination matrix elements are extracted and the asymmetric orbital is satisfactorily reconstructed with a well tailored 800 nm +400 nm bichromatic field.

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* Corresponding author.
  E-mail address: jll_gluck13@163.com (L. Ji).

2. Model

This section includes three independent parts: "Returning probability with momentum $\mathbf{v}_r$", "Simulating the HHG" and "Reconstruction method". In the first part, a semi-classical model used to calculate the probability that an electron returns with momentum $\mathbf{v}_r$ is introduced. In the second part, we describe the modified Lewenstein model in the velocity form, which is applied in this work to simulate the HHG. In the third part, the orbital reconstruction method in the velocity form is presented.

2.1. Returning probability with momentum $\mathbf{v}_r$

To characterize the returning wave packet, the probability that an electron returns with momentum $\mathbf{v}_r$ is calculated semi-classically [22,23]. In detail, the ionization of the electron at the ionization time t_i is described using the Molecular Ammosov–Delone–Krainov (MO-ADK) model for oriented molecules. After ionized by the driving field, the continuum electron is treated classically. Every classical return receives a weight based on the tunnelling probability and a factor τ^{-3} reflecting the effect of wave-packet spreading. τ is the excursion duration and given by $\tau = t_r - t_i$ [36], where t_r is the recombination time of the electron. Considering the effect of wave-packet spreading, the electron trajectories where the excursion duration is larger than a laser cycle are neglected. For a given ionization time t_i, the corresponding t_r is determined by [37–40]

$$\int_{t_i}^{t_r} [\mathbf{p}+\mathbf{A}(t')] \, dt' = 0 \tag{1}$$

$$\frac{[\mathbf{p}+\mathbf{A}(t_i)]^2}{2} = 0 \tag{2}$$

where $\mathbf{A}(t)$ and I_p are the vector potential of the driving field and the ionization energy of the target molecular orbital, respectively. $\mathbf{p}$ is the canonical momentum. When the electron appears in the continuum it is under the influence of a very strong laser field, and when it comes back to the nucleus it has a large kinetic energy, so that the effect of the coulomb potential can be neglected during the propagation of the continuum wave packet. As a result, the canonical momentum $\mathbf{p}$ is constant between t_i and t_r. After obtaining the recombination time t_r, the recombination momentum $\mathbf{v}_r$ is then given by [41] $\mathbf{v}_r = \mathbf{A}(t_r) - \mathbf{A}(t_i)$.

2.2. Simulating the HHG

The HHG is simulated by using the strong field approximation (SFA) model [42–46] for molecules. Instead of the time-dependent dipole moment, we calculate the time-dependent dipole momentum, since for simple molecules the velocity form was shown to give more accurate results within SFA [47]. Within the single active electron (SAE) approximation, the time-dependent dipole velocity is given by

$$\mathbf{v}_{dip}(t;\theta) = i \int_{-\infty}^{t} dt' \left[\frac{\pi}{\zeta + i(t-t')/2} \right]^{3/2} \exp[-iS_{st}(t',t)]$$
$$\times \mathbf{F}(t') \cdot \mathbf{d}_{ion}[\mathbf{p}_{st}(t',t)+\mathbf{A}(t');\theta]$$
$$\times \mathbf{v}_{rec}^*[\mathbf{p}_{st}(t',t)+\mathbf{A}(t);\theta]+c.c. \tag{3}$$

In this equation, ζ is a positive constant. t' and t correspond to the moments of electronic ionization and recombination, respectively. θ is the alignment angle between the molecular axis and the polarization of the probe pulse. $\mathbf{F}(t)$ refers to the electric field of the driving laser pulse, and $\mathbf{A}(t)$ is its associated vector potential. $\mathbf{p}_{st}$ and S_{st} are the stationary momentum and the quasi-classical action, which are given by

$$\mathbf{p}_{st}(t',t) = -\frac{1}{t-t'} \int_{t'}^{t} \mathbf{A}(t'') \, dt'' \tag{4}$$

and

$$S_{st}(t',t) = \int_{t'}^{t} \left(\frac{[\mathbf{p}_{st}+\mathbf{A}(t'')]^2}{2} + I_p \right) dt'' \tag{5}$$

Then the complex amplitudes of the high-order harmonics with a frequency ω_n are given by

$$\tilde{\mathbf{E}}(\omega_n,\theta) = \int e^{i\omega_n t} \frac{d}{dt} \mathbf{v}_{dip}(t;\theta) \, dt. \tag{6}$$

2.3. Reconstruction method

To reconstruct the molecular orbital, one must calculate or measure the high-order harmonic spectra (including the intensity and phase) from the oriented target molecule at various orientation angles θ. In addition, the harmonic spectrum from a reference atom, which has an ionization energy similar to that of the target molecule, should be also calculated or measured with the same driving laser pulse. By calibrating the measured HHG signal of the target molecule with that of the reference system, the recombination dipole velocity element is extracted [18,20]:

$$d_{x/y}^{V}(\omega_n,\theta) = \frac{1}{\eta(\theta_i)} \frac{\tilde{E}_{x/y}^{mol}(\omega_n,\theta)}{\tilde{E}^{ref}(\omega_n)} d_{ref}^{V}(\omega_n), \tag{7}$$

where $\eta(\theta_i)$ is the scaling factor in molecules depending on the ionization angle θ_i and is calculated by the MO-ADK theory [48]. $d_{ref}^{V}(\omega_n)$ is the recombination dipole velocity element of the "known" reference atom.

Using the extracted recombination dipole velocity, the reconstruction of the molecular orbital can be performed in the velocity form:

$$\Psi_q^{D}(x,y) = \mathcal{F}_{k\to r} \left[\frac{d_q^{V}(k_x,k_y)}{k_q} \right] \quad (q=x,y) \tag{8}$$

Here, $\Psi^{D}(x,y)$ is a two-dimensional (2D) projection of the target molecular orbital on the plane orthogonal to the pulse propagation direction.

3. Result and discussion

We take the example of CO molecule to demonstrate the efficient control of the recollision wave packet by the 800 nm+400 nm multicycle laser field. The pulse duration of the bichromatic field used in our simulation is about 30 fs full width at half maximum, and the intensity of the 800 nm and 400 nm pulses are 3.0×10^{14} W/cm^2 and 2.7×10^{13} W/cm^2, respectively. The bichromatic laser pulse is propagated along the z-axis and its two components are both linearly polarized along the y-axis. The electric field is given by [49,50] $E(t) = E_0 \sin^2(\pi t/10T_0) \cos(\omega_0 t) + E_1 \sin^2(\pi t/10T_0) \cos(2\omega_0 t+\phi)$, where ω_0 and T_0 denote the frequency and optical cycle of the fundamental field, respectively. E_0 and E_1 are the amplitudes of the 800 nm and 400 nm pulses, respectively. ϕ is the relative phase between the two pulses, which can be easily adjusted in experiment.

To illustrate how the one-directional recollision of the electron can be achieved by the bichromatic field, we first calculate the classical trajectories of the returning electron in the fundamental field. The dependence of the recombination momentum v_r on the ionization (red dots) and recombination (blue crosses) times is presented in Fig. 1(b). The electric field (dashed green line) of the fundamental laser pulse is also shown in Fig. 1(a). As shown in Fig. 1, the recollision direction of the returning electron is reversed every half optical cycle and is determined by the direction of the electric field at the ionization time. Take the classical trajectories in the fourth optical cycle as an example. In the first half optical