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Interpulse interference of electron emission from an atom irradiated by sinusoidally phase-modulated pulse*

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We theoretically investigate the photoelectron emission from an atom irradiated by an amplitude modulated sinusoidally phase-modulated pulse through solving the time-dependent Schrödinger equation in the momentum space. By controlling the phase amplitude of the pulse in the frequency domain, it can be found that the photoelectron spectra appear as explicit interference phenomena, which originated from the interference between the directly ionized electron and the ionization of the pre-excited atom from different subpulses.

Keywords: high-order harmonic generation, above threshold ionization

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1. Introduction

When atoms are irradiated by strong laser pulses, there are many nonlinear phenomena, such as high-order harmonic generation, nonsequential double ionization, and above threshold ionization (ATI).[1–3] ATI is an ionization progress that an atom can absorb more photons than its ionization threshold.[4] For higher laser intensity, there are two plateaus in the photoelectron spectra, which correspond to the direct ionization and the rescattering process, respectively.[5,6]

Due to the ionized electron rescattering with its parent ion, the rescattering electron carries the real-time structure information.[7,8] The photoelectron with higher energy can be used to detect structures of atoms and molecules.[9] When an atom interacts with a high frequency laser field, the lower order photoelectron also encodes the atomic orbital information.[10,11] Especially, the laser electric field plays an important role in the interaction between the atom and the laser pulse, and the photoelectron emission is sensitive to the laser's parameters. Therefore, photoelectron spectroscopy is one of the important methods to detect the carrier envelope phase of the laser pulse.[12] In order to apply photoelectron spectroscopy well, many researchers made in-depth studies of its generation process.[12–19]

When the atom is driven by the strong laser pulse, according to the incident laser intensity, the ionization mechanism can be divided into multiphoton and tunneling ionization. For the multiphoton ionization process, there are many individual peaks at the photoelectron spectra, and the energy difference between the adjacent peaks is one photon energy (ω_0). The energy position of these peaks is predicted by the conversation of energy $n\omega_0 - (I_p + U_p)$, where the I_p is the ionization energy of an atom and U_p is the ponderomotive energy of the laser electric field.[20] From the time domain perspective, these photoelectron peaks are generated by the interference of the ionized electrons from different cycles in the laser pulse. In the photoelectron spectra, the interference structures from the various instants in one optical cycle also can be observed.[21] Using the interference information between the directly ionized electron and the rescattering electron, one can obtain the differential cross section and the corresponding phase of the electron-ion scattering amplitude.[22]

When the atom is irradiated by the multi-color laser pulse, there are many novel interference processes in the photoelectron emission.[23–29] The interference structure can be investigated by the semiclassical scheme in the tunneling regime.[30–32] Using the additional laser pulse, the interference of wave packets can be controlled. Moreover, the shaped pulse laser provided more convenience to control the photoelectron interference. By controlling the amplitude, phase, and polarization of the pulse in the frequency domain, the pulse with an almost arbitrary shape can be obtained in the time domain.[33,34] For the sinusoidally phase-modulated pulse, one Fourier-transform-limited pulse can be divided into many subpulses. The ionization and excitation of the atom in this pulse were studied.[35,36] In the research, few level models are available and the calculated results are in qualitative agreement

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with the experiment for the weak field. In order to investigate the photoelectron from the modulated pulse, it needs a very large space to include the total ionized electron. Thereby, for the intense laser field, it is rather difficult to investigate the photoelectron emission under the coordinate representation. This problem can be avoided by solving the time dependent Schrödinger equation (TDSE) in momentum space.[37,38] In this work, using this scheme, we investigate the photoelectron emission of an atom irradiated by sinusoidally phase-modulated shaping pulses. It is found that the photoelectron spectra from the phase-modulated laser pulses have many subpeaks, which is caused by the interference from the ionized electrons from different laser subpulses through the analysis of the time-dependent population of the continuum states.

2. Model and method

In order to obtain the photoelectron spectra, we calculated the time-dependent wavefunction under the momentum representation.[39,40] In the velocity gauge and the dipole approximation, the TDSE of an atom in a strong laser pulse can be expressed as (atomic units are used throughout, unless otherwise stated):

$$\mathrm{i}\frac{\partial}{\partial t}\Psi(\boldsymbol{r},t)=\left[\frac{\hat{\boldsymbol{p}}^2}{2}-\boldsymbol{A}(t)\cdot\hat{\boldsymbol{p}}+V(\boldsymbol{r})\right]\Psi(\boldsymbol{r},t), \tag{1}$$

where $\boldsymbol{A}(t)$ is the vector potential of the laser field, and $V(\boldsymbol{r})$ is the atomic Coulomb potential.

By using the momentum eigenfunction as the basis set, the time-dependent wavefunction can be expanded as

$$\Psi(\boldsymbol{r},t)=\frac{1}{(2\pi)^{3/2}}\int\Phi(\boldsymbol{p},t)\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{r})\,\mathrm{d}\boldsymbol{p}. \tag{2}$$

By inserting Eq. (2) into Eq. (1), one can obtain the TDSE in the momentum space

$$\begin{aligned}\mathrm{i}\frac{\partial}{\partial t}\Phi(\boldsymbol{p},t) &= \left[\frac{\boldsymbol{p}^2}{2}-\boldsymbol{A}(t)\cdot\boldsymbol{p}\right]\Phi(\boldsymbol{p},t)\\ &\quad+\int V(\boldsymbol{p},\boldsymbol{p}')\Phi(\boldsymbol{p}',t)\,\mathrm{d}\boldsymbol{p}'\\ &= \hat{H}_0(\boldsymbol{p})+\hat{U}(\boldsymbol{p},t),\end{aligned} \tag{3}$$

where $V(\boldsymbol{p},\boldsymbol{p}')$ is the potential in the momentum space. Here,

$$\hat{H}_0(\boldsymbol{p})=\frac{\boldsymbol{p}^2}{2}\Phi(\boldsymbol{p},t)+\int V(\boldsymbol{p},\boldsymbol{p}')\Phi(\boldsymbol{p}',t)\,\mathrm{d}\boldsymbol{p}'\Phi(\boldsymbol{p},t), \tag{4}$$

$$\hat{U}(\boldsymbol{p},t)=(-\boldsymbol{A}(t)\cdot\boldsymbol{p})\Phi(\boldsymbol{p},t), \tag{5}$$

$$V(\boldsymbol{p},\boldsymbol{p}')=\frac{1}{(2\pi)^3}\int V(\boldsymbol{r})\exp[-\mathrm{i}(\boldsymbol{p}'-\boldsymbol{p})\cdot\boldsymbol{r}]\,\mathrm{d}\boldsymbol{r}. \tag{6}$$

The time-dependent wavefunction can be integrated by the second order split-operator technique

$$\begin{aligned}\Phi(\boldsymbol{p},t+\Delta t) &\simeq \exp(-\mathrm{i}\hat{H}_0\Delta t/2)\\ &\quad\times\exp(-\mathrm{i}\hat{U}(\boldsymbol{p},t+\Delta t/2)\Delta t)\\ &\quad\times\exp(-\mathrm{i}\hat{H}_0\Delta t/2)\Phi(\boldsymbol{p},t)+O(\Delta t^3).\end{aligned} \tag{7}$$

For the hydrogen atom, the potential is given by

$$V(\boldsymbol{p},\boldsymbol{p}')=-\frac{1}{2\pi^2}\frac{1}{|\boldsymbol{p}-\boldsymbol{p}'|^2}. \tag{8}$$

There is a quadratic singularity at $\boldsymbol{p}=\boldsymbol{p}'$, and the singularity can be removed by using the Landé subtraction technique,[41] which can be realized by the partial wave expansion scheme.

Here, we map the semi-infinite domain $[0,\infty]$ to the finite one $[-1,1]$, then use the Gaussian quadrature to discretize the grids in the momentum space.[42] The mapping function is taken as

$$p=p(x)=L\frac{1+x}{1-x}, \tag{9}$$

where L is a mapping parameter. If we choose a smaller value for L, there are more grid points at small p. By projecting the scatter continuum state on the time-dependent wavefunction at the end of the laser pulse, one can obtain the continuum state population amplitude $b_l(\varepsilon,t)$, and the corresponding single differential cross section is:

$$\frac{\mathrm{d}P_\varepsilon}{\mathrm{d}\varepsilon}=\sum_l|b_l(\varepsilon,t)|^2. \tag{10}$$

3. Results and discussions

In the frequency domain, the phase-modulated laser pulse can be changed by the phase function: $\varphi(\omega)=A\sin[(\omega-\omega_{\mathrm{ref}})*T+\varphi_{\mathrm{int}}]$, where A is the amplitude of the modulation function, T is the frequency of the oscillation function, and φ_{int} is the phase offset of the Sinusoidal function. φ_{int} is set to 0 in this paper. In the time domain, the sinusoidal phase modulation pulse produces a sequence of subpulses, the separation of which is determined by the amplitude T. The relative peak amplitudes of subpulses are controlled by the value of A. Figure 1 shows the electric fields of the sinusoidally phase-modulated laser pulses with $A=0$, 0.5, and 1. Here, the envelope of the laser pulse is chosen as the Gaussian shape with a duration of 16.45 fs full width at half maximum. It can be seen from Fig. 1, for $A=0$, that there is only one pulse, the duration of which equals to the Fourier transform limit; for $A=1$, the pulse is mainly split into five subpulses, and the three main subpulses in the middle of the pulse are marked as SP_1, SP_2, and SP_3, respectively. With the increase of A, the intensities of the corresponding subpulses become stronger.

The photoelectron emission irradiated by these sinusoidal pulses are shown in Fig. 2. There are dramatic differences in the photoelectron spectra for different amplitudes. One can notice that, with the enhancement of A, the photoelectron amplitude gets smaller, and more subpeaks appear in the spectra,

in addition, the energies of the main peaks in the photoelectron spectra are increased. For the Fourier-transform-limited pulse, the peak energy can be predicted by $n\omega_0 - I_p - U_p$, where $\omega_0 = 0.057$ is the laser frequency. From the subgraph in Fig. 1, one can see that, with the increase of A, the intensity of the maximum electric field is reduced, and the corresponding U_p is decreased. Therefore, the energies of the main peaks in the photoelectron spectra are enhanced with the increase of the phase amplitude A. Otherwise, there are subpeaks in the photoelectron spectra from Fig. 2, which is caused by the ionization from the excited states.[43]

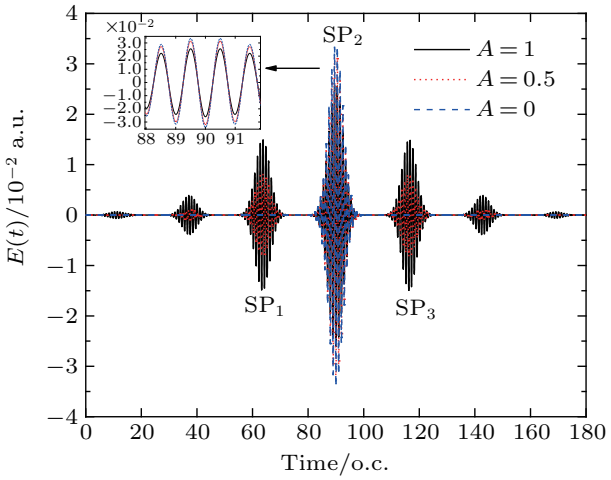


Fig. 1. (color online) Time variation of sinusoidally phase-modulated electric fields with different phase amplitude A. The unit o.c. is short for optical cycle.

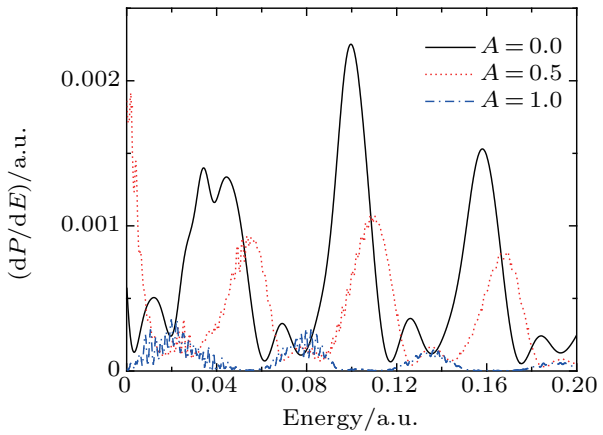


Fig. 2. (color online) Photoelectron spectra of the hydrogen atom irradiated by the sinusoidally phase-modulated electric fields with different phase amplitudes A.

Next, we examine the dependence of the photoelectron spectra on the phase amplitude A of the modulated pulse, as shown in Fig. 3. It can be clearly observed that, as A is increased, the energies of the main peaks in the photoelectron spectra are enhanced. For example, the energy of the first main peak equals to 0.04 for $A = 0$, then it moves to 0.07 when $A = 1$. This result is consistent with that discussed above in Fig. 2, and the variation of the main peak's energy can be attributed to the reduction of the intensity of the subpulse SP$_2$, which also leads to the decrease of the photoelectron amplitude with the increase of A, which can be seen from Fig. 3.

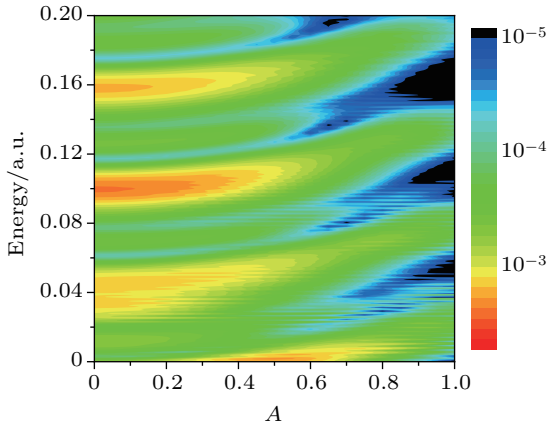


Fig. 3. (color online) The variation of the photoelectron spectra with the phase amplitude of the sinusoidally phase-modulated pulse.

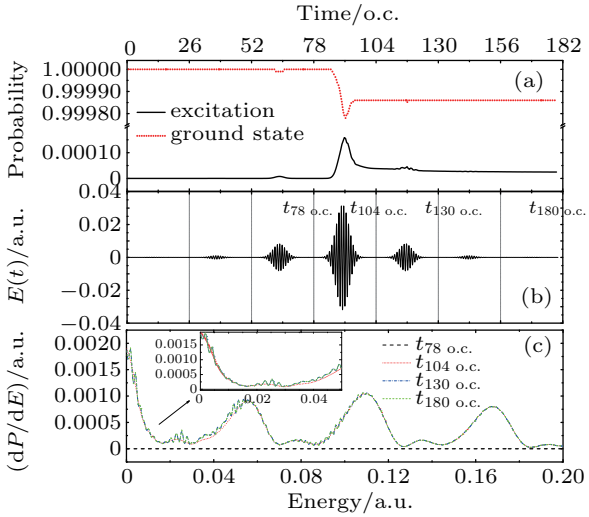


Fig. 4. (color online) (a) The populations of the ground state and the excited states; (b) laser electric field of the sinusoidally phase-modulated pulse with the phase amplitude $A = 0.5$; and (c) photoelectron spectra from different instants.

In order to gain insights into the generation mechanism of the subpeaks in the photoelectron spectra, the transient study of the photoelectron emission is adopted. Figure 4(a) shows the populations of the ground state and the excited states in the case of $A = 0.5$. The corresponding electric field of this modulated laser pulse is presented in Fig. 4(b), and three important subpulses exist. After the atom is irradiated by the set of subpulses, the photoelectron spectra are calculated from the time-dependent wavefunctions at instants (t = 78 o.c., 104 o.c., 130 o.c., and 180 o.c.), as shown in Fig. 4(c). When the atom is irradiated by the subpulse SP$_1$, due to the low electric field intensity of SP$_1$, the ionization is very small and so is the photoelectron intensity. After the action of subpulses SP$_1$ and SP$_2$, the photoelectron emission appears to have no subpeaks, which is similar to that from the Fourier-transform-limited pulse with weak intensity. When the atom is irradiated by three main subpulses, the photoelectron spectrum is

consistent with that calculated from the end of the laser pulse ($t = 180$ o.c.). The above results indicate that the interference between the ionized electrons from SP_2 and SP_3 produces the subpeaks in the photoelectron spectra.

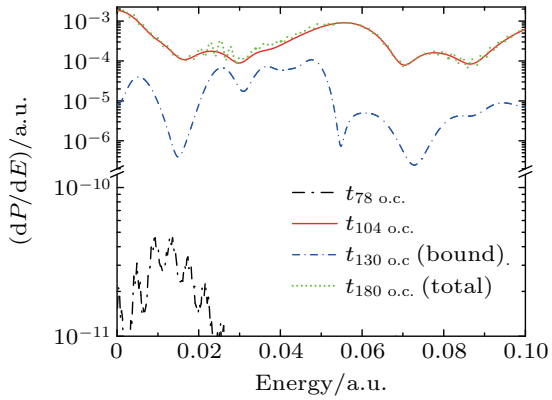


Fig. 5. (color online) Photoelectron spectra calculated from the wavefunction at various instants when the continuum part of the time-dependent wavefunction is subtracted at instant $t = 104$ optical cycles.

In order to clearly see the difference between the photoelectron spectra, we use the logarithm scale to replot the spectra at instants $t = 78$ o.c., 104 o.c., and 130 o.c. After the atom is irradiated by the subpulse SP_1, the intensity of the photoelectron spectrum at $t = 78$ o.c. is very weak, as shown by the black dash line in Fig. 5. When the SP_2 comes, the atom is not only ionized but also excited, which can be clearly observed in Fig. 4(a). Due to the subpulse SP_2 with higher intensity, the photoelectron intensity at $t = 104$ o.c. is increased in excess of six orders of magnitude compared with that at $t = 78$ o.c., and there are no subpeaks in the spectrum, as presented by the red solid curve in Fig. 5. After the action of the subpulse SP_3, the photoelectron spectrum at $t = 130$ o.c. is almost the same as that at $t = 104$ o.c., however, subpeaks appear, as shown by the green dash curve in Fig. 5. From Fig. 4(a), one can notice that the population of the ground state is almost unchanged after the SP_3, and the population of the excited states is significantly reduced. Therefore, the electron ionized from the excited states by the subpulse SP_3 contributes to the subpeaks in the spectrum. In order to obtain deeper insight for the subpeak generation, we calculate the photoelectron spectra at $t = 130$ o.c., and the continuum part of the time-dependent wavefunction is artificially subtracted at the instant $t = 104$ o.c., as presented by the blue dash dot curve in Fig. 5. Compared with the case of the green dash curve, the subpeaks disappear in the photoelectron spectrum. Because the contribution of the ionized electron before SP_3 is artificially removed, there only exists the ionized electron from SP_3 in the case of the blue dash dot curve. It means that the subpeaks generation in the photoelectron spectra can be attributed to the interference between the ionized electrons from the subpulses SP_2 and SP_3.

As explained above, the subpeaks are generated from the interference between the ionization from the subpulses SP_2 and SP_3. The feature of the subpeaks should be associated with the parameters of these subpulses. The laser electric fields with different T are present in Fig. 6(a). This figure reveals that there has a slight difference for the subpulse SP_3. The photoelectron emission irradiated by the two pulses are displayed in Fig. 6(b). There is a significant difference in the subpeaks of the photoelectron emission, although the overall structure of the photoelectron spectra is consistent. The discrepancy is caused by the ionization delay of the atom irradiated by the subpulse SP_3.

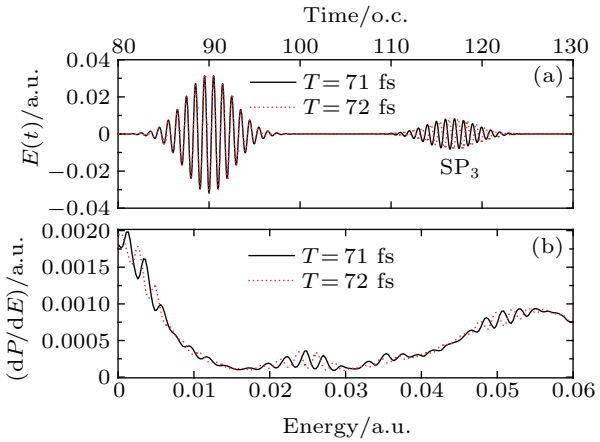


Fig. 6. (color online) (a) Time variation of sinusoidally phase-modulated electric fields with different delay amplitude T; (b) photoelectron spectra of the hydrogen atom irradiated by the laser field of panel (a).

The more detailed photoelectron spectra from the atom irradiated by the modulated pulse with the variety of T is shown in Fig. 7. What stands out in this figure is that the energy of subpeaks is proportional to the phase delay T. One may measure the modulated parameter of the shaping pulse using the sensitivity of the photoelectron emission of the subpulse's behavior.

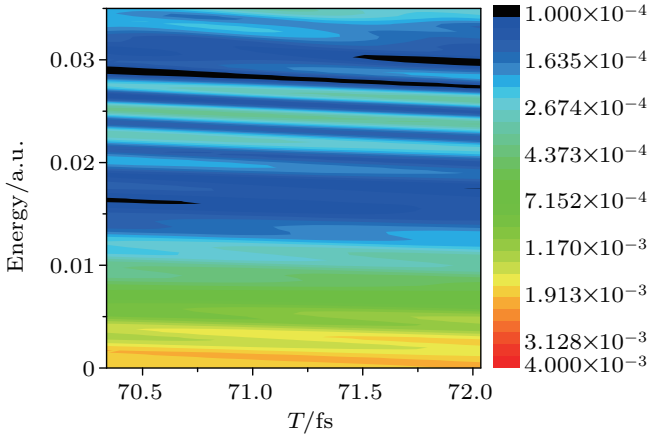


Fig. 7. (color online) Variety of photoelectron spectra of the hydrogen atom irradiated by a sinusoidally phase-modulated pulse with the phase delay T.

4. Conclusion

In summary, the atom is not only ionized but excited by the main subpulse in the modulated pulse, then the excited state can be further ionized by the successive subpulse after the main one, the interference between the ionized electrons from these subpulses leads to the subpeaks in photoelectron spectra. Therefore, the inter-pulse interference of the photoelectron emission can be observed from the atom in the amplitude modulated sinusoidally phase-modulated pulse.

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