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Simultaneous study of the lower order harmonic and photoelectron emission from an atom in intense laser pulse*

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We simultaneously investigate variations of a low order harmonic and photoelectron emission with an incident laser intensity by solving the time-dependent Schrödinger equation in a momentum space. It can be found that, the intensity of low order harmonic and photoelectron are gradually enhanced with the increase of the laser intensity, when the laser frequency is not in resonance with the transition frequency between the laser-induced high excited states and the ground state. If the resonance occurs, the intensity of the lower order harmonic is reduced and the interference can be observed in the lower order photoelectron spectra.

Keywords: high-order harmonic generation, above threshold ionization

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1. Introduction

In recent years, there has been increasing interest in the study of atoms irradiated by ultrashort intense laser pulses due to the development of laser technology.[1–3] Many novel phenomena are observed in the process, such as frustrated tunneling ionization (FTI), above threshold ionization (ATI), high harmonic generation (HHG) and so on.[4–10] HHG is one of table-top soft x-ray laser sources and is used for imaging nano-structure.[11,12] In addition, HHG is an important way to generate an ultrashort pulse, whose duration is a scale of attoseconds. By combining the attosecond pulse with an infrared laser, the electron dynamics of atoms and molecules can probed.[13–17]

In general, these strong phenomena can be understood by the 'three-step model'.[18] The bound electron tunnels out from the atomic potential and then is driven back by the laser electric field to the mother ion with high kinetic energy. The rescattering effect of the electron may result in the generation of high-order harmonics and ATI. There are plenty of works about independently investigating the two phenomena. These studies provided deeper insight into the mechanism of the interaction between atoms and laser electric field.[19,20] The control and applications of HHG and ATI are reported by many groups.[21–23] For instance, the photoelectron emission was taken as an imaging tool for the molecular structure because it carries molecular information through the rescattering process.[24–26] For a few cycle pulses, there is an one-to-one mapping between high-order harmonic and above threshold ionization. Through the simultaneous analysis of HHG and ATI, one can improve the accuracy of the molecular structure imaging.[27]

More recently, there is a tendency to focus on the generation process of low order harmonic and ATI.[28–35] The low order harmonic is an important way to generate a frequency comb in the extreme ultraviolet.[21] The emission intensity of the low order harmonic is highly dependent on a good phase matching. When an atom is irradiated by the mid-infrared laser field, there is a novel low energy structure in the photoelectron spectra.[35] The Coulomb potential plays an important role in the generation of this structure. To further study the physical mechanism of the low order harmonic and photoelectron emission, we simultaneously study these processes by solving the time-dependent Schrödinger equation (TDSE) in the momentum space. This scheme is accurate and contains the effect of Coulomb potential. By simultaneous analysis the variation of the low order harmonic and ATI with incident laser the intensity, it is found that these processes can be affected by the population of higher excited states.

Atomic units are used throughout this paper unless otherwise indicated.

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2. Model and method

In order to study HHG and ATI, one need to numerically solve the time-dependent Schrödinger equation for the interaction between an atom and the laser pulse. There is a significant difference in demand for the numerical calculation of HHG and ATI. The harmonic generation mainly occurs near the nuclear, thereby large numbers of computational grids are needed to describe the rapid change of the potential. In the photoelectron calculation, a large spatial scale is adopted to collect the whole of the ionized electrons. The generalized pseudospectral method in momentum space can satisfy the requirements for the calculation of HHG and ATI.[36–39]

In the velocity gange and the dipole approximation, the time-dependent Schrödinger equation in momentum space is

$$\begin{aligned} \mathrm{i}\frac{\partial}{\partial t}\Phi(\boldsymbol{p},t) &= \left[\frac{\boldsymbol{p}^2}{2} - \boldsymbol{A}(t)\cdot\boldsymbol{p} + \int V(\boldsymbol{p},\boldsymbol{p}')\mathrm{d}\boldsymbol{p}'\right]\Phi(\boldsymbol{p},t) \\ &= [\hat{H}_0(\boldsymbol{p}) + \hat{U}(\boldsymbol{p},t)]\Phi(\boldsymbol{p},t), \end{aligned} \tag{1}$$

where

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

is the Coulomb potential and $\boldsymbol{A}(t) = A_0 f(t)\sin(\omega t)$ is the vector potential of the laser pulse. Here, $A_0 = E_0/\omega_0$, E_0 and $\omega_0 = 400$ nm are the peak amplitude and fundamental frequency of the laser electric field, respectively. The envelope function of the laser pulse is $f(t) = \sin^2(\omega t/(2N_{\mathrm{total}}))$. $N_{\mathrm{total}} = 30$ o.c (o.c. is short for optical cycles) is the number of optical cycles in the pulse. In Eq. (1)

$$\hat{H}_0(\boldsymbol{p}) = \frac{\boldsymbol{p}^2}{2} + \int V(\boldsymbol{p},\boldsymbol{p}')\mathrm{d}\boldsymbol{p}', \tag{2}$$

and

$$\hat{U}(\boldsymbol{p},t) = -\boldsymbol{A}(t)\cdot\boldsymbol{p}. \tag{3}$$

Second-order split-operator scheme was employed to obtain the wavefunction at instant $t+\Delta t$

$$\begin{aligned} \Phi(\boldsymbol{p},t+\Delta t) \simeq\ & \exp(-\mathrm{i}\hat{H}_0\Delta t/2) \\ & \times \exp(-\mathrm{i}\hat{U}(\boldsymbol{p},t+\Delta t/2)\Delta t) \\ & \times \exp(-\mathrm{i}\hat{H}_0\Delta t/2)\Phi(\boldsymbol{p},t) + O(\Delta t^3). \end{aligned} \tag{4}$$

Using the mapping function, fewer grid points are required to obtain accurate results.[40] In the calculation, the mapping parameter is 1.8, the grid number in $\boldsymbol{p}$ is 3000, and the angular momentum number is 40. One can obtain the continuum state population amplitude $b_l(\varepsilon,t)$ by projecting the scatter continuum state on the wavefunction. Here, l is the partial wave quantum number. The single differential cross section of the photoelectron is[41]

$$\frac{\mathrm{d}P_\varepsilon}{\mathrm{d}\varepsilon} = \sum_l |b_l(\varepsilon,t)|^2. \tag{5}$$

The harmonic spectra of the atom are calculated from the Fourier transform of the time-dependent induced dipole in velocity form

$$\mathrm{d}(t) = \langle\Phi(\boldsymbol{p},t)|p_z|\Phi(\boldsymbol{p},t)\rangle. \tag{6}$$

The population of the bound states is calculated by projecting the bound state on the final wavefunction

$$p_{n,l} = |\langle\psi_{n,l}(\boldsymbol{p})|\Phi(\boldsymbol{p},t)\rangle|^2. \tag{7}$$

3. Results and discussions

The laser intensity effect on the ionization and high harmonic generation processes are systematically investigated in this work. Since these processes are related to the ionization and excitation of the atom, we first investigate the population's variation with the laser intensity. In Fig. 1, the population's variation of the ground state (black solid line), excited states (red dotted line) and continuum states (blue dashed–dotted line) with the intensity of the incident pulse is presented. Here, the unit of intensity I_{laser} is $(U_\mathrm{p}+I_\mathrm{p})/\omega_0$, where $U_\mathrm{p} = E_0{}^2/(4\omega_0{}^2)$ is the pondermotive energy of the laser electric field and hydrogen is used in the simulations with the atomic ionization energy $I_\mathrm{p} = 0.5$. The graph shows that there has been a gradual decrease of the ground state population with increasing intensity of the incident laser. There is a significant difference in the population variation of excited and continuum states compared with changing of the ground state. When $I_{\mathrm{laser}} < 4.5$, the populations of the excited and continuum states are quickly increased with increasing laser intensity. When $I_{\mathrm{laser}} > 4.5$, the ionization yield continues to enhance except for a slight decrease near the $I_{\mathrm{laser}} = 5.2$, however there is a stepped structure in the variation of the excited state population, and the step height is reduced with the increase of the laser intensity.

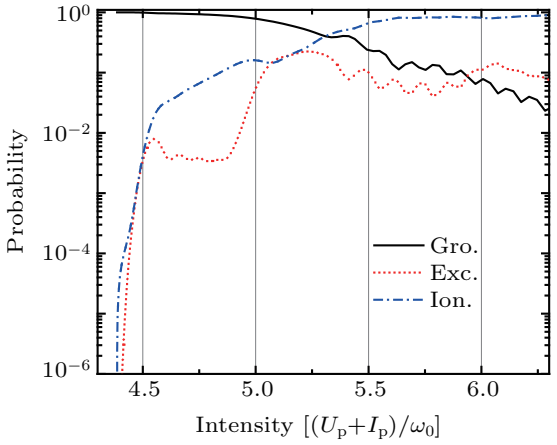


Fig. 1. (color online) The variation of the population of the ground state (black solid line), excited states (red dotted line), and continuum states (blue dashed–dotted line) with the laser intensity in units with $(U_\mathrm{p}+I_\mathrm{p})/\omega_0$.

The ionization of the atom can be further analyzed through its photoelectron spectra. The variation of the photoelectron emission with the laser intensity is presented in Fig. 2. Here, the energy unit of the photoelectron is used $(\text{energy} + I_\text{p} + U_\text{p})/\omega_0$. Under this unit, the energy of the ATI peaks in photoelectron is almost same with the change of laser intensity. The number in the energy is almost the same with change of the laser intensity, and the number in energy equals to the absorbed photon one for the ATI peak. It is noted that there is a clear interference pattern in the ATI peak of photoelectron spectra with some intensities. Furthermore,the energy position of the interference pattern is proportional to the increase of the laser intensity. The low order ATI peak has greater depth of modulation than the high order one, and the first interference appears the laser intensity near the $I_\text{laser} = 5.2$.

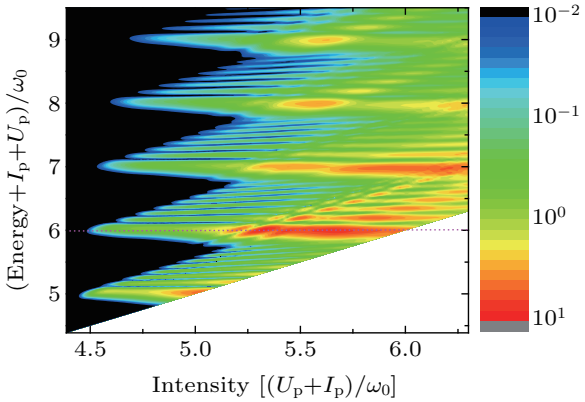


Fig. 2. (color online) Change of photoelectron emission with the laser intensity.

Not only the photoelectron emission is calculated from the time-dependent wavefunction, but also the harmonic spectra are obtained. In Fig. 3, the variation of HHG with the incident laser intensity is shown. Due to the atom symmetry, only odd harmonic should be emitted. Nevertheless, in Fig. 3, in addition to odd harmonics, the radiation with other frequency also can be observed in some laser intensity. As is well known, the intensity of high harmonic generation is proportional to the product of the populations of the ground state and the continuum state. Along with the increase of the laser intensity, this product will continue to increase until the population of the ground state is depleted. In the following, taking the 5th harmonic as an example, we examine the dependence of the HHG efficiency on the laser intensity. From Fig. 3, with the increase of the laser intensity, one can observe that the intensity of the 5th harmonic appears a peak at $I_\text{laser} = 4.8$, and declines until $I_\text{laser} = 5.2$, then quickly increases above $I_\text{laser} = 5.2$. In general, when an atom is irradiated by the strong laser field, there may be a competitive relationship between ionization and excitation. Thereby, in order to explain the decrease of the 5th harmonic intensity about $I_\text{laser} = 5$, it is necessary to investigate the behaviors of the excited state population to understand the variation of the low order harmonic and ATI with the laser intensity.

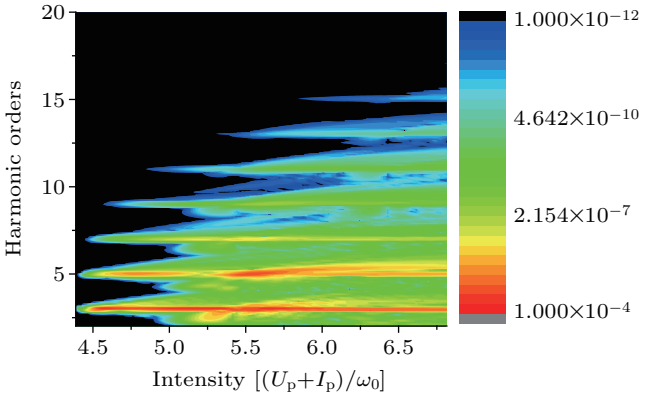


Fig. 3. (color online) Variation of HHG with the intensity of the incident laser.

In Figs. 4(a)–4(c), the change of HHG, photoelectron spectra and the population of the excited states with the laser intensity are presented. Here, the harmonics near the 5th order as example are shown in Fig. 4(c). For $I_\text{laser} < 5$, the energy of the photoelectron peak gradually declines to zero with the increase of laser intensity, which can be ascribed to the stark effect in the continuum state, and only the 5th order harmonic appears at the HHG spectrum. When the intensity $I_\text{laser} > 5$, the harmonic and photoelectron spectra become complex. For example, when $I_\text{laser} = 5.2$, there are clear interference structures in the first main ATI peak from the photoelectron spectrum, especially a significant drop for the intensity of the 5th harmonic and another emission peak near the frequency $\omega = 4.2$ can be found at the harmonic spectrum. From Fig. 4(b), one can observe that, the population of the high excited states near the $I_\text{laser} = 5.2$ is lager than the case of $I_\text{laser} < 5$. It means that there exists a clear corresponding relation between the low order harmonic and the photoelectron emission, and the high excited states play an important role for this relation.

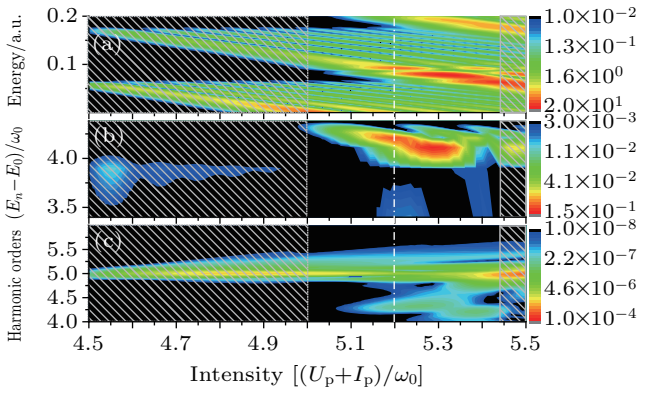


Fig. 4. (color online) The variation of (a) the photoelectron spectra, (b) the excited states population, and (c) high harmonic spectra with the intensity of the incident laser pulse. The unit a.u. is short for arbitrary units.

The corresponding relation between the harmonic and photoelectron spectra can be well understood in Fig. 5. In Fig. 5, the upward tilt solid lines represent the energy shift of the excited state caused by the AC stark effect of the laser field. When the atom is irradiated by the laser field with the low intensity, there is a slight change on the eigenenergy of the atom. Therefore, it is necessary to absorb five photons for ionization of atom ($I_p/\omega_0 = 4.39$). And the photon energy of the fifth harmonic is above the ionization threshold. With the increase of the laser intensity, the ionization threshold of an atom will rise. When the laser intensity $I_{laser} = 5$, six photons are necessary for the atomic ionization, and the first ATI peak disappears. The disappearance of the first ATI peak can be attributed to the channel closing. Therefore, there has been a slight fall of the ionization probability in Fig. 1, which leads to the decrease of the 5th order harmonic, as shown in Fig. 4(c). It is highly consistent with a consequence of channel closing in reference.[42,43] When the intensity of the incident laser is further increased, the energy level of the excited state moves upward, and the five photon resonance between the high excited state the ground one occurs, which causes a sharp enhancement of the population at the excited state, as shown in Fig. 4(b). The ionization from these high excited states interferences with the ionization from the ground state. From Fig. 4(a) one can clearly observe the interference pattern in the photoelectron spectrum. Due to the large population of the highly excited states, the transition probability between the ground state and highly excited states is high. Therefore, the harmonic emission whith the transition frequency about $4.2\omega_0$ can be found in Fig. 4(c). For the higher laser intensity, the energy of the fifth harmonic is lower than the real ionization threshold, and there is the off-resonant between the excited state and the ground state. Hence, the corresponding efficiency of this harmonic is proportional to the laser intensity I_{laser}.

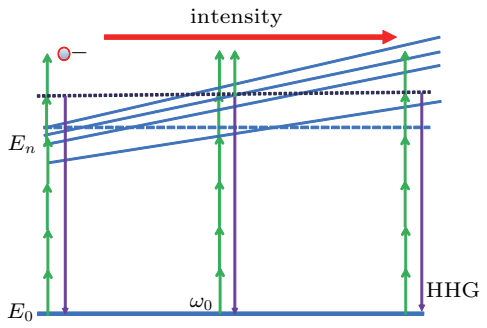


Fig. 5. (color online) Mechanism of generation for the low order harmonic and photoelectron emission.

As discussed above, the excitation of atom plays an important role in the HHG and photoelectron emission. Because the population of the excitation is sensitive to the duration of the laser pulse, which will affect the relation between the harmonic and photoelectron spectra. In Fig. 6, the duration effect on the low order harmonic and photoelectron is presented. Here, the laser's intensity is $I_{laser} = 5.25$ (1.78×10^{14} W/cm^2) and its duration is from 5 optical cycles to 40 optical cycles. From this figure, it is apparent that the duration has a strong effect on the resonance induced harmonic and photoelectron emission. Comparing with the long laser pulse, the frequency width of a short laser pulse is larger, the corresponding population of the excited state is relatively small. The intensity reduction of the 5th order harmonic and the enhanced interference of the low photoelectron has become pronounced.

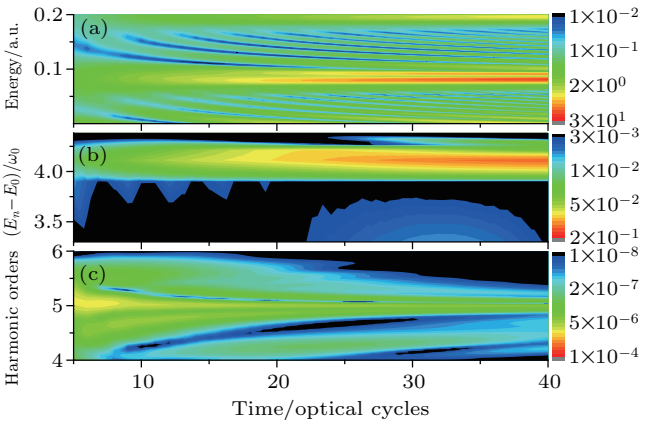


Fig. 6. (color online) The variation of (a) the photoelectron spectra, (b) the excited states population, and (c) high harmonic spectra with the duration of the incident laser pulse with intensity 1.78×10^{14} W/cm^2.

4. Conclusion

In summary, the low order harmonic and photoelectron spectra of an atom irradiated by short laser pulse are systematically investigated by solving the time-dependent Schrödinger equation. When the laser's frequency is not resonant with the energy difference between the excited state and the ground one, the intensities of the low harmonic and photoelectron are enhanced with the increase of the laser intensity. When the resonance occurs, the intensity of low harmonic is reduced and the obvious interference in the photoelectron emission is observed.

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