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To cite this article: Shian Zhang *et al* 2010 *J. Phys. B: At. Mol. Opt. Phys.* **43** 135401

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# Coherent phase control of (2+1) resonantly enhanced multiphoton ionization photoelectron spectroscopy

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Received 6 January 2010, in final form 18 May 2010

Published 14 June 2010

Online at [stacks.iop.org/JPhysB/43/135401](http://stacks.iop.org/JPhysB/43/135401)

## Abstract

In this work, we report a theoretical study of the coherent phase control of typical (2+1) resonantly enhanced multiphoton ionization photoelectron spectroscopy (REMPI-PES) in an atomic system. By properly designing the spectral phase of a femtosecond pulse, we can realize the manipulation of the photoelectron energy, the photoelectron spectral bandwidth and the photoelectron intensity at a certain kinetic energy. Moreover, we can also obtain a high-resolution photoelectron spectrum and fine energy-level structure of the excited states for a complicated quantum system in spite of the broad-width spectrum of the femtosecond pulse. The theoretical results have promising applications for investigating the molecular structure and understanding the laser-induced ionization dynamics.

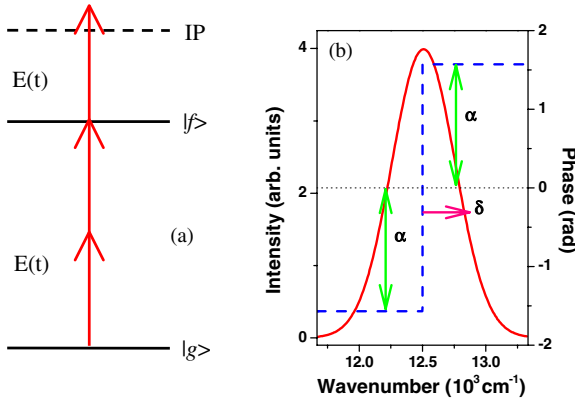
(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

With the advent of the ultrashort pulse shaping technique, an ultrashort pulse has been widely employed to control atomic and molecular dynamics instead of monitoring the dynamical processes, as previously. Recently, quantum coherent control using the ultrashort pulse shaping technique has attracted considerable interest due to its ability to steer a quantum system from an initial quantum state to a pre-selected target state by the light-matter interaction [1–10]. Now it is possible to obtain such a pulse with arbitrary temporal distribution by manipulating the spectral phase and/or amplitude in the frequency domain. In a simple quantum system or simple excitation process, the pre-designed pulse can meet the experimental requirement [1–5]. For a more complicated case, adaptive feedback control based on a learning algorithm is an excellent method to automatically optimize the light field without any information about the quantum system and to achieve the desired outcomes [6–10].

Resonantly enhanced multiphoton ionization photoelectron spectroscopy (REMPI-PES) has proven to be a powerful tool for characterizing excited states and understanding the dynamical ionization process [11–16].

Recently, the coherent control of REMPI-PES induced by a shaped femtosecond pulse with simple spectral phase modulation has been widely investigated [17–22]. These studies were focused on single-photon absorption or resonant multiphoton absorption (i.e. a transition with the intermediate states) from the ground state to the final excited state. However, for the REMPI process with infrared pulse excitation, the population in the ground state is usually pumped to the final excited state by nonresonant multiphoton absorption (i.e. a transition without intermediate states) and then ionized. To our knowledge, reports on the manipulation of the REMPI-PES in the case of nonresonant multiphoton absorption are still scarce. In the present work, we theoretically investigate the coherent phase control of typical (2+1) REMPI-PES in an atomic system. It is necessary to study the model system in the control of photon-induced atomic or molecular dynamics, where the physical dynamical process can be analysed on the basis of theoretical evaluation, and thus more complicated quantum systems may become controllable and understandable. Our theoretical results show that by rationally manipulating the spectral phase of a femtosecond pulse, the photoelectron energy (i.e. multiphoton ionization efficiency),



**Figure 1.** (a) Schematic diagram of energy levels for the typical (2+1) resonantly enhanced multiphoton ionization. Here,  $|g\rangle \rightarrow |f\rangle$  is coupled by the laser field  $E(t)$  with two-photon excitation, and the population in the state  $|f\rangle$  is ionized by one-photon excitation of  $E(t)$ . (b) Schematic diagram of the phase step applied on the laser spectrum, where  $\delta$  and  $\alpha$  represent the phase step position and phase modulation depth, respectively.

the photoelectron spectral bandwidth and photoelectron intensity at a certain kinetic energy can be manipulated, and a high-resolution photoelectron spectrum and an energy-level diagram of a complicated quantum system can also be obtained.

## 2. Theoretical model

We consider the weak interaction of a laser field  $E(t)$  with a two-level atomic system, as shown in figure 1(a). Here  $|g\rangle$  and  $|f\rangle$  are the ground and final excited states, respectively.  $|g\rangle \rightarrow |f\rangle$  is coupled by the laser field  $E(t)$  with two-photon excitation, and the population of the excited state  $|f\rangle$  is ionized by one-photon excitation. We assume that the laser pulse is much shorter than the lifetime of the excited state and initially only the ground state  $|g\rangle$  is populated. The photoelectron spectrum  $P(E_v)$  by one-photon ionization from the final excited state  $|f\rangle$  can be written as [19–23]

$$P(E_v) \propto \int_{-\infty}^{+\infty} E(t) a_f(t) \exp[i(E_v + E_I - E_f)t/\hbar] dt, \quad (1)$$

where  $E_I$  is the ionization energy from the ground state  $|g\rangle$ ,  $E_f$  is the eigenenergy of the final excited state  $|f\rangle$ , and  $a_f(t)$  is the time-dependent probability amplitude in the final excited state  $|f\rangle$ , and given by [24]

$$a_f(t) \propto \int_{-\infty}^t E^2(t) \exp(i\omega_0 t) dt, \quad (2)$$

where  $\omega_0$  is the transition frequency from the ground state  $|g\rangle$  to the excited state  $|f\rangle$ .

As can be seen from equation (1), the photoelectron spectral structure  $P(E_v)$  is correlated with the time-dependent population in the final excited state  $|f\rangle$ , and therefore the population variation in the excited state  $|f\rangle$  will directly affect the photoelectron spectral structure. A previous study demonstrated that by  $\pi$  phase step modulation, the nonresonant two-photon transition probability could be completely eliminated [24, 25]. Here, we also employ the

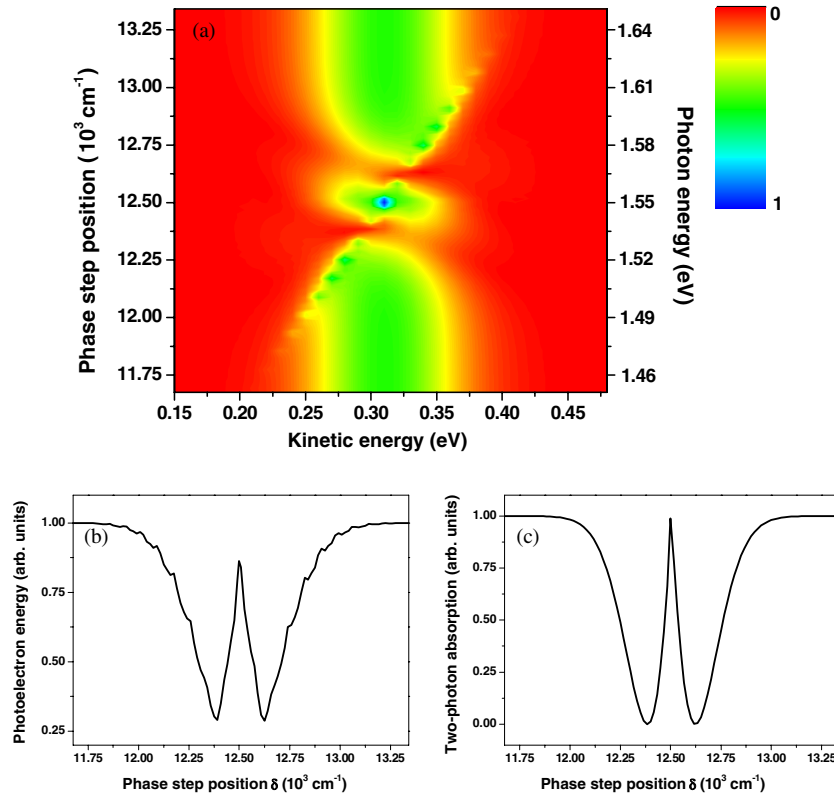
$\pi$  spectral phase step to control the population in the final excited state  $|f\rangle$  and consequently realize the manipulation of (2+1) REMPI-PES. The spectral modulation is shown in figure 1(b), and  $\delta$  and  $\alpha$  represent the phase step position and phase modulation depth, respectively.

## 3. Results and discussion

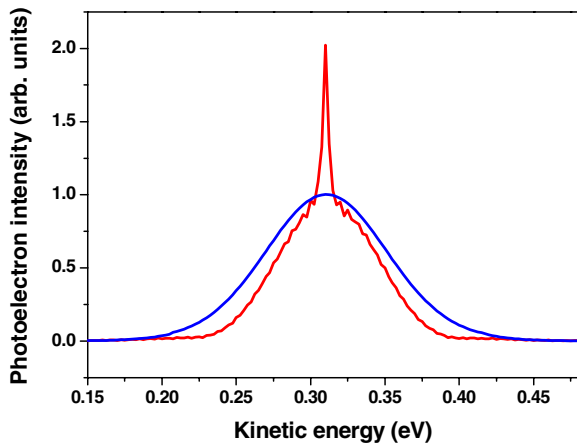
We assume that the laser pulse has a Gaussian profile with a centre frequency of  $12\,500\text{ cm}^{-1}$  and a spectral bandwidth (a full width at half maximum (FWHM)) of  $500\text{ cm}^{-1}$ , a transition frequency from the ground state  $|g\rangle$  to the final excited state  $|f\rangle$  of  $25\,000\text{ cm}^{-1}$  and an ionization energy from the ground state of  $4.34\text{ eV}$ , corresponding to a frequency of  $35\,005\text{ cm}^{-1}$ . Figures 2(a) and (b) show the photoelectron spectra and the photoelectron energy as functions of the phase step position  $\delta$  for  $\alpha = 0.5\pi$ . Here, the photoelectron energy is obtained from the photoelectron spectrum according to  $E = \int_{-\omega}^{+\infty} P(E_v) dE_v$ . As can be seen from figure 2(b), the photoelectron energy can be suppressed by about 85% at about  $12\,375\text{ cm}^{-1}$  and  $12\,625\text{ cm}^{-1}$ , which indicates that the multiphoton ionization efficiency can be effectively controlled by simple spectral modulation. Equation (1) shows that the photoelectron spectrum relies on the time-dependent population in the final excited state  $|f\rangle$ . For comparison, the two-photon transition probability in the final excited state  $|f\rangle$  is calculated according to  $S_2 = |a_f(t \rightarrow \infty)|^2$  and presented in figure 2(c). The two-photon transition probability is completely eliminated at  $\sim 12\,375\text{ cm}^{-1}$  and  $12\,625\text{ cm}^{-1}$ , and the phenomenon has been experimentally demonstrated [24, 25]. It can be found that, by the  $\pi$  phase step modulation, the photoelectron energy has the same evolution as the two-photon transition probability, but the photoelectron energy is not completely eliminated. We believe that this is due to the time-dependent absorption of the excited state  $|f\rangle$  in the ionization process since the absorption and ionization processes employ the same laser field  $E(t)$  [26].

It can also be seen from figure 2(a) that the photoelectron intensities at different kinetic energies achieve maximal enhancement at different phase step positions, while the energy difference between the photon energy at the phase step position and the kinetic energy is constant, which is equal to the ionization energy from the excited state  $|f\rangle$ ,  $E_\Delta$  (i.e.  $E_\Delta = E_I - E_f$ ). So, we can deduce that the photoelectron intensity is maximally enhanced at the kinetic energy corresponding to  $E_\delta - E_\Delta$ ; here  $E_\delta$  is the photon energy at the phase step position. Figure 3 shows the photoelectron spectra induced by the transform limited pulse (blue line) and the shaped pulse with the  $\pi$  phase step at  $12\,500\text{ cm}^{-1}$  (red line). As can be seen, the photoelectron spectrum is narrowed and photoelectron intensity at the kinetic energy of  $0.31\text{ eV}$  is effectively enhanced for the shaped pulse.

If the shaped pulse has an antisymmetric spectral phase distribution around the two-photon transition frequency  $\omega_0/2$ , the two-photon transition probability is independent of the spectral phase and consistent with that of the transform-limited pulse [24]. Next we demonstrate that although the population in the excited state  $|f\rangle$  is invariable, the



**Figure 2.** Simulated photoelectron spectra (a), photoelectron energy (b) and two-photon transition probability in the final excited state  $|f\rangle$  (c) as a function of the phase step position  $\delta$  for  $\alpha = 0.5\pi$ .



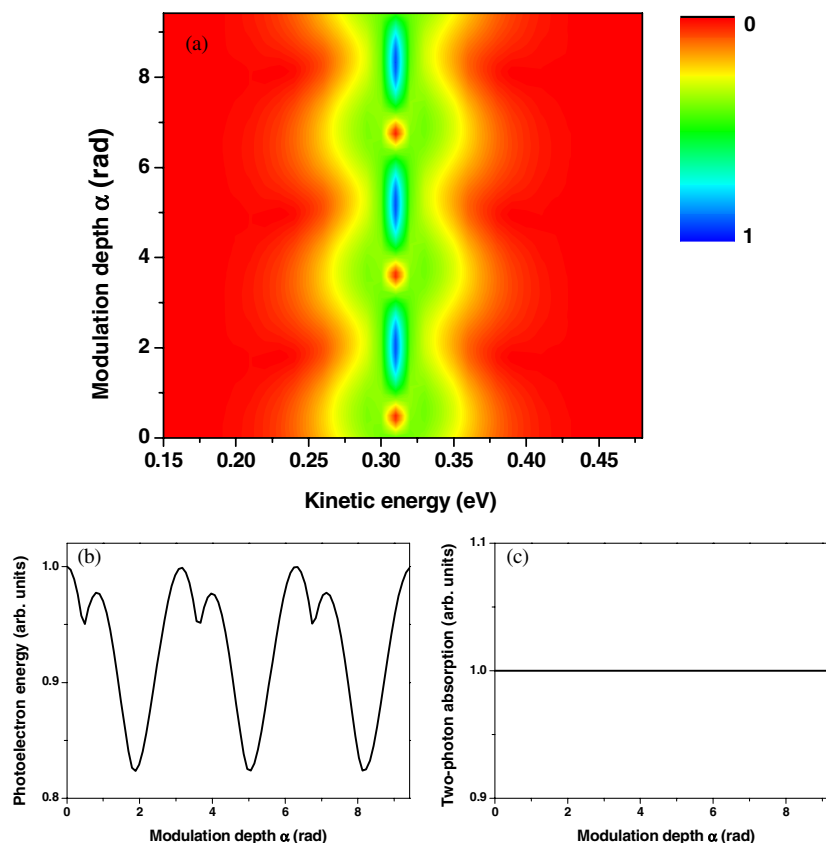
**Figure 3.** Photoelectron spectra induced by the transform limited pulse (blue line) and the shaped pulse with the  $\pi$  phase step at  $12\,500\text{ cm}^{-1}$  (red line). The photoelectron spectrum is narrowed and the photoelectron intensity at a kinetic energy of  $0.31\text{ eV}$  is enhanced for the shaped pulse.

photoelectron spectroscopy can still be manipulated. The photoelectron spectra, photoelectron energy and two-photon transition probability in the excited state  $|f\rangle$  as a function of the modulation depth  $\alpha$  for  $\delta = 12\,500\text{ cm}^{-1}$  are calculated and presented in figure 4. With the increase of the modulation depth, the photoelectron spectra and photoelectron energy are periodically modulated, but the two-photon transition probability remains constant. Interestingly, the photoelectron

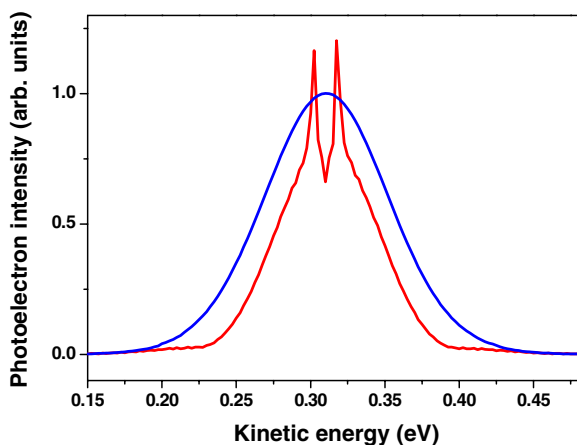
spectrum can be broadened or narrowed, and photoelectron intensity at the kinetic energy of  $0.31\text{ eV}$  can also be enhanced or attenuated. As can be seen, the antisymmetric spectral phase does not affect the population in the excited state  $|f\rangle$ , but it does affect the photoelectron spectral structure and photoelectron energy.

For the femtosecond pulse excitation, several excited states falling within the broad spectrum can be simultaneously populated and ionized, and this may result in their photoelectron spectra being indistinguishable. Since the photoelectron intensity enhancement for the (2+1) REMPI is obtained at the kinetic energy corresponding to  $E_\delta - E_\Delta$ , the photoelectron spectra between the neighbouring excited states can be differentiated by the  $\pi$  phase step modulation. We consider a quantum system including two excited states with transition frequencies of  $25\,065\text{ cm}^{-1}$  and  $24\,935\text{ cm}^{-1}$ , and the separation between the two excited states is  $130\text{ cm}^{-1}$ . The photoelectron spectra induced by the transform-limited pulse (blue line) and the shaped pulse with the  $\pi$  phase step at  $12\,500\text{ cm}^{-1}$  (red line) are presented in figure 5. Two distinct peaks are observed for the shaped pulse, which are related to the two excited states. However, only a single broad peak is obtained for the transform-limited pulse.

Similarly, the energy level diagram for a complicated quantum system can also be labelled by the  $\pi$  phase step modulation according to the kinetic energies of the two peaks, photon energy at the phase step position and the ionization energy. We still employ the above quantum system with two excited states and the calculated results. As can be seen from



**Figure 4.** Simulated photoelectron spectra (a), photoelectron energy (b) and two-photon transition probability in the final excited state  $|f\rangle$  (c) as a function of the modulation depth  $\alpha$  for  $\delta = 12\,500\text{ cm}^{-1}$ .



**Figure 5.** Photoelectron spectra of a quantum system with two excited states induced by the transform limited pulse (blue line) and the shaped pulse with the  $\pi$  phase step at  $12\,500\text{ cm}^{-1}$  (red line). Two distinct peaks are observed for the shaped pulse, which are related to the two excited states.

figure 5, the kinetic energies of the two peaks are 0.302 eV and 0.318 eV, respectively. According to the above expression, the ionization energy from the ground state is 4.34 eV and the photon energy at the  $\pi$  phase step position of  $12\,500\text{ cm}^{-1}$  is 1.55 eV. Thus the eigenenergies of the two excited states can be calculated as 3.108 eV and 3.092 eV, corresponding to the frequencies  $25\,065\text{ cm}^{-1}$  and  $24\,935\text{ cm}^{-1}$ , which are

equal to the transition frequencies of the two excited states described above. It can be concluded that, by the  $\pi$  phase step modulation, the fine energy-level structure of the excited states can be labelled, and therefore this can provide a feasible method to investigate the atomic or molecular structure in spite of the broad spectrum of the femtosecond pulse.

#### 4. Conclusion

In summary, we have theoretically demonstrated that by rational phase modulation, photoelectron spectroscopy through (2+1) REMPI can be expediently manipulated. The photoelectron energy can be effectively controlled, the photoelectron spectrum can be narrowed or broadened and the photoelectron intensity at a certain kinetic energy can also be enhanced or attenuated. Furthermore, the high-resolution photoelectron spectrum and fine energy-level structure of the excited states for a complicated quantum system can also be achieved. Our research provides a theoretical basis for experimental investigation and has the potential application for the control of various REMPI processes.

#### Acknowledgment

This work was supported by Program for Changjiang Scholars and Innovative Research Team in University (PCSIRT), Shanghai Leading Academic Discipline Project

(B408), National Key Project for Basic Research of China (2006CB806006 and 2006CB921105), Ministry of Education of China (30800) and Shanghai Municipal Science and Technology Commission (09142200501, 09ZR1409300 and 10XD1401800).

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