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Mechanisms of infrared-laser-assisted atomic ionization by attosecond pulses

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We propose a mechanism to understand the infrared (IR) laser assisted atomic ionization by attosecond pulses (AP). Atomic structures in an IR laser field are described by Floquet states and atoms can be ionized to a Floquet state by a single AP through different Floquet components. The interference of ionization through different Floquet components results in the oscillation of the ionization yield as a function of the arriving time of the AP. The proposed mechanism explains the recent experimental observations [Johnsson *et al.,* Phys. Rev. Lett. **99**, 233001 (2007)]. Furthermore, we find that, for a specified photoelectron energy, the ionization yield always oscillates as a function of the relative phase between the AP and the IR laser for both He and Ar atoms.

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Photoionization is an important process used to investigate atomic or molecular structures. Each atom or molecule has a unique structure much like a fingerprint and we can identify its constituents by measuring its photoabsorption spectrum. With the advances of laser and synchrotron radiation technologies, one can not only measure but also modify the photoabsorption spectra by merging an X-ray beam with a femtosecond intense laser beam [1,2]. However, due to the lack of a finite phase relationship between the synchrotron X-ray and the femtosecond laser, the ionization process cannot be fully controlled in a coherent way. Johnsson *et al.* [3,4] investigated photoionization of He by an attosecond pulse train (APT) [5], a coherent extreme ultraviolet (euv) light source, merged with an infrared (IR) laser. Since the phase between the APT and the IR field can be controlled by the time delay between the APT and IR laser, the ionization probability can be manipulated in a coherent way. The observed ionization yield was found to oscillate twice per IR optical cycle as a function of the delay between the APT and IR fields. They attributed the oscillation to the interference between electron wave packets created by the different attosecond pulses (AP) in the train. Recently Ranitovic *et al.* [6] found that the IR assisted He photoionization yields by an APT are also sensitive to the wavelength of the IR laser.

Because single attosecond pulses (SAP's) are already available in several laboratories [7–10], we investigate the IR assisted atomic ionization by both an SAP and an APT for both He and Ar atoms. Our calculated total ionization yields by the APT are consistent with the available experimental measurements [4,6]. By analyzing the energy distribution of the ejected electrons in the region where the IR and SAP (or APT) temporally overlap, we find a strong oscillation exists even for the SAP ionization. Since the IR pulse duration is much longer than that of the SAP or APT, we can treat it as an infinitely long pulse and use Floquet theory [11] to explain the dynamics involved in the system. Since we use a relatively weak IR laser, we can treat the ground state as unperturbed. Each excited state and the continuum form Floquet states, with each Floquet state made up of many sidebands separated by one IR photon energy. The SAP can excite atoms from the ground state to different sidebands of any Floquet state coherently and the interference between the transitions through different sidebands results in the oscillation of the ionization yield. The SAP has a very broad energy distribution and it can excite or ionize atoms from the ground state to many different Floquet states. There is no phase relation between the different Floquet states so the ionization amplitudes through different states add up incoherently and this results in a small oscillation for the total ionization yield. On the other hand, when we use an APT, due to the sharp energy resolution we typically select a single dominant Floquet state, which results in a large oscillation. Based on this explanation, we should also expect to observe a large oscillation when an SAP is used if, instead of selecting a narrow band of euv energies, we select a narrow energy range in the final electron energy.

We investigate theoretically the ionization process of He and Ar by an IR laser combined with an APT or SAP by solving the time-dependent Schrödinger equation. The time-dependent field strength $E_x(t)$ of the SAP is of the form (atomic units, $m = \hbar = e = 1$, are used hereafter unless otherwise stated),

$$E_x(t) = F_x e^{-2\ln 2\, t^2/\tau_x^2} \cos(\omega_x t), \tag{1}$$

with F_x the peak field strength of the SAP and $\omega_x = 23.0$ eV the center energy of the SAP and $\tau_x = 0.3$ fs the full width at half maximum (FWHM) of the SAP. The field strength of the APT can be expressed as

$$E_{\rm apt}(t) = \sum_j (-1)^j E_x(t - jT_h) e^{-2\ln t^2/\tau_T^2}, \tag{2}$$

with T_h the time interval between the AP's within the APT and $\tau_T = 10$ fs the FWHM of the APT.

The IR field strength is

$$E_{\rm IR}(t) = F_{\rm IR} e^{-2\ln 2\,(t-t_d)^2/\tau^2} \cos[\omega(t - t_d)]. \tag{3}$$

Here, $F_{\rm IR}$ is the field strength of the laser, $\tau = 45$ fs is the FWHM of the IR pulse, ω is the center frequency of the IR pulse (here we choose the laser wavelength as 796 nm), and t_d

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is the time delay between the IR pulse and APT (or SAP). The Hamiltonian of the system is written as

$$H(t) = \left(-\frac{\nabla^2}{2} + V(r)\right) - \mathbf{rE}(t) = H_0 + V^{\text{ext}}(t). \quad (4)$$

Here H_0 is the Hamiltonian with the model potential $V(r)$ [12] and $V^{\text{ext}}(t) = -z[E_{\text{IR}}(t) + E_x(t)]$ is the interaction of the electron with the external fields under the assumption that all the external fields are polarized along the z direction. The time evolution of the ground-state wave function Ψ_g can be expressed as

$$|\Psi(t)\rangle = -i \int_{-\infty}^{t} e^{-i\int_{t'}^{t} H(t'')dt''} V^{\text{ext}}(t') e^{-iH_0 t'} |\Psi_g\rangle dt' + e^{-iH_0 t}|\Psi_g\rangle. \quad (5)$$

We discretize the space in the pseudospectral grid and propagate the wave function by the split-operator method in the energy representation [13]. The numerical simulations are performed in a finite box. To avoid the unphysical reflection at the boundary, we separate the space into two regions within the box, the inner region and the outer region. When the time-dependent wave function in space reaches the outer region, we project the outer region wave function into momentum space (Volkov state) and remove it from the wave function in real space. This procedure allows us to keep all the phase information for a long time propagation without the reflection from the boundary. The detail of the numerical procedure can be found in our previous article [14]. When the pulses are over, the total wave function is separated into two parts. One is the wave function in momentum space (or the outer region), which describes the ionization process and the other is located in the inner region in real space and describes the excitation process. The second term of Eq. (5) does not contribute to any dynamical process. To improve the numerical accuracy we remove this part in the calculation. In the following calculations, we choose the IR laser intensity as 5×10^{12} W/cm^2 and the AP pulse peak intensity as 3.5×10^{12} W/cm^2. The ionization probabilities depend on the AP intensity linearly up to 10^{14} W/cm^2 so that the choice of AP intensity is not important as long as the AP intensity is less than 10^{14} W/cm^2. The time interval T_h between the AP's in the APT is chosen as a half optical cycle of the IR laser. To illustrate the mechanism, we also investigate the ionization probabilities by a combined field

$$E_{H13+H15}(t) = F_x[\cos(13\omega t) + \cos(15\omega t)]e^{-2\ln 2t^2/\tau_T^2}, \quad (6)$$

of the 13th and 15th harmonics (noted by $H13 + H15$) with an IR laser.

Figure 1 shows the total ionization probabilities of He and Ar atoms by the SAP or APT in the presence of the IR laser as a function of the time delay between the SAP (or APT) and the IR laser. We see that there is a significant oscillation for He ionized by the APT and there is almost no oscillation for Ar ionized by the APT. If we replace the APT with a SAP, there are almost no oscillations for either He or Ar. All these observations are consistent with the experimental results for the APT [4] and their simulations for the SAP.

Why is there a large oscillation for He with the APT but not for the SAP? To answer this question, we show the ejected

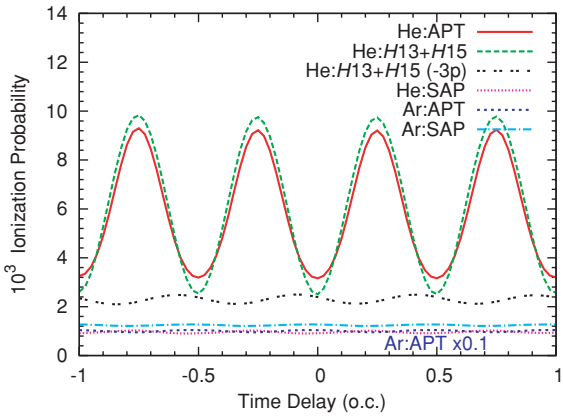

FIG. 1. (Color online) The ionization probabilities of He and Ar by the APT and the SAP in the presence of the IR field as a function of the time delay between the SAP (or APT) and the IR laser. The ionization probabilities of He by the $H13 + H15$ field, the combined field of harmonics 13 and 15 (dashed line) and without the $3p$ state (double dotted line) are also presented; o.c. denotes optical cycle.

electron energy and time delay distributions by the APT and SAP in Fig. 2 [the color bars in Figs. 2 through 4 indicate the relative strength.] For the APT case, the ejected electron shows a clear above-threshold-ionization (ATI) structure, with many sharp peaks separated by one IR photon energy. For a given ATI peak, the yield oscillates twice per optical cycle. For the SAP case, there is a continuum of ATI energies because

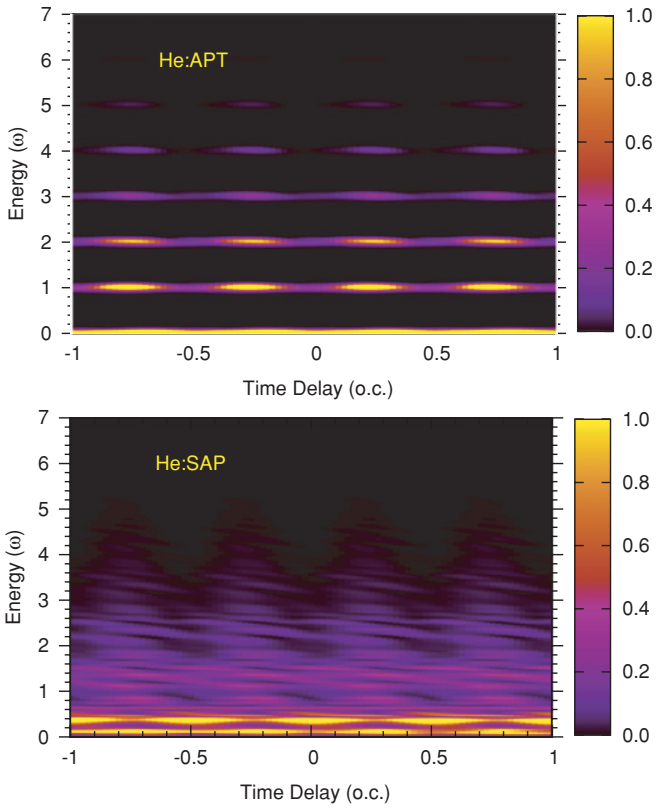

FIG. 2. (Color online) The delay time and energy distributions of the ejected electron of He ionized by the APT (upper panel) and the SAP (lower panel).

of the broad energy distribution of the harmonics. If we look at a particular ATI energy, the yield also oscillates twice per optical cycle with the amplitude comparable to the APT case, but because the oscillations have different phases for different ATI energies, the sum no longer oscillates.

Since the IR pulse duration is longer than that of the APT or AP, the atomic excited and continuum states in the IR laser field become dressed states, or more specifically, Floquet states. A Floquet state Ψ_α can be expressed, as a function of time, as [11]

$$\Psi_\alpha(t) = e^{-i\epsilon_\alpha t} \sum_n e^{-in\omega t} \psi_{\alpha,n}, \tag{7}$$

with ϵ_α the quasienergy, n the sideband index, and $\psi_{\alpha,n}$ the time-independent wave function of the nth sideband. The euv comes from the high-order-harmonic generation by the same laser source and it covers the energy range from the 11th to 17th harmonics of the IR laser [4,6]. The euv can excite He to any of several Floquet states and for each of these, through any of several sidebands. The relative importance of these amplitudes is very sensitive to resonances between the euv energy and the location of the sidebands. For any particular Floquet state, more than one sideband can contribute. For example, if we excite He at time t_0 from the ground state, to the nth sideband or mth sideband of a particular Floquet state, the relative phase difference between the two amplitudes is $(n-m)\omega t_0$. Due to parity conservation, the leading term is $|n-m| = 2$ and thus the amplitude for this process should show an oscillation twice per optical cycle. To confirm this argument, we also present the ionization probabilities by the $H13 + H15$ field and they agree with that of the APT as shown in Fig. 1 with a scaling factor. In the case of the SAP, the bandwidth of the euv is so large that there are many Floquet states created and the oscillating amplitude of the total ionization yield is suppressed. When using an APT, we select Floquet states or one dominant Floquet state within a narrow energy range, which results in a large oscillating amplitude. Note that the present calculations are equivalent to the calculation of the photoabsorption assisted by periodic fields [15]. Here we focus on the ejected electron energy distribution other than the photoabsorption cross sections.

To support this explanation, we enlarge the energy distributions of ATI energies around 2ω in Fig. 3. For the energy distribution of the SAP, we multiply an energy filter function $\{F(E) = \exp[-200 \times (E - 2.02\omega)^2]\}$ to the ATI spectra in Fig. 2 to select the Floquet states around 2ω as shown in the lower panel of Fig. 3. With this manipulation, the distributions from the APT and SAP are close to each other and the oscillating amplitudes of the ionization yields from SAP and APT are comparable to each other.

Why does Ar differ from He in showing no significant oscillation for either the SAP or APT? The center energy of the APT or SAP is 23.0 eV, below the He ionization threshold and above the Ar ionization threshold. So the Floquet states involved in He ionization are reached through (quasiresonant) sidebands of atomic bound states. In this case, there can be one dominant Floquet state and we can observe

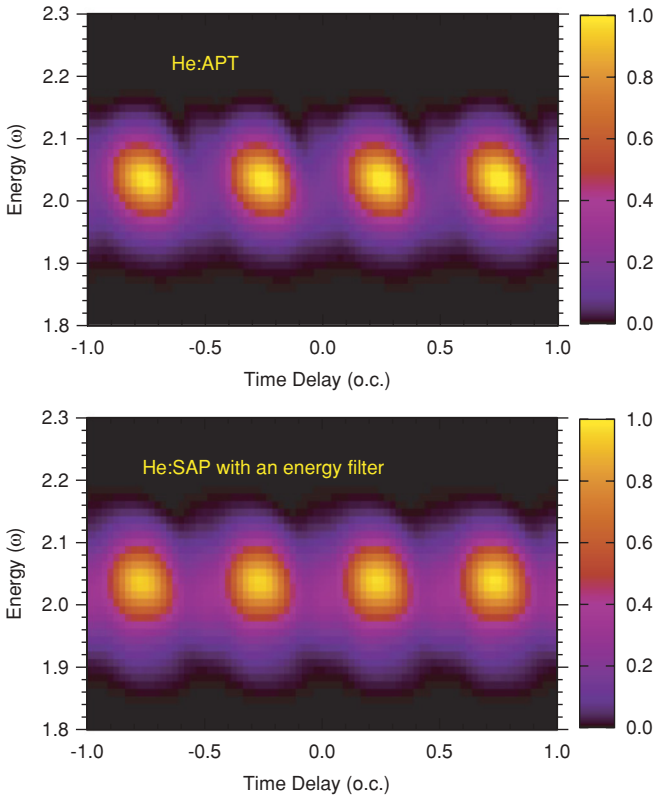

FIG. 3. (Color online) The enlargement of Fig. 2 in a narrow energy range. For the SAT case, we multiply a filter function to select the Floquet states around 2ω in the lower panel.

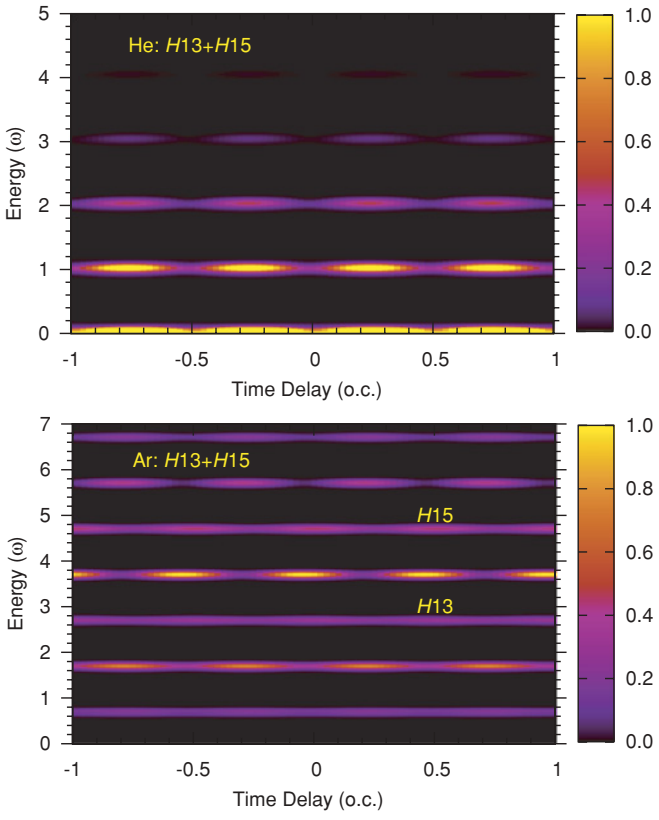

FIG. 4. (Color online) The energy distributions of the ejected electrons as a function of time delay for He (upper panel) and Ar (lower panel) atoms ionized by $H13 + H15$ field with an IR laser.

oscillation not only for a single electron energy, but also in the total ionization yield. Since we use a split-operator method in the energy representation [13] to propagate the wave function, we can remove an arbitrary atomic bound state in the calculation to identify its contribution. We identified that the major contributor for He by the $H13 + H15$ field is the $3p$ state as shown in Fig. 1 by comparing the spectra with and without the $3p$ state. For the Ar case, the transitions are through sidebands of the continuum. Figure 4 shows the IR laser assisted ionization of He (upper panel) and Ar (lower panel) by the $H13 + H15$ field. For the He case, the results are very similar to that by the APT. All the ATI peaks oscillate in the same pattern, which indicates that there is one dominant Floquet state. For the Ar case, the oscillating amplitude of a specified ATI peak is comparable to that of He atoms. However, the oscillation in the total ionization yield is suppressed because different Floquet states oscillate with different phases.

To summarize, we studied the mechanism of atomic ionization by AP's in the presence of a moderately intense IR laser by solving the time-dependent Schrödinger equation. We explain the results using a Floquet picture. The IR field produces dressed atomic states (or Floquet states), each of which is described by a quasienergy with many sideband energies separated by one IR photon energy. A transition from the ground state to a Floquet state via a specified sideband has a well-defined phase and the interference of the transitions to the Floquet state via different sidebands results in the oscillations as a function of the relative time delay between the IR laser and the transition time. This is a very general phenomenon that exists in any situation, such as Aharonov-Bohm oscillations in an H atom in a radiation field [16], when we excite a physical system by a coherent source in the presence of a periodically time-varying field. The oscillation amplitude of the total ionization yield by a SAP is suppressed because many Floquet states are contributing and the oscillations must be added up incoherently. Using an APT, we select particular Floquet states so that the oscillating amplitude is enhanced. Alternatively, selecting the final photoelectron energy will also tend to select a single Floquet state, leading to the oscillation as a very general phenomenon.

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[1] M. P. Hertlein *et al.*, Phys. Rev. A **73**, 062715 (2006).
[2] S. H. Southworth *et al.*, Phys. Rev. A **76**, 043421 (2007).
[3] P. Johnsson *et al.*, Phys. Rev. Lett. **95**, 013001 (2005).
[4] P. Johnsson, J. Mauritsson, T. Remetter, A. L'Huillier, and K. J. Schafer, Phys. Rev. Lett. **99**, 233001 (2007).
[5] P. M. Paul *et al.*, Science **292**, 1689 (2001).
[6] P. Ranitovic *et al.*, New J. Phys. **12**, 013008 (2010).
[7] M. Drescher *et al.*, Science **291**, 1923 (2001).
[8] G. Sansone *et al.*, Science **314**, 443 (2006).
[9] E. Goulielmakis *et al.*, Science **320**, 1614 (2008).
[10] I. Thomann *et al.*, Opt. Express **17**, 4611 (2009).
[11] S. I. Chu and D. A. Telnov, Phys. Rep. **390**, 1 (2004).
[12] X. M. Tong and C. D. Lin, J. Phys. B **38**, 2593 (2005).
[13] X. M. Tong and S. I. Chu, Chem. Phys. **217**, 119 (1997).
[14] X. M. Tong, K. Hino, and N. Toshima, Phys. Rev. A **74**, 031405(R) (2006).
[15] T. Shirahama, X. M. Tong, K. I. Hino, and N. Toshima, Phys. Rev. A **80**, 043414 (2009).
[16] M. Kalinski, Phys. Rev. A **57**, 2239 (1998).