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Angularly resolved electron spectra of H⁻ by few-cycle laser pulses

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We study theoretically the detachment of H^- by a linearly polarized few-cycle laser pulse. We show that the angularly resolved distribution of the electrons is very sensitive to the carrier envelope phase (CEP) and the duration of the short laser pulse. This in turn provides an additional means to measure the CEP of a laser pulse at lower laser intensities.

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The rapid development of modern laser technology has made it possible to generate few-cycle pulses both in the infrared [1] and in the xuv region [2]. A very important feature of these few-cycle pulses is that the carrier envelope phase (CEP) plays a crucial role in the nonlinear interaction with matter, which provides an additional laser parameter to control the relevant physical and chemical processes [3]. In particular, experiments have demonstrated striking CEP dependence in high-order harmonic generation (HHG) [4], multiphoton ionization [5], and multiphoton excitation [6]. Strong CEP dependence has also been found in the nonsequential double ionization of Ar [7] and in the electron localization of dissociating D_2^+ [8]. These phenomena can in turn be used to measure the CEP of the pulse. An accuracy of measurement better than $\pi/10$ has recently been achieved by a stereo detection of the spatial distribution of electrons [9]. More recently, Peng *et al.* [10,11] have investigated the CEP effects of an attosecond pulse on the momentum and energy distributions of the ionized electrons for H and He atoms. They have also studied the influences of an additional short infrared laser pulse on the ionizing dynamics.

There have been recent experiments for H^- in intense laser fields [12,13]. The angular distribution of the detached electron has been shown to be sensitive to the laser intensity and wavelength. Various theories have also shown similar phenomena near the detachment threshold (see e.g., Refs. [14,15]). However, these experiments have been carried out with rather long pulses of duration around a few hundred femtoseconds and most theories are done for a monochromatic laser. Very recently, theoretical calculations have found strong CEP dependence and pulse length dependence of above-threshold detachment of F^- by a circularly polarized few-cycle laser pulse [16].

In the present work, we provide angularly resolved electron spectra of H^- detached by few-cycle infrared laser pulses of frequencies near the two-photon detachment threshold. We show that the spatial distribution of the electron is strongly dependent on the CEP of the pulse. Because of the low binding energy of negative ions, the low laser intensity needed for detachment allows a robust and convenient determination of the CEP [9].

The fact that there is only one bound state for H^- allows an accurate theoretical treatment of photodetachment by short laser pulses [17]. We assume the ir laser, with a frequency ω and a pulse duration τ, is polarized along the z axis, whose vector potential is given by

$$\mathbf{A}(t) \equiv A(t)\hat{\mathbf{z}} = A_0 \sin^2\left[\frac{\pi}{\tau}\left(t+\frac{\tau}{2}\right)\right]\sin\left[\omega\left(t+\frac{\tau}{2}\right)+\phi\right]\hat{\mathbf{z}}, \tag{1}$$

where ϕ is the CEP. The peak value of the vector potential is related to the peak laser intensity I_0 by $A_0=E_0/\omega=\sqrt{I_0/I_{\text{a.u.}}}/\omega$. The period of the laser T_{L} is given by $2\pi/\omega$. According to Eq. (22) of Ref. [17], after the interaction of the ir laser pulse, the S-matrix transition amplitude from an initial state with bound energy E_{b} to a final state with energy $E=\mathbf{k}^2/2$ is given by

$$S_{\mathbf{k}} = i\int_{-\infty}^{+\infty} dt e^{i(E+E_{\text{b}})t}\mathbf{d}[\mathbf{k}+\mathbf{A}(t)]\cdot\mathbf{E}(t) \times\exp\left\{-i\int_t^{\infty} dt'\left[\mathbf{k}\cdot\mathbf{A}(t')+\frac{A^2(t')}{2}\right]\right\}, \tag{2}$$

where $\mathbf{E}(t)=-\frac{\partial}{\partial t}\mathbf{A}(t)$ is the electric field strength. For H^-, the dipole transition operator from the ground state to the continuum state in momentum space is given by [17]

$$\mathbf{d}[\mathbf{k}+\mathbf{A}(t)] = \frac{4C_i}{\sqrt{2\pi}}\frac{[\mathbf{k}+\mathbf{A}(t)]}{\{[\mathbf{k}+\mathbf{A}(t)]^2+2E_{\text{b}}\}^2}, \tag{3}$$

where $C_i=0.315\,52$ and $E_{\text{b}}=0.027\,751$ a.u.

Because of the symmetry in k_x and k_y, we can consider the representative case where $k_y=0$. Thus Eq. (2) can be rewritten as

$$S_{k_xk_z} = i\frac{4C_i}{\sqrt{2\pi}}\int_{-\infty}^{+\infty} dt e^{i(E+E_{\text{b}})t}\exp\left(-i\int_t^{+\infty} dt'\left[k_zA(t')+\frac{1}{2}A^2(t')\right]\right)\frac{E(t)[k_z+A(t)]}{\{k_x^2+[k_z+A(t)]^2+2E_{\text{b}}\}^3}. \tag{4}$$

The transition probability to the final state $(k_x, k_y=0, k_z)$ is calculated by

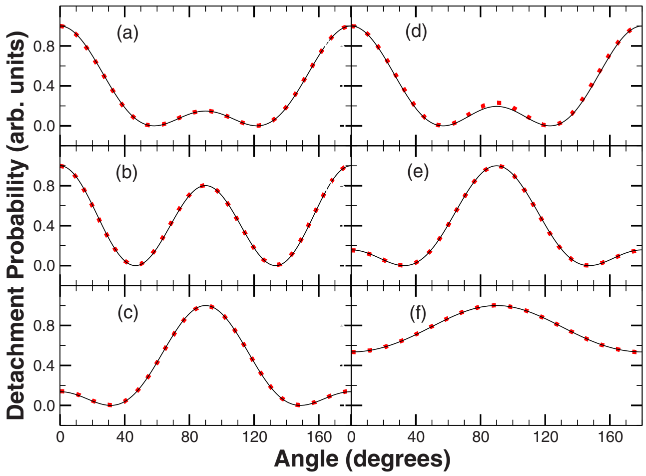

FIG. 1. (Color online) Angular distributions of two-photon detachment by a long pulse of $50T_L$ (solid curves), compared with the results of Ref. [15] (dotted curves), for laser intensity of $I_0=1\times10^{10}$ W/cm^2 (left-hand column) and $I_0=2\times10^{11}$ W/cm^2 (right-hand column) at different wavelengths: 1800 nm (first row); 2400 nm (second row); 2700 nm (third row). The probability in each plot is normalized to unity at its maximum.

$$P(k_x,k_z) = |S_{k_xk_z}|^2, \tag{5}$$

which gives the corresponding energy distribution

$$P(E,\theta) = P(k_x,k_z), \tag{6}$$

where θ is the angle between the observation direction and the laser polarization $\hat{\mathbf{z}}$. The two distributions are normalized such that $\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}P(k_x,k_z)dk_xdk_z \equiv \int_0^{\infty}\int_0^{2\pi}P(E,\theta)dEd\theta$.

For a laser pulse with the vector potential of Eq. (1), we can carry out the integration over t' in Eq. (4) analytically. The overall integration over t is carried out numerically using the Gauss-Kronrod-Patterson method with a precision of 10^{-14}. In order to compare with other theoretical results for the monochromatic laser case, we calculate the electron angular distribution for a rather long pulse of duration $\tau=50T_L$ at various wavelengths and intensities. In Fig. 1, our results are compared with those of [15], in which a nonperturbative quantum electrodynamics scattering theory is used. In each plot of Fig. 1, the angular distribution is shown at the electron energy $E=2\omega-U_p-E_b$, where U_p is the ponderomotive energy, $U_p=E_0^2/4\omega$. One notices that both calculations are almost identical except for the case in (d) where a very small difference around 90° appears.

Now let us consider the short pulse case where the duration $\tau=3T_L$. In Fig. 2, we present angle-resolved electron spectra of H^- detached by a laser of wavelengths 1800 nm and 2400 nm for three different values of the CEP ϕ (0, 0.5π, and 1.5π). The peak laser intensity I_0 is taken to be 4×10^{11} W/cm^2 for all plots in Fig. 2. The general feature for both wavelengths is that the momentum distribution $P(k_x,k_z)$ is symmetric about $k_x=0$. When the CEP $\phi=0$, it is symmetric about $k_z=0$ as well. For other values of ϕ, the symmetry about the $k_z=0$ axis is broken while the symmetry about the $k_x=0$ axis remains. For $\lambda=1800$ nm and $I_0=4\times10^{11}$ W/cm^2, the maximum value of k_z for two- and three-photon detachment is estimated to be 0.192 and 0.296 a.u. by assuming $k_x=0$ and $k_z=\sqrt{2(n\omega-U_p-E_b)}$, where n is the number of photons absorbed. Indeed, one observes that the detachment probability $P(k_x,k_z)$ peaks at these two values in Fig. 2(a) along the $k_x=0$ axis. Similarly in Fig. 2(d), the detachment probability peaks at the predicted k_z values for two- (0.068 a.u.), three- (0.206 a.u.), and four-photon (0.284 a.u.) detachment for $\lambda=2400$ nm and $I_0=4\times10^{11}$ W/cm^2.

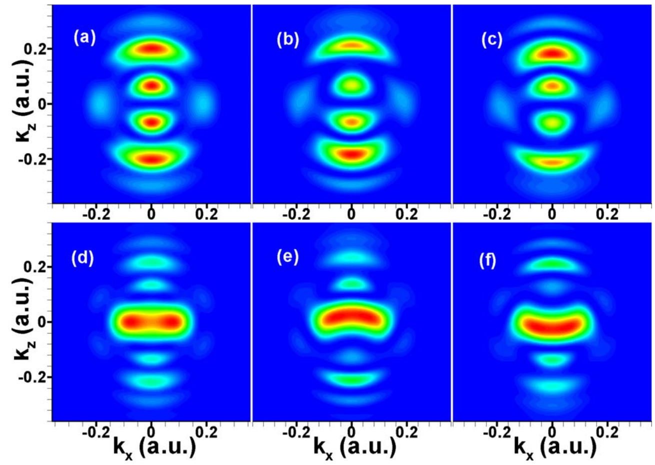

FIG. 2. (Color online) Angularly resolved electron distributions by short laser pulses of $\tau=3T_L$ and $I_0=4\times10^{11}$ at $\lambda=1800$ nm (upper row) and 2400 nm (lower row). The CEP ϕ is taken to be 0 (left-hand column); 0.5π (middle column); 1.5π (right-hand column).

Besides these expected peaks, one observes additional peaks. In Fig. 2(a), an obvious peak appears around $k_z=0.056$ a.u. While in Fig. 2(d), an additional peak occurs at around $k_z=0.131$ a.u. Actually, this can be understood as a feature of few-cycle laser pulses: The bandwidth is so broad that one-photon detachment becomes possible. For a short pulse of $\tau=NT_L$, the value of the vector potential $A(t)$ in Eq. (1) can be rewritten as [3]

$$A(t)=\frac{A_0}{2}\sin\left[\omega\left(t+\frac{\tau}{2}\right)+\phi\right]-\frac{A_0}{4}\sin\left[\omega\left(1+\frac{1}{N}\right)\left(t+\frac{\tau}{2}\right)+\phi\right] - \frac{A_0}{4}\sin\left[\omega\left(1-\frac{1}{N}\right)\left(t+\frac{\tau}{2}\right)+\phi\right], \tag{7}$$

from which one observes that there exist three different frequency components, i.e., $\omega_0=\omega$, $\omega_1=(1+\frac{1}{N})\omega$, and $\omega_2=(1-\frac{1}{N})\omega$.

In the calculations in Fig. 2, the number of cycles $N=3$. For $\lambda=1800$ nm and $I_0=4\times10^{11}$ W/cm^2, one-photon detachment with ω_1 becomes possible, which gives the additional peak around $k_z=0.056$ a.u. However, for $\lambda=2400$ nm and $I_0=4\times10^{11}$ W/cm^2, one-photon detachment with ω_1 is still not possible. But we find that the peak around $k_z=0.131$ a.u. originates from two-photon detachment with the combination of ω_0 and ω_1. Also the other lower frequency combination $\omega_0+\omega_2=5\omega/3\approx0.0316$ a.u. will also be available and generate electrons with momentum around 0.088 a.u.

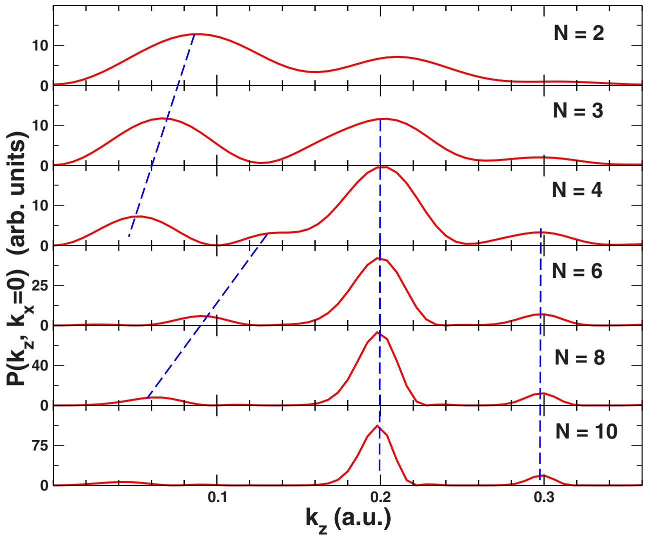

FIG. 3. (Color online) Momentum distributions along $k_z > 0$ and $k_x = 0$ (i.e., $\theta = 0$) for wavelength $\lambda = 1800$ nm, peak intensity $I_0 = 4 \times 10^{11}$ W/cm^2, and CEP $\phi = 0$ for different pulse durations $\tau = NT_L$, where the number of cycles N is indicated in each plot. Dotted lines indicate the evolution of particular features with changing pulse length.

In order to see the effects of these new frequency components due to the short pulse duration, we show in Fig. 3 the momentum distributions along $k_z > 0$ and $k_x = 0$ (i.e., $\theta = 0$) for a short laser pulse of wavelength $\lambda = 1800$ nm, and peak intensity $I_0 = 4 \times 10^{11}$ W/cm^2, and CEP $\phi = 0$ at different pulse durations $\tau = NT_L$. One can immediately note that the two-photon $(2\omega_0)$ peak near 0.2 a.u. and the three-photon $(3\omega_0)$ peak near 0.3 a.u. does not shift when the number of cycles N of the short laser pulse is varied. However, as the pulse length is gradually increased from $N = 2$ to $N = 10$, one clearly observes that momentum peaks lower than 0.2 a.u. are shifted to lower electron energy. These observations indicate that these lower momentum peaks indeed come from the one photon $(1\omega_1)$ process and the pair of two photon processes $\omega_0 + \omega_{1,2}$ discussed in the previous paragraph.

In Fig. 4, for the case $\lambda = 1800$ nm, we provide energy distributions $P(E, \theta)$ along $\theta = 0$ and π as a function of the CEP ϕ for different laser pulse lengths τ. Obviously, the energy distribution has perfect symmetry between the forward $(\theta = 0)$ and backward $(\theta = \pi)$ direction: The distribution in the forward direction for ϕ is exactly the same as that in the backward direction for $2\pi - \phi$. This symmetry can also be seen in Fig. 2 for (b) and (c), and for (e) and (f). As expected, the variation of the energy distribution against the change of ϕ is most noticeable for shorter pulse lengths. As the pulse length τ is increased to $7T_L$, the dependence on ϕ almost disappears although the bandwidth effects are still shown as the broad distribution of two- (0.501 eV) and three-photon (0.296 eV) detachment. The one-photon peak by ω_1 is also barely observable in Figs. 4(e) and 4(f) at the energy 0.05 eV.

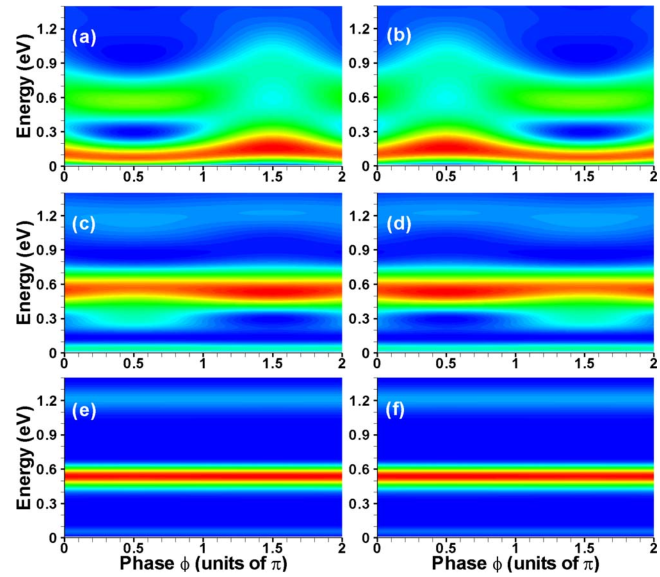

FIG. 4. (Color online) Energy distribution of the detached electron as a function of the CEP ϕ for $\lambda = 1800$ nm and $I_0 = 4 \times 10^{11}$. The pulse duration τ is $3T_L$ (first row); $5T_L$ (second row); $7T_L$ (third row). The observation angle θ is 0 (left-hand column), π (right-hand column).

In summary, we have provided angularly resolved electron spectra resulting from photodetachment of H^- by few-cycle laser pulses near the two-photon threshold. The distributions exhibit sensitive dependence on the CEP and the pulse duration. This sensitivity provides another means to determine the CEP of a laser pulse.

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