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Asymmetry in Photoionization of Atoms Irradiated by Few-cycle Laser Pulses

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Abstract: Photoionization of atoms in intense, few-cycle laser pulses is inversion asymmetry. An asymmetric parameter is used to quantitatively analyze the asymmetry degree. By means of a non-perturbative quantum scattering theory and employing a three-mode laser field to mimic the short pulse, the variation of the asymmetric parameter are researched with the carrier-envelope phase and duration of the pulses. It is found that the asymmetry degree varies with the carrier-envelope phase as a sine-like pattern, and the maximum of asymmetry degree varies with pulse intensity and pulse duration. Along with the increasing laser intensities, the maximal asymmetry firstly decreases and then increases after it reaches a minimal value. At higher intensities, the asymmetry is still distinctive for relative-long few-cycle pulses. Thus, increasing the pulse intensity is helpful to observe the carrier-envelope phase-dependence.

Key words: Inversion asymmetry; Few-cycle laser pulse; Photoelectron angular distribution; Carrier-envelope phase

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0 Introduction

The ultrashort intense laser pulses with duration as short as few optical cycles, as a powerful research tool, are widely used in many fields^[1-3]. For few-cycle pulses, the temporal shape of the electric field varies with the initial phase of the carrier wave with respect to the pulse envelope, i. e., the carrier-envelope (CE) phase. All physical processes depending on the electric field of the laser pulses may depend on its CE phase^[1-4].

In the photoionization of atoms irradiated by few-cycle pulses, the numbers of ionized electrons in the direction of the positive and the negative optical fields are not always the same, which is termed as inversion asymmetry. The inversion asymmetry has attracted much attention, both experimentally and theoretically^[4-10]. Because the inversion asymmetry varies with the CE phase, the measurement of asymmetry manifests directly the CE phase of a short pulse^[11-13].

The inversion asymmetry is intensity-dependent and varies with the pulse duration. In Ref. [10], very large asymmetry was

demonstrated for the pulses persisting up to eight cycles at a higher intensity, while the asymmetry decreases quickly with the increase of pulse duration at a lower intensity. In the calculations performed by means of the time-dependent Schrodinger equation (TDSE), the asymmetry varies dramatically with the laser intensity and the pulse duration^[14].

To quantitatively study the inversion asymmetry, an asymmetry parameter is introduced^[9,10]. The asymmetry parameter is defined as the ratio of the difference to the sum of partial ionization rates in opposite directions:

$$a = \frac{P_+ - P_-}{P_+ + P_-} \quad (1)$$

where P_+ and P_- are the partial ionization rates in a pair of opposite directions, respectively. This parameter is zero for symmetric ionization and reaches its extrema ± 1 when the ionization occurs completely in one side^[9]. Thus we prefer to term this parameter as the asymmetry degree. The asymmetry degree disclosed by Chelkowski *et al.* is of maximum about 60%^[9], and the experimentally observed asymmetry in radio-frequency domain is far larger than that in infrared

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domain^[7,10].

In our study on the photoionization of atoms in few-cycle pulses, distinctive asymmetry appears in the photoelectron angular distributions (PADs)^[15-17]. The asymmetric PADs vary with the CE phase, the pulse duration, and the kinetic energy of photoelectrons. A large asymmetry appears in the PADs of certain kinetic energies, and the corresponding asymmetry degree can be very close to 100%, both in linearly polarized pulses and in circularly polarized pulses. Then, how about the asymmetry in the overall ionizations?

Based on our earlier works, in this paper, we study the asymmetry degree in the ionizations in few-cycle pulses of various peak intensities and of different pulse durations. We will show that increasing the pulse intensity is helpful to observe the CE phase-dependence.

1 Theory

We use a nonperturbative scattering theory of photoionization^[18], which treats the Volkov states as intermediate states while the final state of a photoelectron is an electron plane wave. The electric field of n -cycle pulses is written as

$$\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t + \phi_0) \sin^2(\pi t / \tau + \pi/2) \quad (2)$$

where ϕ_0 is the CE phase. The pulse duration is defined as $\tau = 2n\pi/\omega$, which differs a little from the pulse width τ_p defined as the half width at half maximum (FWHM). For 800 nm laser pulses, $\tau = 2.67n$ fs while $\tau_p = 0.97n$ fs. We use a three-mode laser field to mimic a train of identical n -cycle pulses

$$\omega_1 = \omega; \omega_2 = \omega(1+1/n); \omega_3 = \omega(1-1/n) \quad (3)$$

with the intensity of the sideband modes being a quarter of that of the central mode. The advantage of our treatment is that the ionization rate can be obtained analytically^[15-17]. Our treatment reproduces the asymmetric PADs, and also shows the reversal of asymmetry with shifting the CE phase by π .

The differential ionization rate for photoelectrons with given kinetic-energy is ($\hbar = 1, c = 1$)

$$\frac{dW}{d\Omega_{\text{pf}}} = \frac{(2m_e^3 \omega^5)^{1/2}}{(2\pi)^2} (q - \epsilon_b)^{1/2} (q - u_p)^2 |\Phi_i(\mathbf{P}_f - \mathbf{k})|^2 \left| \sum_{q_i, j_i} \chi_{j_1 - q_1, j_2 - q_2, j_3 - q_3}(\mathbf{z}_f) \chi_{j_1, j_2, j_3}(\mathbf{z}_f)^* \right|^2 \quad (4)$$

where m_e is the electron rest mass, \mathbf{P}_f is the final momentum of photoelectrons, and \mathbf{k} the central wave-vector of the laser pulse. The integer q

relates the final kinetic energy of photoelectrons as

$$E_k \equiv \frac{\mathbf{P}_f^2}{2m_e} = q\omega - E_b \quad (5)$$

thus is named as the absorbed-photon number, since it denotes the absorbed energy of electrons in unit of ω . The number of transferred photons in the i th mode in overall process, say $q_i (i=1,2,3)$, satisfies the following integer equation

$$q = q_1 + q_2 + q_3 + (q_2 - q_3)/n \quad (6)$$

and the sum over q_i is performed over all the possible q_i that satisfy the above relation. The sum over j_i is performed on the energy shell; j_p is the ponderomotive parameter defined as

$$u_p = \frac{e^2 \Lambda^2}{m_e \omega^3} \quad (7)$$

with 2Λ being the classical-field amplitude of the laser pulse. The generalized phased Bessel (GPB) functions are given by

$$\chi_{j_1, j_2, j_3}(\mathbf{z}_f) = \sum_{m_i} X_{-j_1 + 2m_1 + m_4 + m_5 + m_6 + m_7}(\zeta_{1f}) \cdot X_{-j_2 + 2m_2 + m_4 - m_5 + m_8 + m_9}(\zeta_{2f}) \cdot X_{-j_3 + 2m_3 + m_6 - m_7 + m_8 - m_9}(\zeta_{3f}) \cdot X_{-m_1}(\mathbf{z}_1) \cdots X_{-m_9}(\mathbf{z}_9) \quad (8)$$

where the sum is performed over $m_i (i=1,2,\dots,9)$; $-\infty < m_i < \infty$, and $X_n(z)$ are phased Bessel functions defined by

$$X_n(z) = J_n(|z|) e^{inarg(z)} \quad (9)$$

The arguments of the GPB function are defined as

$$\begin{aligned} \zeta_{1f} &= \frac{2|e|\Lambda_1}{m_e \omega_1} \mathbf{P}_f \cdot \boldsymbol{\epsilon}_1, \zeta_{2f} = \frac{2|e|\Lambda_2}{m_e \omega_2} \mathbf{P}_f \cdot \boldsymbol{\epsilon}_2, \\ \zeta_{3f} &= \frac{2|e|\Lambda_3}{m_e \omega_3} \mathbf{P}_f \cdot \boldsymbol{\epsilon}_3, \mathbf{z}_1 = \frac{e^2 \Lambda_1^2}{2m_e \omega_1} \boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_1, \\ \mathbf{z}_2 &= \frac{e^2 \Lambda_2^2}{2m_e \omega_2} \boldsymbol{\epsilon}_2 \cdot \boldsymbol{\epsilon}_2, \mathbf{z}_3 = \frac{e^2 \Lambda_3^2}{2m_e \omega_3} \boldsymbol{\epsilon}_3 \cdot \boldsymbol{\epsilon}_3, \\ \mathbf{z}_4 &= \frac{2e^2 \Lambda_1 \Lambda_2}{m_e (\omega_1 + \omega_2)} \boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2, \mathbf{z}_5 = \frac{2e^2 \Lambda_1 \Lambda_2}{m_e (\omega_2 - \omega_1)} \boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*, \\ \mathbf{z}_6 &= \frac{2e^2 \Lambda_1 \Lambda_3}{m_e (\omega_1 + \omega_3)} \boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_3, \mathbf{z}_7 = \frac{2e^2 \Lambda_1 \Lambda_3}{m_e (\omega_3 - \omega_1)} \boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_3^*, \\ \mathbf{z}_8 &= \frac{2e^2 \Lambda_2 \Lambda_3}{m_e (\omega_2 + \omega_3)} \boldsymbol{\epsilon}_2 \cdot \boldsymbol{\epsilon}_3, \mathbf{z}_9 = \frac{2e^2 \Lambda_2 \Lambda_3}{m_e (\omega_3 - \omega_2)} \boldsymbol{\epsilon}_2 \cdot \boldsymbol{\epsilon}_3^* \end{aligned} \quad (10)$$

where $\boldsymbol{\epsilon}_j (j=1,2,3)$ is the polarization vector of the j th mode defined by

$$\boldsymbol{\epsilon}_j = [\boldsymbol{\epsilon}_x \cos(\xi/2) + i\boldsymbol{\epsilon}_y \sin(\xi/2)] e^{i\phi_j} \quad (11)$$

where $\boldsymbol{\epsilon}_x$ and $\boldsymbol{\epsilon}_y$ are unit vectors vertical to each other; ξ determines the degree of polarization, such that $\xi = \pm \pi/2$ denotes circular polarization and $\xi = 0$ and π linear polarization. The phase angle $\phi_j (j=1,2,3)$ is related to the CE phase as

$$\phi_1 = 0, \phi_2 = -\phi_0/n, \phi_3 = \phi_0/n \quad (12)$$

In Eq. (10), $2\Lambda_i (i=1,2,3)$ is the classical-field amplitude of the i th mode given by

$$\Lambda_1 = \frac{\sqrt{m_e \omega u_p}}{2|e|}; \Lambda_2 = \frac{n\Lambda_1}{2(n+1)}; \Lambda_3 = \frac{n\Lambda_1}{2(n-1)} \quad (13)$$

The photoelectron rate with given kinetic-energy is obtained by integrating over the solid angle $d\Omega_{\rho_i} = \sin\theta_i d\theta_i d\phi_i$ of photoelectrons, where θ_i is the scattering angle and ϕ_i is the azimuthal angle. The overall ionization rate is obtained by the sum of rates of all the possible q . All the calculations are performed in the polarization plane defined by $\theta_i = \pi/2$.

It has shown that the inversion asymmetry of photoionization is caused by the interference effect among different transition channels^[15-17]. Here one transition channel is identified as one set of q_i that satisfying Eq. (6) for a fixed integer q . The number of channels is determined by the kinetic energy and the pulse duration in Eqs. (5) and (6). More transition channels are available in shorter pulses. Because the kinetic-energy spectrum of photoelectrons varies with the laser intensity, the asymmetry degree in overall ionization varies with the laser intensity and the pulse duration.

2 Numerical results and discussions

We choose Kr as sample atoms in our calculations. The initial wavefunction is chosen as that of the outermost shell $4P_{3/2}$ with binding energy 13.99 eV. The circularly polarized laser pulses are of central wavelength 800 nm and of peak intensity varying from 10 TW/cm² to 90 TW/cm². The asymmetry degree is calculated by means of the partial ionization rates along one pair of opposite directions, and we set $\phi_i = 0$ for P_+ and π for P_- . The choice of circular polarization has the advantage that GPB functions in the transition-rate formulae is simplified largely^[15], thus time-consumed calculations are avoided. We also find that the maximal asymmetry degree along the polarization vector in linearly polarized five-cycle pulses, at $I = 50$ TW/cm², is larger than that in circularly polarized five-cycle pulses. In what follows, we just discuss variation of the asymmetry degree with the CE phase, the pulse intensity, and the pulse duration in circular polarization case. Some calculation results are shown in Figs. 1~3.

Our calculations show that, the asymmetry degree varies with the CE phase as a sine-like pattern, with a periodicity of 2π . Here the sine-like variation pattern means the regular oscillation of the asymmetry degree from its positive maximum to its negative maximum along with the change of CE phase. Similar variation also appears

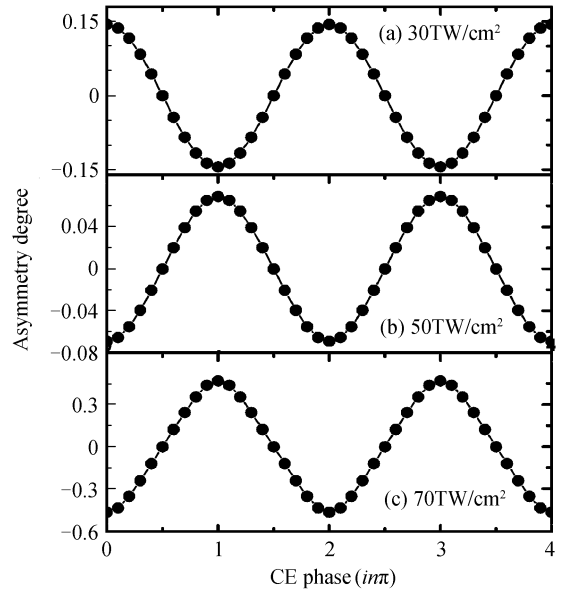


Fig. 1 Variation of the asymmetry degree with CE phase in seven-cycle pulses for several pulse intensities

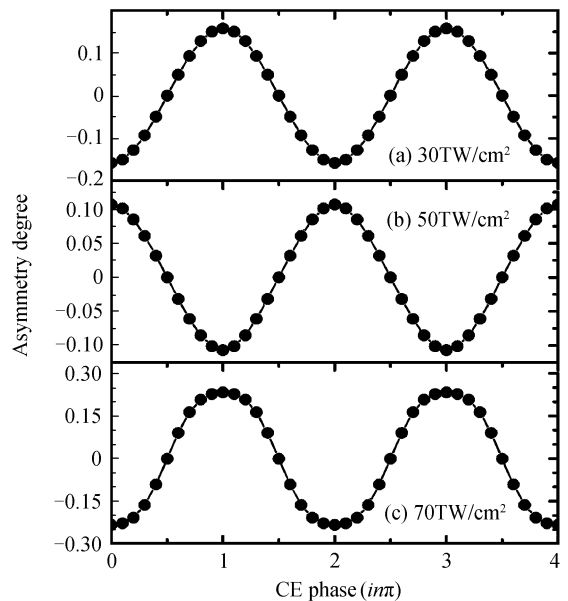


Fig. 2 Variation of the asymmetry degree with CE phase in five-cycle pulses for several pulse intensities in the linearly polarized pulses. This variation pattern reflects the change of electric field in the pulse envelope. For circular polarization, it reflects the change of maximal electric field along one pair of opposite directions with CE phase. For linear polarization, it reflects the change of maximal electric field along the polarization vector. Thus, this variation pattern does not depend on the polarization of laser pulses. According to the observation of Paulus *et. al*^[6-7], the modulation of the CE phase on the ionization in opposite directions shows the oscillation with a periodicity of 2π , which agrees with our calculations.

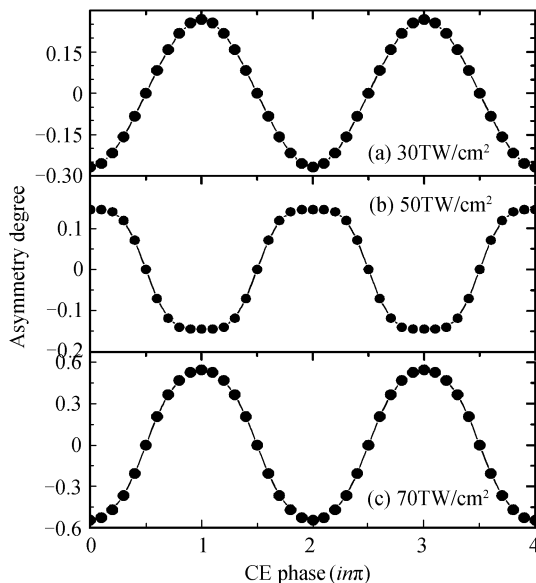


Fig. 3 Variation of the asymmetry degree with CE phase in three-cycle pulses for several pulse intensities

The maximal asymmetry, both positive and negative maximal degrees, appears for the CE phase being the integer times of π . For some special CE phases, such as $\pi/2$ and $3\pi/2$, the asymmetry degree is zero. These phenomena do not vary with the pulse intensity and the pulse length. Noting that the electric field is symmetrically distributed in the pulse envelope when the CE phase equals to $\pi/2 \pm k\pi$, these phenomena mean that the symmetric distribution of electric field produces symmetric ionization, and the asymmetric distribution of electric fields leads to asymmetric electron emissions^[16].

The investigation using TDSE methods also discloses the sine-like variation pattern of the asymmetry degree with the CE phase, and the asymmetry reversal when the CE phase shifts a quantity of π ^[8-9,14]. These phenomena agree with our calculations, but the differences are obvious yet. The calculations using 3D TDSE methods show that the symmetric emission always corresponds to $\phi_0 = -0.3\pi$ and 1.5π , and the maximal asymmetry always corresponds to about -0.5 and 0.71 , when the laser intensity is about $10 \sim 100 \text{ TW/cm}^2$ ^[9]. The calculations using 1D TDSE method show that the value of CE phase corresponding to symmetric ionization varies with the pulse intensity for higher pulse intensity, while at lower intensities, the corresponding value of CE phase is always 0.5π and 1.5π ^[14]. Meanwhile, the Coulomb attraction of parent core to the ionized electron has important influence on the asymmetry^[8].

The total ionization rate, say $P_+ + P_-$, also

varies with the CE phase. Our calculations show that the total ionization rate varies with the CE phase with a periodicity of π , but the modulation is less than 5%, even for 3-cycle pulses (of which the FWHM is about 2.9 fs). The CE-phase dependence of the total ionization rate is hard to be observed, as shown in a recent experiment^[19].

The asymmetry degree varies with the pulse duration. Although we have shown that the inversion asymmetry arises from the interference among transition channels, a more direct picture is that the inversion asymmetry reflects the asymmetric distribution of the electric field in the pulse envelope. Since the asymmetry in the electric field becomes more dramatic in shorter pulses, it is natural that the asymmetry becomes more prominent in shorter pulses. Our calculations indicate that this is the case when the laser intensity is less than 50 TW/cm^2 , as shown in the three figures. Take the asymmetry degree at 30 TW/cm^2 as an example. The maximal asymmetry degree in seven-cycle pulses is about 14.4%, and that in five-cycle pulses is a little higher, reaching 15.7%. The asymmetry degree in three-cycle pulses increases greatly and reaches 26.8%. Meanwhile, the corresponding CE phases to the positive and the negative maximal asymmetry degree vary with the pulse duration. For example, at pulse intensity 50 TW/cm^2 , the asymmetry degree reaches a maximum at $\phi_0 = \pi$ in five-cycle pulses, while that occurs at $\phi_0 = 0$ in seven-cycle pulses, as shown in Fig. 1(b) and Fig. 2(b), respectively. Similar phase shift appears when the pulse intensity changes.

Besides the pulse duration, the pulse intensity also affects the asymmetric photoionization as well as the corresponding asymmetry degree. When the laser intensity is very low, the multiphoton ionization will not occur. Increasing laser intensity, the ionization becomes more and more probable. The ionization cannot be significant until the laser intensity exceeds the ionization threshold. For further higher intensities, it is not always the case that a higher intensity corresponds to higher ionization probability, because the target materials may be depleted before the laser intensity reaches its maximum. Correspondingly, the maximal asymmetry degree varies with the pulse intensity, as shown in Fig. 1~3 for several pulse durations. Fig. 1 shows the variation of the asymmetry degree with CE phase in seven-cycle pulses (with FWHM about 6.8 fs) at several calculated pulse

intensities. The maximal asymmetry degree for pulse intensity 30 TW/cm^2 is about 14.4%. The maximal asymmetry degree decreases with the increasing pulse intensity. When the pulse intensity is 50 TW/cm^2 , the maximal asymmetry degree is only about 7.4%. But for further increasing pulse intensities, the maximal asymmetry increases, for example, the asymmetry degree reaches 45.7% when $I = 70 \text{ TW/cm}^2$. Besides the change of the maximal asymmetry degree, we also find a shift in the corresponding CE phases. For example, the asymmetry degree reaches its positive maximum at $\phi_0 = 0$ for $I = 30 \text{ TW/cm}^2$, but at $\phi_0 = \pi$ for $I = 50 \text{ TW/cm}^2$. That means the optimal emission reverses along with the increase of pulse intensity. Similar phenomena are found for other pulse lengths, as shown in Fig. 2 for five-cycle pulses (with FWHM about 4.86 fs) and in Fig. 3 for three-cycle pulses (with FWHM about 2.92 fs).

Why the maximal asymmetry first decreases then increases, with increasing laser intensity? This phenomenon comes from the nonlinear dependence of ionization on the laser intensity. At lower intensities, a sub-strongest half-cycle ionizes less electrons (here, the sub-strongest half-cycle denotes the one weaker than the strongest half cycle but stronger than others in the envelope, generally it points to the opposite direction against the strongest one), while if the value of electric field in the following strongest half cycle exceeds the ionization threshold, the ionization will be increased significantly. Thus, a larger value of asymmetry can be reached at lower laser intensities. With increasing laser intensity, although the strongest half-cycle may ionize much more electrons than that in the sub-strongest half-cycles, while the latter is also notable, the asymmetry is less than that of lower intensities. Correspondingly, the asymmetry degree decreases with increasing pulse intensity, on the condition that the sub-strongest half-cycles ionize less photoelectron than that in the strongest half-cycle. For further higher intensities, the asymmetry reverses. On the other hand, along with the increasing pulse intensity, the ionization in other half-cycles contributes more and more to the overall ionization. When the pulse intensity reaches a critical value, the ionization will occur mainly at the leading edge of laser pulses, i. e., before the electric field in laser pulses reaching its

maximum. Because the electric field in the leading edge varies more dramatically than that in the top of pulse envelope, the asymmetry degree gets a larger maximal value. The possible maximum of the asymmetry degree depends largely on the pulse envelope. For sine-square pulse envelope, the asymmetry becomes more and more distinctive with the increasing pulse intensity, as long as the pulse intensity exceeds the critical value. Correspondingly, the asymmetry degree becomes larger and larger, but in an oscillation manner.

The critical value of pulse intensity varies with the ionization potential of the target atom, the frequency of the carrier wave, and the pulse duration. In our treatment, the synthesized laser comprises a sequence of identical, few-cycle laser pulses. The electron wave-packet ionized by the laser field comes from many cycles, which leads to the critical value of pulse intensity less than that of single few-cycle pulses.

The maximal asymmetry degree depends not only on the pulse intensity and the pulse length, but also on the kinetic energy of photoelectrons. The maximal asymmetry degree of overall ionizations is found to be about 60%, although the asymmetry degree for photoelectrons at given kinetic energies may be very large. Not all the photoelectrons with different energies have the same optimal direction, and the asymmetry degree may vary with the kinetic-energy of photoelectrons dramatically. For circularly polarized laser pulses, the difference behaves as the CE phase corresponding to the maximum of PADs varying with the kinetic energy of photoelectrons. Our study on PADs has found that along with increasing kinetic energies of photoelectrons, the original maximum of PAD splits into two maxima; the newly produced two maxima evolve to the opposite pole of the symmetric axis, and finally incorporate as a new maximum located in the symmetric axis; thus, after the evolution, the CE phases corresponding to the maximum of PADs differ from each other by a quantity of $\pi^{[20]}$. As a result, the asymmetry degree in the overall ionization decreases. The variation of the asymmetry with the kinetic energy of photoelectrons is observed by Paulus *et. al.*^[6-7].

3 Conclusion

The asymmetry degree in photoionization depends on the CE phase, the pulse duration, and the pulse intensity. The maximal asymmetry

degree decreases with increasing pulse intensity, while increases when the pulse intensity exceeds the critical value. For relatively-longer few-cycle pulses, the inversion asymmetry becomes weak at low intensities while still notable at higher intensities. Considering the dependence of ionization yields on the pulse intensity, we conclude that adopting higher intensity pulses will be helpful to observe the CE-phase effects.

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周期量级激光脉冲作用下原子电离不对称性研究

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摘要:应用不对称性参量对周期量级强激光脉冲下原子电离分布的反演不对称性进行了定量分析. 采用非微扰的散射理论解析方法和三个激光模式模拟超短脉冲, 研究不对称性参量随激光强度、包络位相和脉冲宽度的变化. 计算表明, 这种不对称性是随着波包的绝对位相以正弦形式变化而变化, 其最大不对称程度依赖于脉冲强度和脉冲宽度. 随着激光强度的提高, 不对称性参量是先降低到最小值然后增加. 对脉冲宽度相对长、有几个周期量级的高强度激光, 其不对称性具有显著的特点. 因此, 提高脉冲强度有助于对包络绝对位相变化的观察.

关键词:反演不对称; 周期量级激光脉冲; 光电子角分布; 包络位相