PHYSICAL REVIEW A, VOLUME 61, 033402

Rescattering effect on phase-dependent ionization of atoms in two-color intense fields

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(Received 17 May 1999; published 10 February 2000)

Influence of rescattering process on the two-color laser ionization is investigated by using an improved two-step quasistatic model, in which the Coulomb focusing effect is considered. We focus on phase-dependent rescattering process and ''phase-control.'' It is found that the rescattering leads to further forward/backward asymmetry, breaks the symmetry of rate-phase relation about $\phi=\pm\pi/2$, and accounts for the departure of the prediction of a simple two-step model from the experimental data [Phys. Rev. Lett. **73**, 1344 (1994); Phys. Rev. **A54**, 4271 (1996)]. Our results are in good agreement with the experimental observations.

PACS number(s): 32.80.Rm

In the past two decades, the study of the interaction of atoms with intense laser fields led to a comprehensive understanding of the nonlinear physics in the underlying dynamics of the ionized electrons [1]. This advance was driven by significant progress in both experimental and theoretical capabilities. The recognition of the rescattering process and its leading to phenomena [2–4] was one of the most important steps in complete understanding the atom in laser fields. In fact, this thinking merely comes from a simple quasiclassical notion: Once an electron in a strong field has undergone a transition into continuum from its initial bound state, its motion is dominated by its interaction with the laser field. In the case of linearly polarized field , a majority of these electrons will be driven back into the vicinity of ion core and undergo elastic or inelastic scattering, or be recaptured into the ground state by emitting a high-energy photon. This process is the so-called rescattering process. Now, it is commonly believed that the rescattering is responsible for many unusual observations, such as the cut-off law in high-order harmonic generation, a plateau formed by high-order ATI peaks, and the singular angular distributions of the photoelectrons in the plateau regime [2–9]. Various theories are developed to treat the rescattering. Fully quantum-mechanical calculations were presented both for simplified delta Source and real atomic potential [5,7]; Lewenstein and coworkers have made analyses in the semiclassical framework [6]. Another treatment towards rescattering is directly modifying the quasistatic model by considering Coulomb focusing effect in its second step [8,9].

In the other aspect, two-color laser ionization of atoms became an interesting topic recently, benefiting from the advances in laser technology of ''phase-locking.'' It shows many important novel features that cannot be seen with single-frequency driving [10–20]. Additional interests have risen from the application implemented successfully in the past for ''coherent control'' of atomic and chemical processes [14]. Experimentally, Muller *et al.* [11]. observed the dependence of the ionization yields and ATI spectra on the relative phase in the MPI regime; With more intense lasers, Watanabe [15] observed the phase-dependence ionization of $1\omega-3\omega$ lasers, their experiment falls into tunneling regime and was explained satisfactorily by the quasistatic model. Recently, a fully high-precision experiment was performed by Schumacher *et al.* [16,17]. Their measurements confirm many predictions of the quasistatic model, but there exists an important discrepancy: An asymmetry in the ATI rates relative to the sign of the two-color phase. Although some attempts are made [20] to explain the departure, the results are not satisfactory.

The main purpose of this paper is to observe the rescattering effect on the phase-dependence ionization dynamics in two-color intense fields. To this end, we introduce a three-dimensional (3D) quasistatic model which generalizes the well-known quasistatic model by including the effect of the Coulomb potential on the electron motion after tunneling ionization, and therefore, can describe the rescattering process. Our calculations show that, the rescattering effect leads to further forward/backward asymmetry and the breaking of the symmetry about $\phi=\pm\pi/2$, and resulting in a shift of the phase yielding the peak signal of the total ionization rate at high intensity. Our discussions also give a satisfactory explanation for the experiment [16,17].

As a beginning, we briefly represent the improved two-step quasistatic model adopted in our calculations. The first step, i.e., the ionization of the electron from the bound state to the continuous state, is treated by tunneling ionization theory generalized by Delone *et al.* [21]. In the second step, the motion of an electron in the combined Coulomb potential and the laser fields is described by a classical Newtonian equation.

The initial condition of the Newtonian equation is determined by a equation including the effective potential given in Ref. [22] and a generalized tunneling formula obtained by Delone *et al.* [21]. In parabolic coordinates, the Schrodinger equation for a hydrogen atom in a uniform field ϵ is written,

$$\frac{d^2\phi}{d\eta^2}+\left(-\frac{1}{4}+\frac{1}{2\eta}+\frac{1}{4\eta^2}+\frac{1}{4}\epsilon\eta\right)\phi=0. \tag{1}$$

The above equation has the form of an one-dimensional Schrodinger equation with the potential $U(\eta)=-1/4\eta-1/8\eta^2-\frac{1}{2}\epsilon\eta$ and the energy $K=-\frac{1}{8}$. The turning point, where the electron burn at time t_0, is determined by $U(\eta)=K$. In the quasistatic approximation, the above field parameter ϵ relates to the laser field amplitude $F(t)$ by $\epsilon=F(t_0)$.

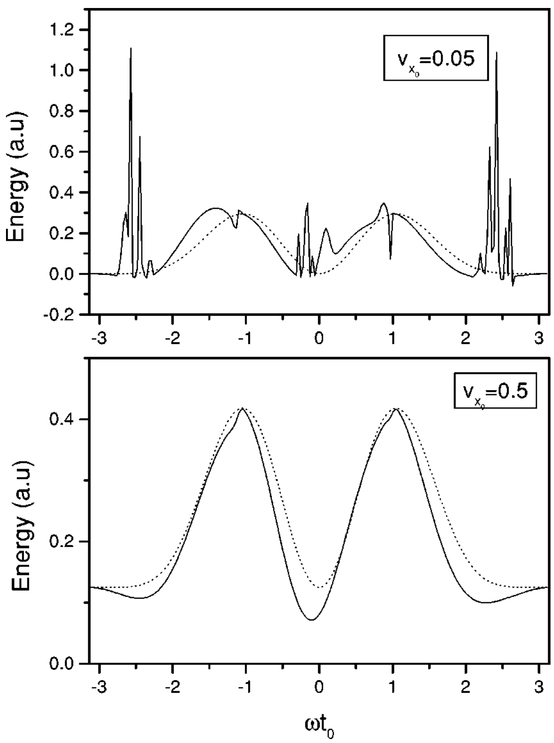

FIG. 1. Electron kinetic energy versus ωt_0. $\phi=0$. Improved two-step model (Solid); Simple two-step model (dotted).

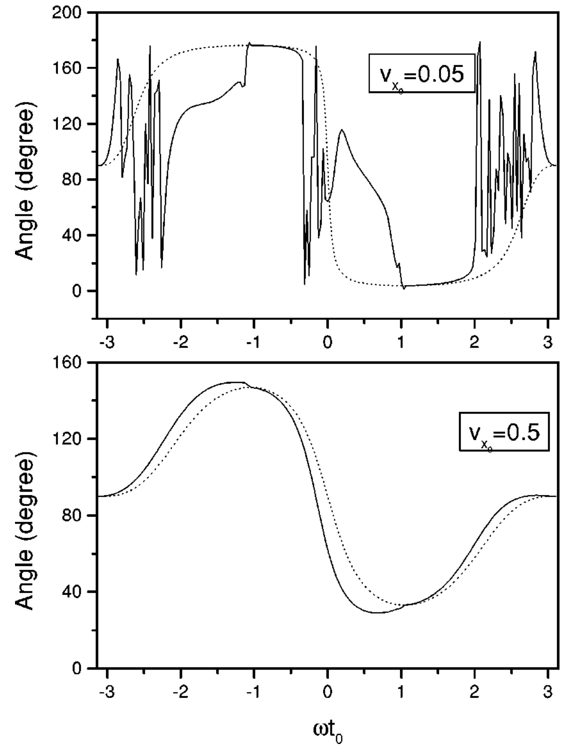

FIG. 2. Emission angle of the electron versus ωt_0. $\phi=0$. Improved two-step model (Solid); Simple two-step model (dotted).

Since the system is azimuthal symmetric about the polarization axis, we can restrict the motion of the electron on the plane (x,z). The initial velocity is set to be $v_z=0, v_x=v_{x0}$. The weight of each trajectory is evaluated by [21]

$$w(t_0,v_{x0})=w(0)\overline{w}(1), \quad w(0)=\frac{4}{\epsilon}\exp(-2/3\epsilon),$$

$$\overline{w}(1)=\frac{v_{x0}}{\epsilon\pi}\exp(-v_{x0}^2/\epsilon). \tag{2}$$

The Newtonian equation describing the motion of the electron after tunneling ionization is

$$m_e\ddot{\mathbf{r}}=-\frac{e^2\mathbf{r}}{r^3}-eF(t)\mathbf{e}_z. \tag{3}$$

Compensated energy E_c advocated by Leopold and Percival [23] is introduced by

$$E_c=\frac{m_e}{2}\left[\dot{\mathbf{r}}+\frac{e}{m_e}\int F(t)dt\mathbf{e}_z\right]^2-e^2/r. \tag{4}$$

When an electron is ionized completely, the Coulomb potential is weak enough and E_c tends to be a positive constant which is just the ATI energy in an ultrashort pulse laser.

We apply our theory to hydrogen atom in the two-color laser field. The laser field is expressed as

$$F(t)=F_1\cos\omega_1 t+F_2\cos(\omega_2 t+\phi), \tag{5}$$

where ϕ is the phase difference between two fields. The field parameters are chosen as $F_1=F_2=0.025$ a.u. ($I_1=I_2=2.25\times10^{13}$ W/cm^2) and $\omega_1=0.04242(\lambda_1=1064$ nm), $\omega_2=0.08484(\lambda_2=532$ nm). Thus, the ponderomotive po-

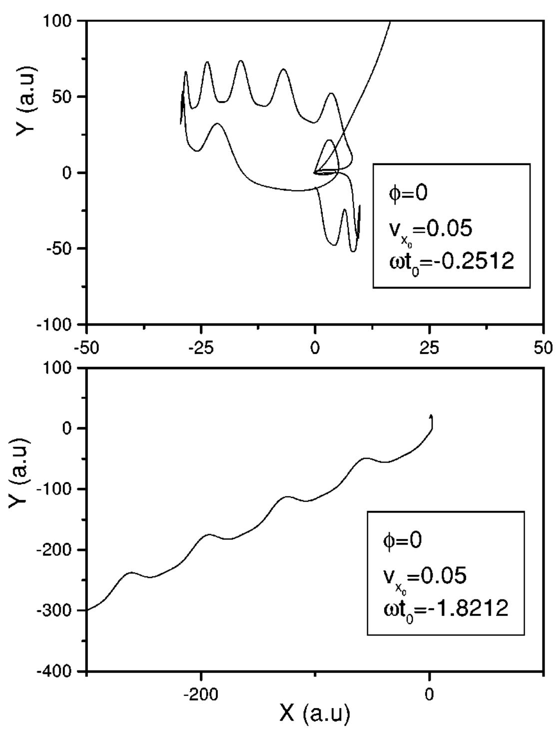

FIG. 3. Typical trajectories of the electron. (a) Multiple return occurs. (b) Direct escape.

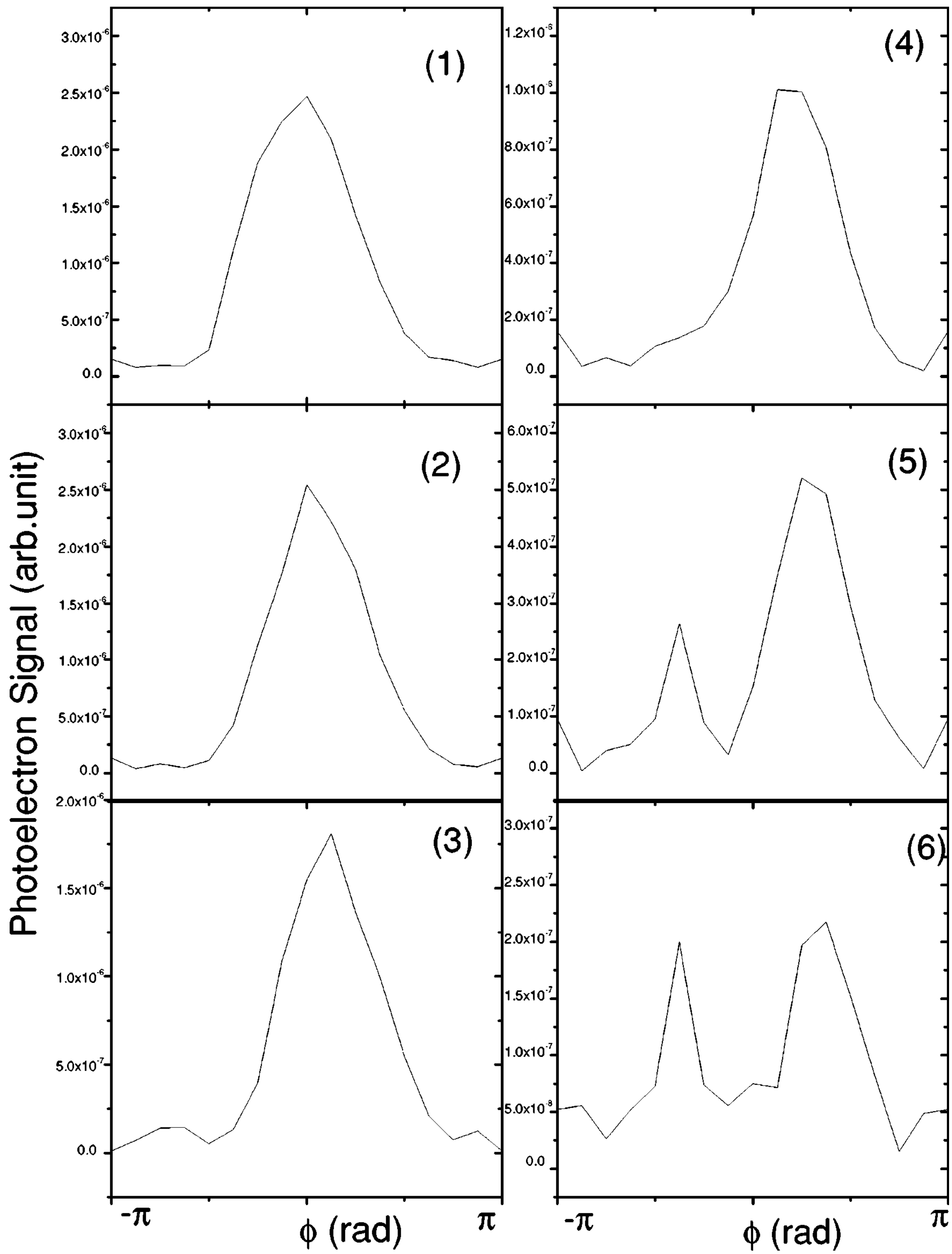

FIG. 4. Phase dependence of ATI: (1)-(6) are the first through sixth forward ATI peaks.

tential $U_p = e^2F^2/4m_e\omega^2 = 0.08683$. The ionization potential $I_0 = 0.5$ a.u.(13.6 eV). Then the Keldysh parameter $\gamma = (I_0/2U_p)^{1/2} \approx 1.69$.

The parameter is so chosen to match the experiment [16,17]. As is well known, the Keldysh parameter is defined as the ratio of the tunneling time and the inverse optical frequency. The tunneling ionization applies as long as the electric field may be regarded constant during the tunneling time, i.e., $\gamma < 1$. Otherwise, the multiphoton ionization dominates in the regime $\gamma > 1$. Even so, there is no definite boundary between multiphoton and tunneling ionization, especially in the practical applications. In the regime where γ is around 1, one find that both multiphoton and tunneling characters present [16,17]. This is the so-called ''mixing regime.'' Tunneling theory was extended to calculate the ion yield in many experiments [10,24–28], and the calculations are consistent with the experimental data even in the regime γ around 1 [25]. It has been shown in Refs. [16] and [17] that although $\gamma>1$ and the ATI spectrum evidently showed the multiphoton character, the phase-dependent variation of the ATI peaks was largely consistent with the phase-dependent predictions of tunneling theory. In practice, we recognize the two essential conditions for the tunneling ionization. That is, the frequency of the laser field is much smaller than the frequency of the motion of the bound electron and the strength of the laser field is less than the value of the critical field (critical field is defined by equating the ionization potential of the bound state to the potential of the saddle point of the combined potential consisting of the laser field and the Coulomb potential). These two conditions are essential and are satisfied by our case. So, we try to use the improved quasistatic model to study the phase dependent rescattering effect. As will be shown later, our results are in good agreement with the experiment.

In our computations, 10^5 initial points are randomly distributed in the parameter plane $-\pi<\omega t_0<\pi$, $v_{x_0}>0$ so that the weight of the chosen trajectory is larger than 10^{-11}. Each trajectory is traced by numerical evaluation of Eq. (3) for such a long time that the electron is actually ionized. As shown above, in our model the rescattering process of the electron after tunneling is described by the Newtonian equation (3) including the Coulomb potential. ATI spectra and angular distribution can be obtained by calculating the statistics on an ensemble of trajectories corresponding to various initial field phases and perpendicular velocities. To investigate the detailed dynamical mechanism of the process and to show the rescattering effect due to the Coulomb focusing, we fix the perpendicular velocity and calculate the initial phase (ωt_0) dependence of the ATI energy and emission angle of different ϕ by using both the improved two-step model and the simple two-step model [in which the Coulomb potential is neglected in Eq. (3)]. Figure 1 shows the energy-ωt_0 dependence and Fig. 2 shows the emission angle-ωt_0 dependence. As the initial velocity (v_{x_0}) perpendicular to the polarization of the electric field is relatively large, the improved two-step model predicts a smooth phase dependence of the ATI energy and emission angle almost coincident with those predicted by the simple two-step model. This is because the Coulomb focusing effect on the electron with large perpendicular velocity is small, i.e., the electron does not rescatter with the nucleon when it quiver in the external field with a large initial perpendicular velocity. Things change in cases where the perpendicular velocity of the electron is quite small. The electron has large probability to collide with the nucleon. This leads to an obvious difference between the predictions of two models. The dependence of ATI energy and the emission angle on the initial phase is poorly resolved in many regions indicating that chaotic scattering happens [9]. In these regions, any arbitrary small change in the initial phase may result in a substantial change of the final electron energy and emission angle. Multiple returns and even an infinitely long time trapping can occur in these regions and they will give the main contribution to the novel distribution in the plateau region of the ATI spectrum [9]. Some typical regular or irregular trajectories calculated by the improved two-step model are demonstrated in Fig. 3. In recent works of G. Sand and J.M. Rost [29], these irregular orbits are also shown to play a dominating role in the high-order harmonic generation.

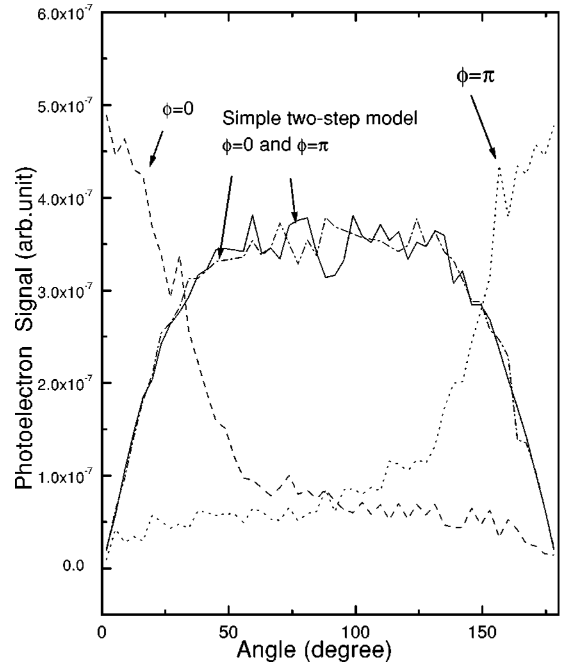

FIG. 5. Angular distribution calculated by the improved two-step model and the simple two-step model.

We calculate the statistics on the ionized electron energy corresponding to different ATI peaks [since the electron energy spectrum obtained by Eq. (4) is continuous, so the count of the nth ATI peak is defined as the sum of the points leading to the energy between $(n-\frac{1}{2})\hbar\omega$ and $(n+\frac{1}{2})\hbar\omega$ in our calculation]. Figure 4 shows the relative ϕ dependence for the first six different ATI peaks. The ATI peaks correspond to electrons in the forward direction (the direction of $\mathbf{e}_z$) just as same as the ATI peaks detected in the experiments [16,17]. Comparing with predictions of simple two-step model, the most essential difference is that, only one phase not two phase as the simple two-step model predicts where the peak signal appears for lower ATI peaks. Obviously, a symmetry about $\phi=\pm\pi/2$ appears in the ϕ dependence of the ATI peaks in the prediction of the simple two-step model but the symmetry no longer remains in the prediction of the improved two-step model. This kind of breakdown of the symmetry can also be found in the experimental data of Refs. [16] and [17], especially remarkable in the low energy ATI peaks of Xenon spectrum.

Inspection shows that this asymmetry is due to the breakdown of forward/backward symmetry showed in the angular distribution Fig. 5. This is one of the important rescattering effects and will lead to the fore-aft asymmetry showed in Fig. 8 of Ref. [17].

Figure 6 shows the relative ϕ dependence for the backward ATI peaks. It is clear that the symmetry about $\phi=\pm\pi/2$ also breaks down but the symmetry with respect to $\phi\to\phi+\pi$, $z\to-z$ and $\omega t\to\omega t+\pi$, which leads to that the

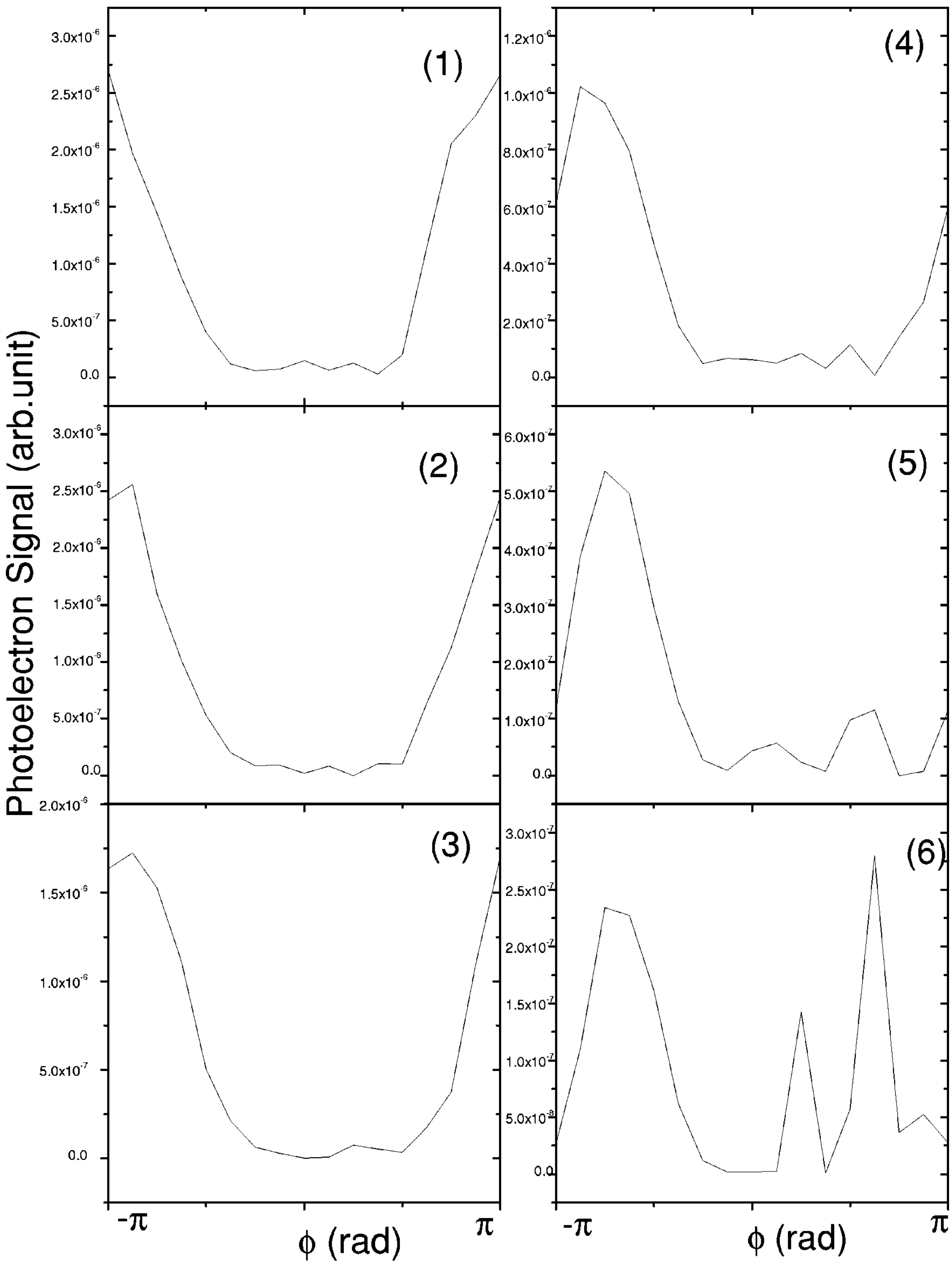

FIG. 6. Phase dependence of ATI: (1)-(6) are the first through sixth backward ATI peaks.

backward rate at phase ϕ is the forward rate at phase $\phi+\pi$ remains. Figure 7 shows the ϕ dependence of the total (summed forward and backward) rates for the ATI peaks. It is qualitatively consistent with the experiment. The reason for the shift of the phase, which yields the peak total rate can be explained as: with respect to the forward ATI electrons, as stated in Ref. [17], the contribution to the high energy electron increases as ϕ increases from 0 to $\pi/2$. So the phase yielding the forward peak signal shifts from 0 to $\pi/2$ as the energy increases. On the other hand, the phase yielding the backward peak signal also shifts from $-\pi$ to $-\pi/2$ for the same reason. As a result, the phase yielding the peak total rate, which is the sum of forward and backward peak signals shifts to positive as the energy of the peak increases.

We would like to point out that the breakdown of the symmetry of the rate-phase relation about $\phi=\pm\pi/2$ has also

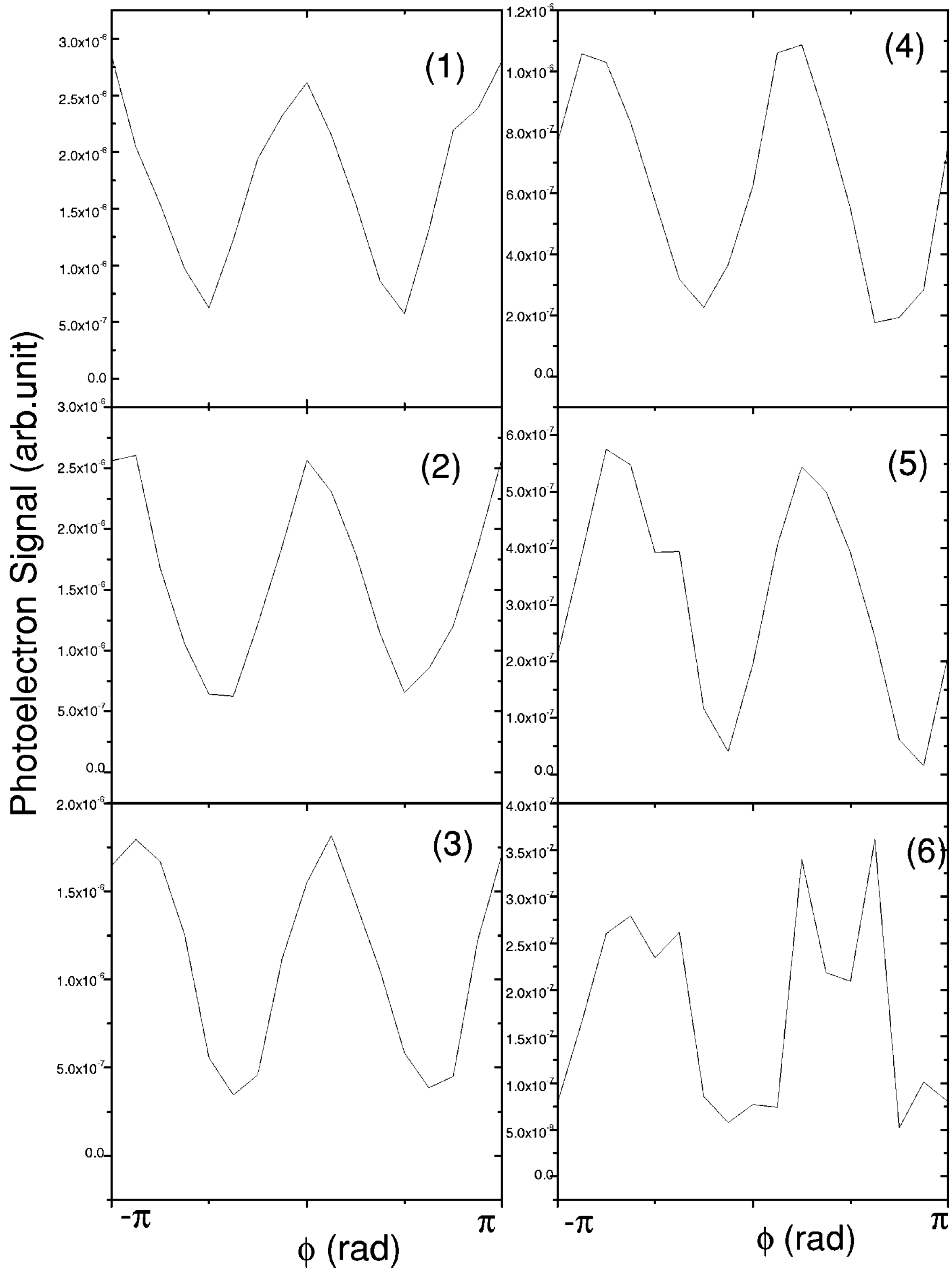

FIG. 7. Phase dependence of the total (summed forward and backward) yield for the ATI peaks.

been shown in paper of K.J. Schafer and K.C. Kulander [13]. However, without including the rescattering process in the calculation, the phase yielding the peak total rate keeps at $\phi=\pm\pi/2$ [corresponding to $\phi=0$ in the Eq. (5) adopted in our calculation] and then the shift cannot be predicted by their theory.

Another important rescattering effect is the increment of the photoelectrons with higher ATI energy. This effect leads to that the contribution from the field with $\phi=0$ to the high-energy ATI peaks increases relatively. Then the shift of the phase yielding peak signal of high energy forward photoelectron from $\phi=0$ is smaller than that predicted by the simple two-step model. This can be shown by comparing Fig. 8 with Fig. 4 and is readily proved by the fact that the prediction of

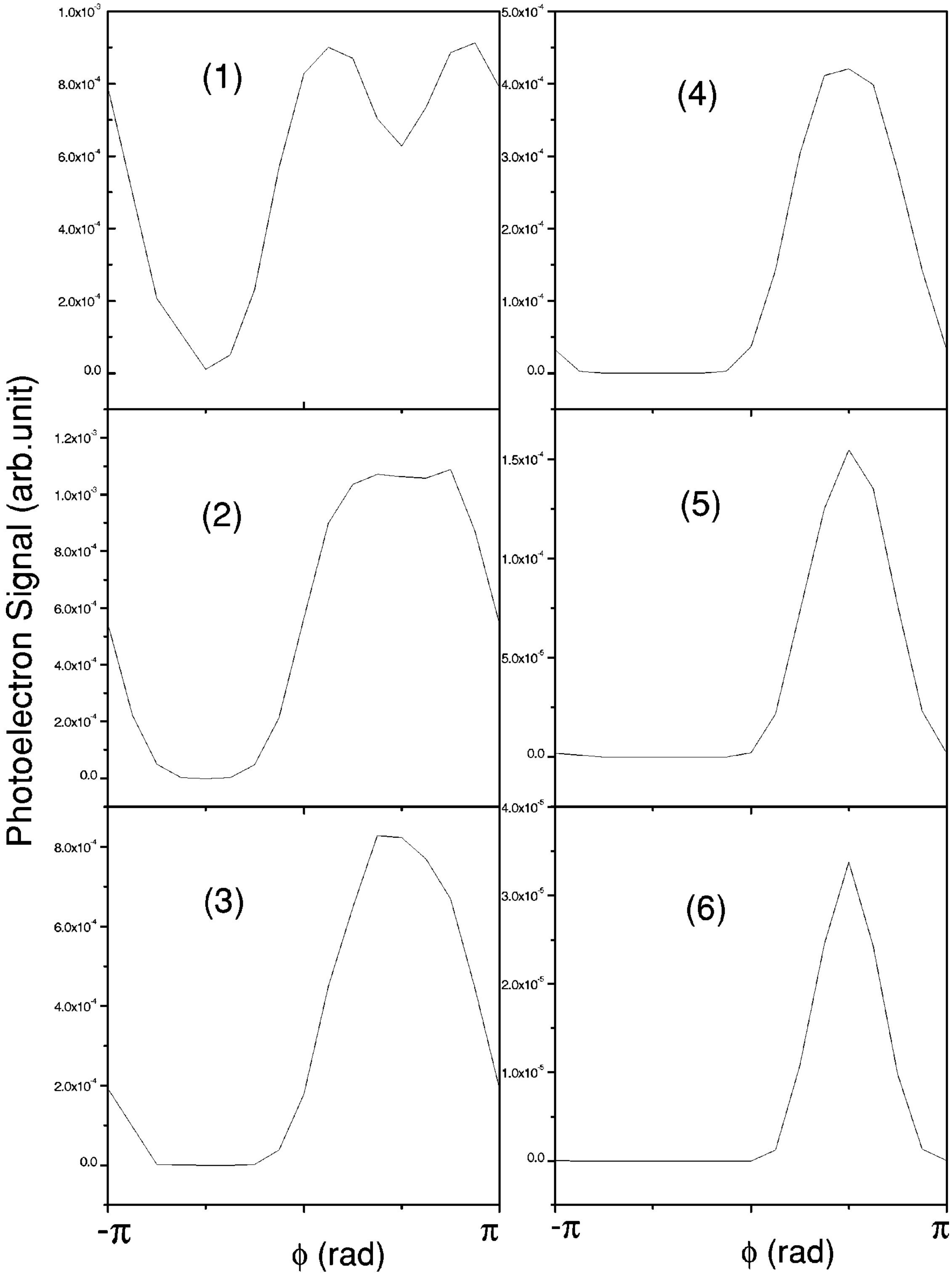

FIG. 8. Phase dependence of the forward ATI peaks predicted by the simple two-step model.

the simple two-step model is more consistent with the experimental data for low intensity than for high intensity.

In conclusion, we have studied the influence of rescattering on the phase-dependent effect in the two-color laser field ionization by using an improved two-step quasistatic model developed recently. The behavior of the classical trajectories of an electron after tunneling are analyzed. We find that the rescattering influences the phase-dependent ionization in two-color intense fields dramatically. It is shown that the departure of the predictions of the simple two-step quasistatic model from the experiment is indeed due to the rescattering effect. Rescattering breaks the symmetry of the ATI rate-phase relation about $\phi = \pm\pi/2$. This breakdown of the symmetry leads to the shift of the phase, which yields the peak signal of the total rate of the ATI peaks. Our results can give a satisfactory explanation to recent experiments on two-color ionization in intense fields.

It also should be pointed out that in our model the rescattering process is treated classically. Although it is commonly believed that transition between continuum can be reasonably described classically, a fully quantum wave evolution is worthy of developing in the future work.

The author J. Chen want to thank Dr. Cheng for helpful discussions. This project is supported by the National Nature Science Foundation/19674011,973 Project and the CAEP Foundation.

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