# Phase dependence in two-color excitation of a model atom by intense fields

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We solve numerically the one-dimensional Schrödinger equation with a model potential for the case of excitation by two commensurate (the fundamental and third harmonic) intense laser fields and find a strong dependence of the atomic evolution and high-harmonic generation on the relative phase between the two fields. Our method employs the Kramers-Henneberger frame to exploit the insights gained from emphasizing the wave-packet dynamics of an electron oscillating under the influence of the strong laser fields.

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The excitation of an atom by a very strong laser field continues to attract attention from experimentalists and theorists [1,2]. The application of a single laser frequency has led to the discovery of above-threshold ionization [3] (ATI), high-harmonic generation [4], and the ejection of electrons by very strong fields in a tunneling process [5]. Recently, attention has been focused on the issue of stabilization [1,2,6] and suppression of ionization in strong fields. Theoretical concepts such as the production of dressed wave packets [7], the use of the Kramers-Henneberger frame [8] to describe strongly perturbed atomic electrons, the adoption of Floquet methods [9], and the use of computationally intensive numerical methods [10] have all contributed to our understanding of strong-field atomic physics. Often, one-dimensional models [11,12] have proved useful in developing theoretical insights which retain their utility in fully three-dimensional (and therefore more complicated) models [13].

When an atom is excited by two distinct frequencies, phenomena related to the phase-dependent modification of the electronic motion are produced [14]. Early work on two-color excitation of atoms by the fundamental and second harmonic of a laser field demonstrated how ATI and ionization yields can be significantly changed by phase-dependent interactions [15,16], suggesting that a form of "coherent control" [17] over the multiphoton dynamics is possible. In the present paper, we extend these considerations of two-color excitation to the intense-field stabilization regime in which the ponderomotive quiver motion dominates the dynamics. We address the problem of two-color excitation by a fundamental and its *third* harmonic: For such a choice of frequencies, there is direct competition between the two fields to access near-resonant states [18,19] so that phase dependence is expected to be significant even at modest intensities. At higher intensities the tunneling followed by a quiver picture of ionization and high-harmonic generation [20–22] can be modified in a phase-dependent way by such a combination of frequencies. These interfere to enhance or diminish that part of the quiver close to the nucleus, as we shall show.

The nonperturbative results we present here have been obtained by solving numerically the one-dimensional Schrödinger equation using a finite-difference approach [10] which exploits the intensity-dependent deformation of the atomic potential. The Schrödinger equation is transformed into the Kramers-Henneberger (KH) frame [8] which oscillates with the laser frequency; it is given by

$$i\hbar\frac{\partial\psi_{KH}}{\partial t}(x,t)=-\frac{\hbar^2}{2m}\frac{\partial^2\psi_{KH}}{\partial x^2}(x,t)+V(x+\alpha(t))\psi_{KH}(x,t),$$

(1)

where the effect of the laser electric field is contained in

$$\alpha(t)=-(e/m)\int_{-\infty}^{t}A(\tau)d\tau ,$$

which is the classical displacement of the electron in the electric field. The "Rochester potential," in atomic units [11],

$$V(x)=-\frac{1}{\sqrt{1+x^2}}$$

(2)

is used as it is continuous across the origin and behaves asymptotically like the Coulomb potential. It produces a Rydberg-like series of high-lying bound states, and retains parity as a good quantum number [11].

The electric field is taken to be classical and of the form

$$E(t)=[E_1\sin(\omega t)+E_3\sin(3\omega t +\phi)]f(t) ,$$

(3)

where $E_1$ and $E_3$ are the fundamental and third-harmonic field amplitudes, $\omega$ is the fundamental frequency, and $\phi$ the relative phase between them. The function $f(t)$ is an envelope function which is chosen to be either a sine-squared pulse: $f(t)=\sin^2(\pi t/\tau)$, where $\tau$ is the pulse length, or a sine-squared ramped turn-on: $f(t)=\sin^2(\pi t/2\tau)$ for $t<\tau$ and $f(t)=1$ for $t>\tau$, where $\tau$ is the length of the turn-on. Because we consider two fields, one of them is chosen to parametrize the length of the pulse: For example, all the sine-squared pulses used in this work are 50 *fundamental* periods long.

Once $\psi_{KH}$ is obtained, it is used to calculate three main quantities: the displacement of the electron wave packet $x(t)$, the probability of ionization (which is defined as

unity minus the bound-state populations), and the acceleration of the electron wave packet, from which the harmonic spectra can be calculated [25].

The results of the numerical integration and their interpretation depend sensitively on the form of the KH wave function, and it is informative to see how these evolve under the influence of the pulse. Let us first examine the case of a smooth sine-squared pulse. In Fig. 1, where we have equal intensities of fundamental and third harmonic, we can clearly see that during the turn-on part of the pulse the initial ground-state wave packet is quickly ionized, producing outward-going ripples of ionization. However, once this is over, the wave function quickly stabilizes (by stabilization, we mean there is no further significant ionization) [11,12]. In Fig. 1(a), where the relative phase $\phi$ is equal to zero, we observe a dichotomous wave packet being formed and destroyed in a way which follows the envelope function of the pulse. The peak separation is a maximum at the peak of the pulse. Although not shown, if $\phi = \pi/2$, we obtain a similar evolution; the initial wave packet bifurcates and then reforms; the bifurcation, however, is not as clearly developed as the $\phi = 0$ case, as more of the wave-packet density lies between the two peaks, which as a consequence are smaller in magnitude. Figure 1(b) shows the evolution for $\phi = \pi$. There is a drastic difference in wave-packet shape; the dichotomy

has now disappeared.

The difference in stabilized wave-packet shape can easily be explained by a simple extension of the one-color stabilization theory [1]. In this case once the frequency and intensity conditions for stabilization are satisfied:

$$\omega \gg |E_{KH}| \tag{4}$$

and

$$\alpha_0^2 \omega \gg 1 , \tag{5}$$

where $E_{KH}$ is the Kramers-Henneberger frame binding energy of the potential, $\omega$ the angular frequency, and $\alpha_0$ the amplitude of oscillation of a free electron in the field in steady state, then the time-dependent KH frame potential $V[x + \alpha(t)]$ is dominated by its time average $V_0(\alpha_0, x)$. This time-averaged dress potential is given by

$$V_0(\alpha_0, x) = \frac{1}{T} \int_{-T/2}^{T/2} V[x + \alpha(t)] dt , \tag{6}$$

where $T$ is the period. For a bichromatic field we have a degree of ambiguity about how to obtain a time-averaged potential. We investigate $V_0(\alpha_1, \alpha_3, \phi, x)$ which is obtained from an average over the longest period; therefore, in our case we take

$$V_0(\alpha_1, \alpha_3, \phi, x) = \frac{1}{T_1} \int_{-T_1/2}^{T_1/2} \frac{-1}{\sqrt{1 + [x - \alpha_1 \sin(\omega t) - \alpha_3 \sin(3\omega t + \phi)]^2}} dt . \tag{7}$$

Here, $\alpha_1$ and $\alpha_3$ are the amplitudes for the fundamental and third harmonic in steady state, respectively; $T_1$ is the period of the fundamental; and $\phi$ is the relative phase between them. The result of this is shown in Fig. 2, in which it is easily seen that for $\phi = 0$ and $\phi = \pi/2$,

$V_0(\alpha_1, \alpha_3, \phi, x)$ is a double-well structure and would therefore be likely to support a dichotomous wave packet. However, for $\phi = \pi$, $V_0(\alpha_1, \alpha_3, \phi, x)$ is fairly flat with small modulations, which we would expect to see support a single-peaked wave packet, as is observed in the numeri-
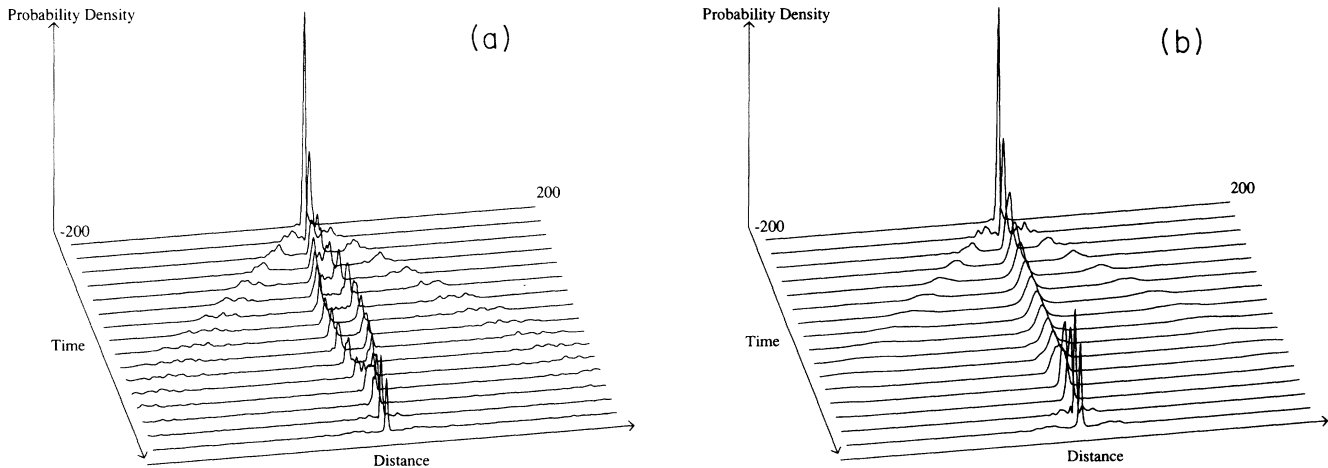


FIG. 1. This shows the spatial and temporal evolution of an atomic wave packet formed under strong-field excitation. We plot $|\psi_{KH}(x,t)|^2$ in arbitrary units over a range of 400 atomic units for $(\omega, 3\omega)$ two-color excitation by a 50-cycle, sine-squared pulse with equal intensities ($E_1 = E_3 = 18.49$ in atomic units) of fundamental frequency ($\omega = 1$ a.u.) and third-harmonic frequency for the relative phases (a) $\phi = 0$ and (b) $\phi = \pi$. Each profile is separated from its predecessor by a time step of $\frac{1}{16}$ of the full pulse duration of 50 cycles.
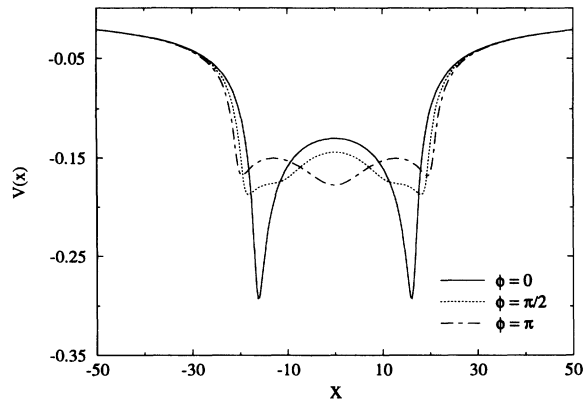
FIG. 2. The time-averaged two-color Kramers-Henneberger potentials as a function of $x$ (in atomic units) for three choices of relative phase with the same field parameters as in Fig. 1.

cal calculations using the full time-dependent dynamics.

By changing the phase, we are therefore able to alter drastically the shape of the stabilized wave packet and by doing so also change the stabilization characteristics. If we plot the probability of ionization $[P(t)]$ for the parameters of Fig. 2, the results shown in Fig. 3 are obtained. Because $\alpha_1$ is greater than $\alpha_3$ by a factor of 9, we expect the fundamental field to affect the dynamics of the wave packet most and this is verified by the fact that the number of oscillations in $P(t)$ is about twice the number of cycles in the pulse, as expected from a stabilization calculation using the fundamental alone. In other words, the electron quiver motion is dominated by the fundamental field. However, the degree of stabilization during and at the end of the pulse depends on the relative phase $\phi$. By changing the phase, we find that the maximal amount of stabilization (basically the lower extrema of the oscillations) is greatest for $\phi = \pi$.

As mentioned previously, the main governing parameter in the intense regime is $\alpha$; therefore, the natural next



FIG. 4. Kramers-Henneberger frame wave packets taken from the peak of a 50-cycle, sine-squared pulse for the case of identical ponderomotive swing amplitudes $\alpha_1 = \alpha_3 = 6.16$ ($E_1 = 6.16$, $E_3 = 55.44$ a.u., and fundamental frequency $\omega = 1$ a.u.) for three choices of relative phase.

step is to arrange $E_1, E_3$ such that $\alpha_1 = \alpha_3$. In this case we find that all three phases produce single-peaked wave-packet evolutions. However, the magnitude of this peak varies with phase; it is a minimum for $\phi = 0$ and a maximum for $\phi = \pi$ (Fig. 4), the $\phi = 0$ wave packet being consequently wider than the $\phi = \pi$ wave packet. One would expect the greatest stabilization for $\phi = \pi$ and this is confirmed in the probability of ionization plots of Fig. 5 where we can see that $P(t)$ during, and at the end of, the pulse is smallest for $\phi = \pi$ than for the other choices of $\phi$.

Taking a closer look at the probability of ionization for the two cases, equal intensities (case 1) and equal $\alpha$'s (case 2) reveal interesting features. In Fig. 6(a) we have plotted $P(t)$ for case 1 over a small section in its time evolution. It is clear that the position of the oscillations in time change with choice of relative phase of the excitation
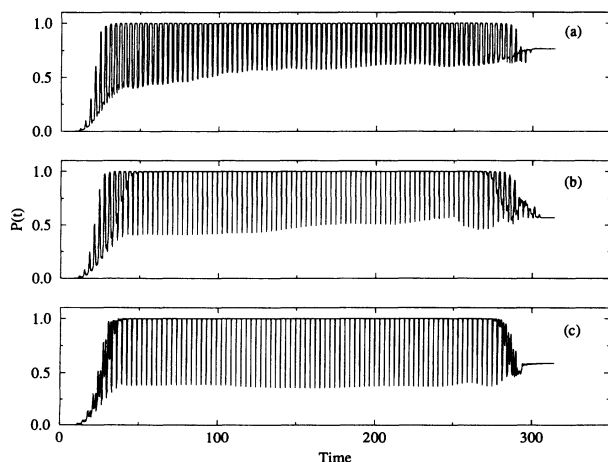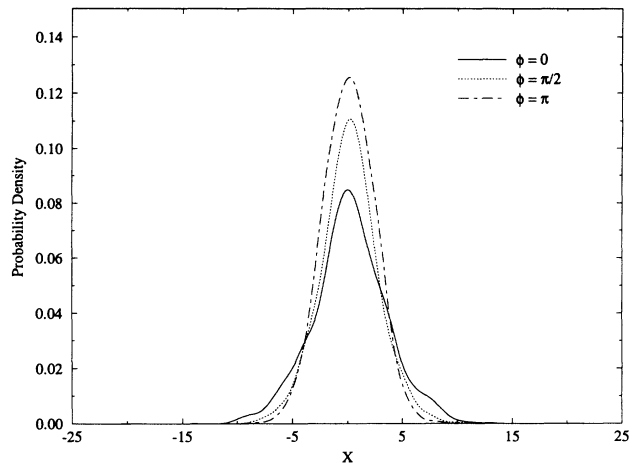


FIG. 3. The probability of ionization as a function of time (in atomic units) for three choices of relative phase (a) $\phi = 0$, (b) $\phi = \pi/2$, and (c) $\phi = \pi$ with the same field parameters as in Fig. 1.
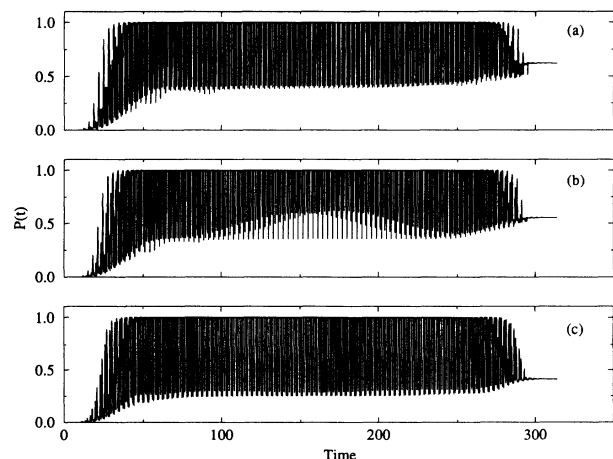


FIG. 5. The probability of ionization as a function of time for the field parameters of Fig. 4 with (a) $\phi = 0$, (b) $\phi = \pi/2$, and (c) $\phi = \pi$.
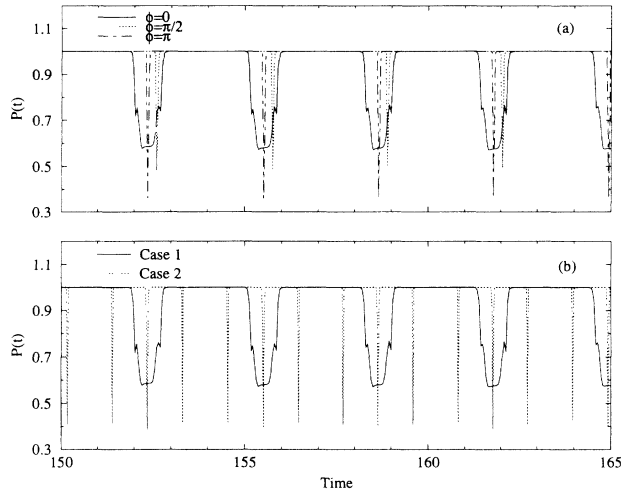
FIG. 6. (a) This is a magnification of a section of Fig. 3 with the three phases shown simultaneously. (b) This is a magnification of the probability of ionization for $\phi=0$, case 1 (equal intensities) for the parameters of Fig. 1 and case 2 (equal $\alpha$'s) for those of Fig. 4.

fields. In Fig. 6(b) we have plotted $P(t)$ for both case 1 and case 2, with $\phi=0$, over the same period of time; we can see that the wide single oscillation in case 1 (where $\alpha_1 \neq \alpha_3$) corresponds to three distinct oscillations in case 2. Both of these features can be explained by looking at the laboratory frame displacement $x(t)$. At each turning point in $x(t)$, the electron has zero kinetic energy, and from the conservation of energy its total energy will be negative. This means that the electron is quasibound and harder to ionize, and so a minimum in $P(t)$ is observed. Therefore, if it is possible to change the position in time and number of turning points in $x(t)$, one should be able to alter the position in time and number of oscillations in $P(t)$. This is possible by varying $\phi$ and $\alpha_3$. Figure 7 demonstrates this: In Fig. 7(a) the expectation value of displacement $x(t)$ is plotted for case 1 and all three

phases. It is easily seen that the shift observed in Fig. 6(a) is due to the shift in the turning points of $x(t)$. To clarify this further, Fig. 7(b) shows $x(t)$ and $P(t)$ for case 1 with $\phi=0$, simultaneously. It is also worth noting that the large width of the oscillations for $\phi=0$ comes from the long period of time that the electron velocity is close to zero. Figure 7(c) shows a similar plot but for case 2 and $\phi=\pi$, from which it is clear that by changing $\alpha_3$, one can change the number of turning points in $x(t)$ and therefore the number of oscillations in $P(t)$.

We have shown that by changing $\phi$, it is possible to change the stabilization characteristics of the atom. As we shall now show, this phase dependence of the quivering electron also has an effect on the harmonic spectra produced. At this point, we change our excitation to that of a smoothly turned on pulse which is then held constant after 5.25 periods of the fundamental, so that we can study the steady-state behavior. In Fig. 8 we show the power spectra for $\phi=0$ and $\phi=\pi$ in the case of equal intensities. A common feature of both spectra is that both odd and even harmonics are present. This happens when atoms are driven by the fundamental and the *second* harmonic and is explained by the emission of $n$ fundamental photons plus or minus $m$ harmonic photons [23]. Here, however, such a simple explanation does not suffice given normal dipole selection rules, and the origin of the even harmonics derives from the breaking of left-right symmetry by the ionization in the initial stages of the short-pulse excitation. It has also been shown theoretically that intense laser stabilized states will radiate both odd and even harmonics [24]. However, by changing the phase from 0 to $\pi$, we see a large change in spectral shape; the cutoff where the highest harmonics diminish into the background has moved to approximately half its value for $\phi=0$. This phenomenon can be explained by looking at the way the harmonic spectra are computed. They are calculated by the method developed by Burnett *et al.* [25] in which Ehrenfest's theorem is used to find the dipole acceleration and it is from this that the power
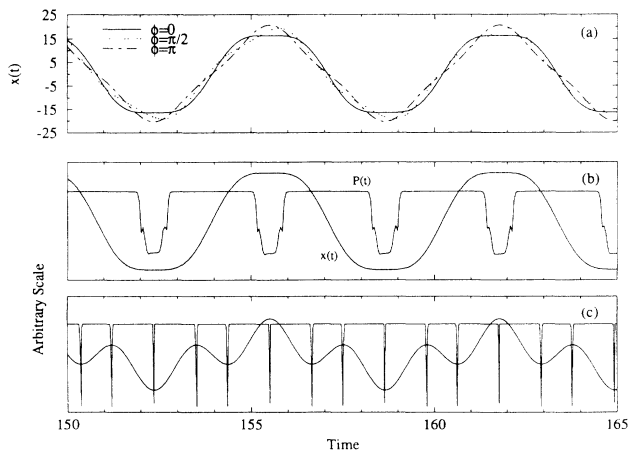


FIG. 7. (a) Displacement as a function of time (in a.u.) for the field parameters of Fig. 1, (b) displacement and the probability of ionization as a function of time for the field parameters of Fig. 1 with $\phi=0$, and (c) the same as (b) but for the field parameters of Fig. 4 with $\phi=\pi$.
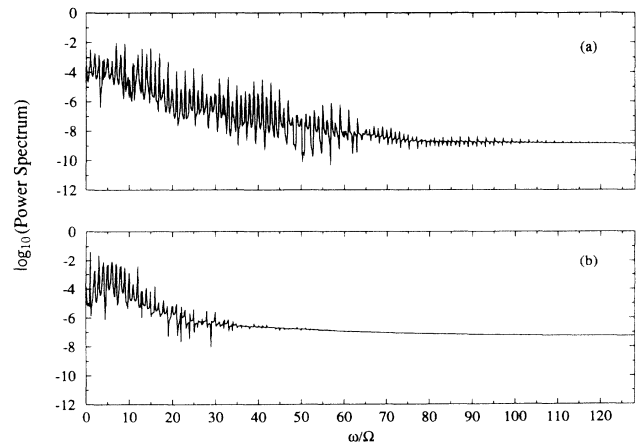


FIG. 8. The harmonic power spectra calculated from the last 8 cycles of a 50-cycle, 5.25-cycle, sine-squared ramped turn-on pulse with the field parameters of Fig. 1 ($\Omega$ being the fundamental frequency) for the relative phases (a) $\phi=0$ and (b) $\phi=\pi$.

spectra of the harmonics is calculated. Following this procedure, using the "Rochester" potential of Eq. (2) yields the expression

$$\ddot{d}(t) = \left\langle \psi_{KH} \left| \frac{x + \alpha(t)}{\{1 + [x + \alpha(t)]^2\}^{3/2}} \right| \psi_{KH} \right\rangle \tag{8}$$

for the acceleration in the laboratory frame. Therefore, the function $x / (1 + x^2)^{3/2}$ sweeps through $|\psi_{KH}|^2$ and acts as a measure of the rate of change in $|\psi_{KH}|^2$. So, if $|\psi_{KH}|^2$ has many spatial components, there will be pronounced high-frequency components in the power spectrum. Our results agree well with this idea; because of the relatively short turn-on of the pulses used in the harmonic analysis, the wave packet is not as clean as those presented earlier in Fig. 1, but the same behavior is observed: The wave packet for $\phi = 0$ has more structure than for the $\phi = \pi$ case and so one would expect an extended harmonic spectrum, as is observed. We can say that these high harmonics are mainly due to the differences in $|\psi_{KH}|^2$ since $\alpha(t)$ for both cases is very simi-

lar, as reflected in Fig. 7(a); however, because being near the nucleus is important for harmonic generation, the phase which makes $x(t)$ closest to the nucleus for longer will produce more harmonics. This can also be seen from Fig. 7(a) where $x(t)$ for $\phi = 0$ is closer to $x = 0$ for longer than $x(t)$ for $\phi = \pi$. The effect of changing $\alpha(t)$ on the harmonic spectra, together with an extended analysis of the above results, will be published elsewhere.

We have shown that, by using two commensurate intense fields, we are able to control the shape of the stabilized wave packet and its ponderomotive motion. Using this control, we are able to change the atomic response, such as the degree of stabilization and the harmonic spectra emitted.

## ACKNOWLEDGMENTS

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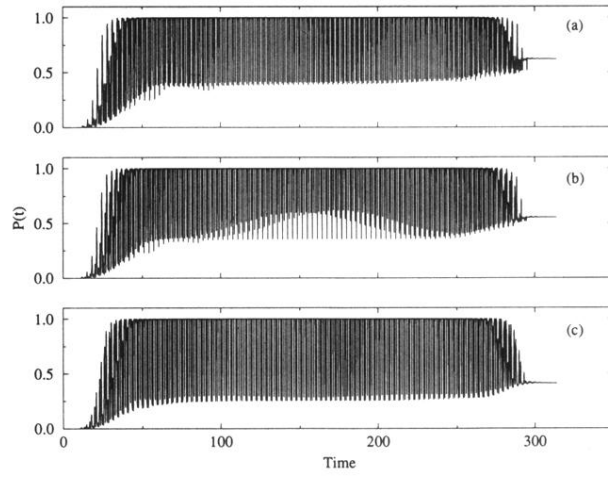[25] K. Burnett, V. C. Reed, J. Cooper, and P. L. Knight, Phys. Rev. A **45**, 3349 (1992).

FIG. 5. The probability of ionization as a function of time for the field parameters of Fig. 4 with (a) $\phi=0$, (b) $\phi=\pi/2$, and (c) $\phi=\pi$.