Carrier-Envelope Phase Effect on Atomic Excitation by Few-Cycle rf Pulses

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We present an experimental and theoretical study of the carrier-envelope phase effects on population transfer between two bound atomic states interacting with intense ultrashort pulses. Radio frequency pulses are used to transfer population among the ground state hyperfine levels in rubidium atoms. These pulses are only a few cycles in duration and have Rabi frequencies of the order of the carrier frequency. The phase difference between the carrier and the envelope of the pulses has a significant effect on the excitation of atomic coherence and population transfer. We provide a theoretical description of this phenomenon using density matrix equations. We discuss the implications and possible applications of our results.

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Modern pulsed lasers can produce ultrashort intense bursts of light with only a few cycles of carrier oscillation [1]. The carrier-envelope phase (CEP) strongly affects many processes involving few-cycle pulses. In particular, CEP effects on high-harmonic generation [2], strong-field photoionization [3], the ionization of Rydberg atoms [4], the dissociation of HD^+ and H_2^+ [5], and the injected photocurrent in semiconductors [6] have been demonstrated by few-cycle pulses. A stabilized and adjustable CEP is important for applications such as optical frequency combs [7] and quantum control in various media [8–10]. Several techniques have been developed to control the CEP of femtosecond pulses [11,12]. A crucial step in attaining this control is measuring the CEP to provide feedback to the laser system. Promising approaches use, for instance, photoionization [13,14] and quantum interference in semiconductors [6].

In this Letter, we report the CEP effect in the population transfer between two bound atomic states interacting with few-cycle pulses. For our experiment, we use intense rf pulses interacting with the magnetic Zeeman sublevels of rubidium (Rb) atoms. We have found that short pulses can be crafted to cause significant population transfer, the CEP of the pulse strongly affects that transfer, and relatively large population transfer may be observed far from resonance.

The significance of our experiment is the following. On the one hand, it provides the insight of CEP effect in a new regime. Unlike the processes mentioned above, our experiment is the first, to our knowledge, to observe the CEP effect on a transition between two *bound* atomic states. The transition is driven by rf pulses with the Rabi frequency comparable to the carrier frequency, a regime studied mainly theoretically to date [15]. Our study in the rf domain suggests new experiments with bound states and few-cycle pulses in the optical domain. This will furnish another way to measure the CEP. Furthermore, the observed phase dependent excitation, as a result of the interference between one-photon and multiphoton transitions [6,16], is important to quantum-control experiments [8–10].

On the other hand, our experiment provides a unique system serving as an experimental model for studying ultrashort optical pulses. Zeeman sublevels are well isolated from other states and provide a good approximation to a two-level system. The level splitting can be tuned over an exceptionally large fractional range. For rf pulses, we have full control of all parameters such as duration, Rabi frequency, CEP, pulse shape, chirp, and more. Currently such control cannot be achieved in the optical domain. Thus, our system is suitable for studies in which the technology is not available in optical domain. It may also lead to further suggestions for optical experiments.

The main result of this Letter is shown in Fig. 1. The ultrashort pulses described in Figs. 1(a) and 1(b) interact with a two-level atom; states $|c\rangle$ and $|d\rangle$ refer to the excited and ground states, respectively, as shown in Fig. 1(c). These pulses have the same envelope but different CEPs. One pulse may be called a cosine pulse [see Fig. 1(a)]; the other a sine pulse [see Fig. 1(b)]. The amount of excited population depends on the shape of the pulses [17]. This is clearly demonstrated in Fig. 1(d) for our cosine and sine pulses.

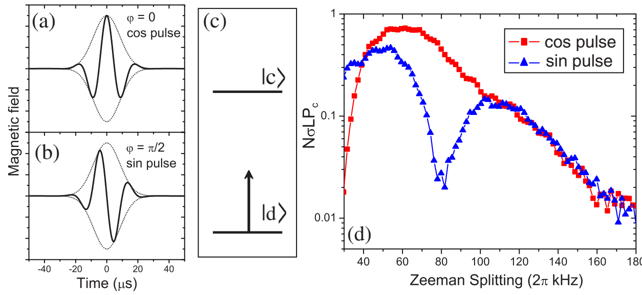

FIG. 1 (color online). A few-cycle rf pulse with different carrier-envelope phases: (a) $\varphi = 0$; (b) $\varphi = \pi/2$. Panel (c) shows a two-level system. Panel (d) shows the population of $|c\rangle$ excited by cosine ($\varphi = 0$) and sine ($\varphi = \pi/2$) pulses.

Our experiment is performed in a gas of rubidium atoms. A 2.5-cm-long cell containing ^{87}Rb (and 5 torr of neon) is located within a magnetic shield. The cell is heated in order to reach an atomic density of the order of 10^{12} cm^{-3}. The configuration of the optical and magnetic fields is shown in Fig. 2. A longitudinal static magnetic field is applied along the laser beam to control the splitting of the Zeeman sublevels of the ground state. A pair of Helmholtz coils produces a transverse rf field at a frequency of 50 kHz.

The energy level scheme is shown in Fig. 2. The ground state (^{87}Rb, $5^2S_{1/2}$, $F = 1$) has three sublevels ($|S_{1/2}1, m_F\rangle$, $m_F = 0, \pm 1$); a circularly polarized laser pulse optically pumps the system and drives the atoms to the sublevel with $m_F = +1$. This is followed by a few-cycle rf pulse, which excites some population to the sublevels with $m_F = 0$ and $m_F = -1$. The population in the sublevels with $m_F = 0, -1$ is subsequently determined by

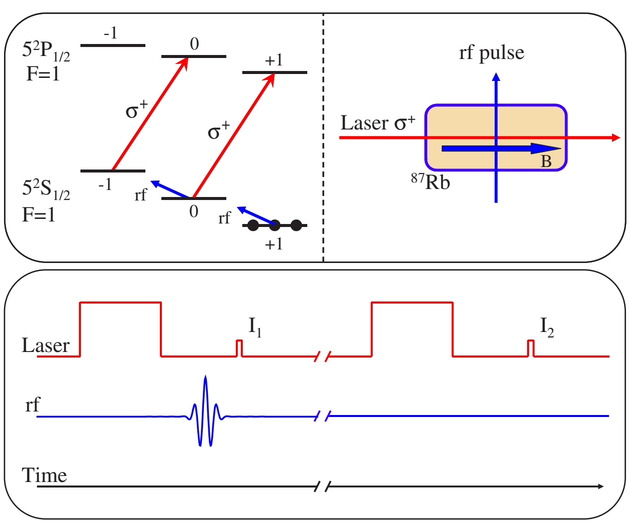

FIG. 2 (color online). The upper block shows a diagram of the relevant energy levels and the geometry of the Rb cell and the applied fields. The lower block shows the time sequence of the laser pulses and the rf pulses.

measuring the transmission of a weak circularly polarized probe pulse.

The Hamiltonian for an atomic state with $F = 1$ in a magnetic field $B = (B_x, B_y, B_z)$ is given by

$$\hat{H} = -g\mu_0 \begin{pmatrix} B_z & \frac{B_x + iB_y}{\sqrt{2}} & 0 \\ \frac{B_x - iB_y}{\sqrt{2}} & 0 & \frac{B_x + iB_y}{\sqrt{2}} \\ 0 & \frac{B_x - iB_y}{\sqrt{2}} & -B_z \end{pmatrix}, \quad (1)$$

where $g = -1/2$ is the Lande factor for the ground state of ^{87}Rb state, μ_o is the Bohr magneton, $B_z = B_0$ is the static magnetic field that is chosen in the direction of the z axis, and B_x and B_y are the transverse components that are driven by a digital function generator. The function generator can be programmed to provide short pulses with controllable parameters, such as the pulse duration and the CEP.

The time sequence of the laser pulses and rf pulses is detailed in Fig. 2. The sequence of laser pulses includes a 1.5 ms strong pulse to optically pump the Rb atoms, and a 5 μs weak pulse (100 μs later) to probe the population of the upper Zeeman sublevels. The sequence repeats every 20 ms. The linearly polarized magnetic field component of the rf field, $B_x = B(t)\cos(\nu t + \phi)$ and $B_y = 0$, has a Gaussian-shaped envelope $B(t) = B_{ox}\exp[-(\frac{t}{\tau})^2]$, where $\tau = T/(2\sqrt{\ln 2})$, and T is the FWHM duration of the pulse. The rf pulse is delayed by 50 μs with respect to the optical-pumping laser pulse, and its duration T varies from 20 to 28 μs.

To determine the population transfer due to the rf excitation, the experiment is performed with a sequence of laser pulses with a rf pulse followed by a sequence of laser pulses without rf pulse. For the first sequence, the transmitted probe pulse intensity is given by $I_1 = I_0\xi e^{-N\sigma L(1-\rho_{gg})}$, where I_0 is the probe pulse input intensity, ξ is a factor due to the relaxation, N is the atomic density, σ is the absorption cross section, L is the cell length, ρ_{gg} is the population of the ground level, and $1 - \rho_{gg}$ is the population of the upper sublevels due to the rf excitation. For the second sequence, in which there is no rf excitation, the transmitted probe pulse intensity is given by $I_2 = I_0\xi$. Therefore, the population due to the rf excitation is given by the quantity $-\ln(I_1/I_2) = N\sigma L(1 - \rho_{gg})$, which is presented as our experimental result in Fig. 1(d).

The main experimental result is shown in Fig. 1(d), where the quantity $N\sigma LP_c$ is plotted as a function of the Zeeman splitting. The behavior is dramatically different for the sine and cosine pulses, as is shown by the blue curves and the red curves, respectively. The rf pulses all have the same carrier frequency (50 kHz), and we modulate the Zeeman splitting. The rf pulse has a Gaussian envelope with the duration (FWHM) of 20 μs (one cycle). For the sine pulse, we observe a well-defined one-photon resonance at 50 kHz and a three-photon resonance that is shifted to about 110 kHz (Bloch-Siegert shift [18]). For the cosine pulse, the upper state population decreases almost exponentially as the atomic frequency is tuned away from one photon resonance. An important feature is that, at 80 kHz, the rf excitation for the cosine pulse is larger by over an order of magnitude compared to that of the sine pulse. This shows that it is crucial to take into account the CEP.

To calculate the behavior of this system, we employ the density matrix approach, which naturally incorporates the relaxation processes. We assume that the optical-pumping step transfers all of the population to the $|S_{1/2}F, m_F\rangle = |S_{1/2}1, +1\rangle$ state, and then we simulate atomic dynamics using the Hamiltonian given by Eq. (1). The set of density matrix equations is given by

$$\dot{\rho} = -\frac{i}{\hbar}[\hat{H}, \rho] - \Gamma(\rho - \rho_0), \qquad (2)$$

where $\hat{H}$ is the Hamiltonian given by Eq. (1), Γ is the relaxation of the system, and ρ_0 is the thermal equilibrium density matrix of atoms.

We also studied the effect of the CEP for different rf pulse durations. As an example, Fig. 3 shows the results for both the cosine and sine pulses with different durations varying from 20 to 28 μs. We compared these experimental results with the theoretical simulations from the set of Eq. (2). The short pulse transfers population from the ground state $|S_{1/2}1, +1\rangle$ to the states $|S_{1/2}1, 0\rangle$ and $|S_{1/2}1, -1\rangle$ (see Fig. 2). The circularly polarized probe laser is absorbed because it is equally coupled to the transitions $|S_{1/2}1, 0\rangle \to |P_{1/2}1, +1\rangle$ and $|S_{1/2}1, -1\rangle \to |P_{1/2}1, 0\rangle$, as shown in Fig. 2 (the dipole moments of both transition are the same). The depletion of population

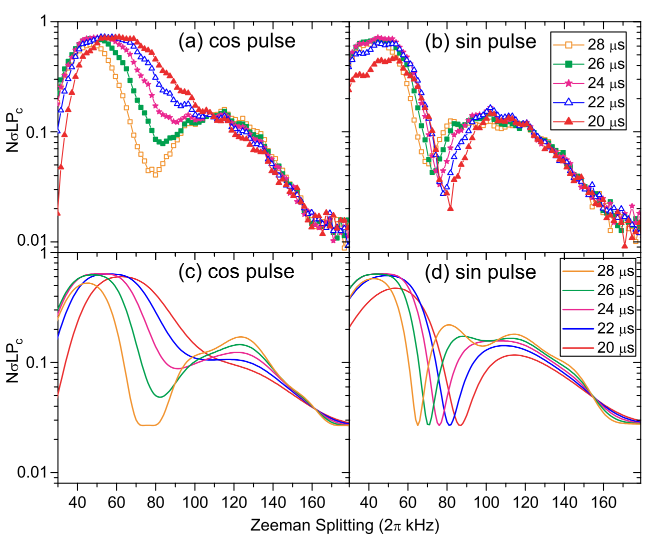

FIG. 3 (color online). The population of the upper Zeeman sublevels excited by cosine (a) and sine (b) rf pulses with different pulse durations ranging from 28 to 20 μs. Corresponding theoretical simulations are shown in (c) and (d).

from the ground state is given by $1 - \rho_{gg}$, where $|g\rangle = |S_{1/2}1, +1\rangle$. The calculated signals shown in Figs. 3(c) and 3(d) agree well with the experimental results shown in Figs. 3(a) and 3(b).

The results can be understood as the CEP dependent interference between one-photon and multiphoton transitions as suggested in [6,16]. When the pulse is so short that its frequency spectrum is broad enough to cover two frequencies for both one- and three-photon transitions, the overall excitation depends on the interference between the one- and three-photon pathways. As shown in Fig. 1(d), the sine pulse has a destructive interference while the cosine pulse has a constructive one. If the pulse duration becomes longer, the narrower spectrum makes the interference and the effect of CEP less pronounced. As shown in Fig. 3, the difference of excitations is less influenced by the CEP as the pulse duration goes from 20 to 28 μs. The results also suggest that a few-cycle pulse with adjustable CEP can manipulate the atomic excitation.

For a simple mathematical description of the results, let us neglect relaxation and consider only two levels coupled by the radiation field [see Fig. 1(c)]. The Rabi frequency is given by $\Omega(t) = \Omega_0 f(t)\exp(-\alpha^2 t^2)$, where $f(t) = f_0(e^{i\nu t} + pe^{-i\nu t})$ is either $\sin(\nu t)$ or $\cos(\nu t)$ (ν is the frequency of the rf field). For $\cos(\nu t)$, $p = 1$ and $f_0 = 1/2$; for $\sin(\nu t)$, $p = -1$ and $f_0 = -i/2$. The equations for the state vector $|\Psi\rangle = Ce^{i\omega_c t}|c\rangle + D|d\rangle$ (ω_c is the frequency of the transition between levels $|c\rangle$ and $|d\rangle$) are

$$\dot{C} = -i\tilde{\Omega}D, \qquad \dot{D} = -i\tilde{\Omega}^* C, \qquad (3)$$

where $\tilde{\Omega} = \Omega_0 f(t)\exp(-\alpha^2 t^2 + i\omega_c t)$. For the lowest order in the coupling field ($D \simeq 1$), the solution of Eq. (3) is

$$C^{(1)} \simeq -\frac{i\sqrt{\pi} f_0 \Omega_0}{\alpha}(e^{-(\omega_c-\nu)^2/4\alpha^2} + pe^{-(\omega_c+\nu)^2/4\alpha^2}). \quad (4)$$

One can see that for long ($\alpha \ll \omega_c$) and short ($\alpha \sim \omega_c$) pulses, both the sine and cosine pulses have a similar profile: we have a sum of two spectral components at $\pm\omega_c$. The profile has no zero points for $\nu > 0$. For the next nonzeroth order, we have four terms that correspond to spectral components at $\pm\omega_c$ and $\pm\omega_c/3$ and their spectral widths are broader. As in Eq. (4), the positive and the negative frequency components do not strongly influence each other. The components at ω_c and $\omega_c/3$ are given by

$$C^{(3)} \simeq i\frac{\sqrt{\frac{\pi}{3}} f_0^3 \Omega_0^3}{\alpha(\omega_c - \nu)^2}(p\xi e^{-(\omega_c-\nu)^2/12\alpha^2} + e^{-(\omega_c-3\nu)^2/12\alpha^2}), \qquad (5)$$

where $\xi = (3\omega_c - \nu)/(\omega_c + \nu)$. For long pulses, these components are easily resolved. But for short pulses (a few periods of oscillations, $\alpha \sim \omega_c$), the widths of these components are broader than the widths of the components of $C^{(1)}$. For the cosine pulse, the components are not

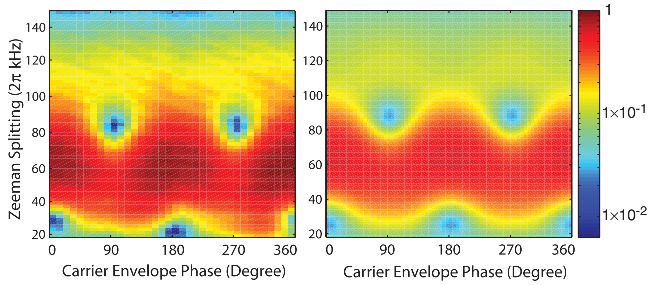

FIG. 4 (color online). Excited state population versus CEP: Experiment (left), simulation (right).

resolved: the profile resembles one broad spectral component. For the sine pulse, the situation is completely different: the total profile has zero at $\omega_c = 2\nu + 3\alpha^2 \frac{\ln\xi}{\nu}$ where the components (having opposite signs) cancel.

Another insight into these observations can be related to the Ramsey effect [19]. Namely, the two pulses delayed with respect to each other produce interference. Sine pulse (see in Fig. 1) can be viewed as a two "subpulses" of the carrier field (one up and one down) where changing the carrier frequency changes the time between these subpulses. Meanwhile, a cosine pulse is more like a one pulse of the carrier field, and changing the carrier frequency just changes its duration. Then, what we have observed is that the first half of the sine pulse is interfering with the second half of the sine pulse, and this interference produces the dip in the excitation of the upper level (see in Fig. 1).

Thus, sine and cosine pulses (which correspond to a change of $\pi/2$ in CEP difference) can be clearly distinguished. Our last study presented here involves changing the CEP in smaller steps. In particular, we performed experiments using pulses with CEPs varying from 0° to 360° by steps of 10° for various Zeeman splittings. The result is shown as an image in Fig. 4 (left-hand panel). The rf pulses used in the experiment have frequency 50 kHz and pulse duration (FWHM) of 20 μs. The obtained data are plotted in Fig. 4. The range of the population in the excited state is the same as in Fig. 3. One can see that the excited population is a continuous function of the CEP at a given Zeeman splitting, and consequently, this can be used to determine and/or to control the CEP of the ultrashort pulses. We have performed simulations that reproduce the observed results, and the results are shown in Fig. 4 (right-hand panel). We note that measuring population in the excited state might be easier than the asymmetry of the direction of produced ions or electrons.

In conclusion, we have theoretically and experimentally studied the carrier-envelope phase effect on population transfer between two bound atomic states interacting with intense ultrashort pulses. We have found that the effect is particularly strong if the pulses are only a few cycles in duration and have the Rabi frequencies of the order of the carrier frequency. The obtained result can be understood in terms of a new analytical solution obtained for a two-level atom under the action of laser radiation with an arbitrary pulse shape and polarization [17]. The interaction of intense ultrashort pulses with atomic system can be studied with rf pulses as a model system. We acknowledge and honor here the ground-breaking experiments performed with cw rf radiation (see [20]). These experiments in the rf region might furnish physical insight for the development of CEP control in optical fields, which are important for the generation of pulses with a predetermined absolute phase.

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