Carrier-envelope phase-dependent atomic coherence and quantum beats

Ying Wu[1,2] and Xiaoxue Yang[1]
[1]*Physics Department, Huazhong University of Science and Technology, Wuhan 430074, People's Republic of China*
[2]*State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, People's Republic of China*
(Received 30 November 2006; published 26 July 2007)

It is shown that the carrier-envelope phase (CEP) of few-cycle laser pulses has profound effects on the bound-state atomic coherence even in the weak-field regime where both tunneling and multiphoton ionization hardly take place. The atomic coherence thus produced is shown to be able to be mapped onto the CEP-dependent signal of quantum beats (and other quantum-interference phenomena) and hence might be used to extract information about and ultimately to measure the carrier-envelope phase.

DOI: 10.1103/PhysRevA.76.013832 PACS number(s): 42.65.Re, 42.50.Md

I. INTRODUCTION

The carrier-envelope phase (CEP) of ultrashort laser pulses of a few cycles [1,2] plays an important role in strong-field ionizations [3–11], strong-field dissociation [12], high-harmonic generation [13–15], and attosecond electron dynamics [16–18]. The CEP may be determined by measuring spatial asymmetry in ionization [11,16], and extreme ultraviolet (xuv) and soft-x-ray radiations [19].

Nakajima and Watanabe [20] have recently studied the CEP dependence in bound-state populations rather than ionization or photoelectron yields in the multiphoton ionization regime where tunneling ionization hardly takes place. It thus enables one to observe the phase-dependent population in some bound states via photoemission detection, which is technically simpler than the energy and angle-resolved photoelectron detection at considerably lower intensities than explored previously [20].

In this paper, we investigate the *atomic coherence* properties produced by an ultrafast laser pulse with a few cycles. It should not be confused with the optical coherence properties of high-harmonic generation studied previously [21,22]. We show that the CEP of few-cycle laser pulses has profound effects on the bound-state atomic coherence even in the weak-field regime (the Rabi frequency is much less than the transition frequencies involved) where both tunneling and multiphoton ionizations hardly take place. This CEP-dependent atomic coherence is then shown to be mapped directly onto the signal of quantum beats [27–29] between two channels of spontaneous emission, and hence it might be used to extract information about the carrier-envelope phase by measuring the quantum beats. It is well known that quantum-mechanical interference due to the coherent superposition of several states (atomic coherence) inevitably leads to oscillations in the amplitude of the detected signal and has been seen in a wide variety of optical experiments, including fluorescence spectroscopy [23], photon-echo techniques [24], transient grating experiments [25], and Raman spectroscopy [26]. Closely related phenomena are the quantum beats [27–29] and versatile wave mixing in cold atomic media [30–38]. It is thus anticipated that the sensitive dependence of the bound-state atomic coherence on the CEP of few-cycle laser pulses revealed in this paper opens entirely different ways of exploring and utilizing the CEP-dependent coherence effects of ultrafast pulses in both the weak-field regime (quite unlike the previous strong-field ionization regimes) and different spectral regions such as spectral regimes from the infrared up to the ultraviolet spectral domains instead of soft-x-ray radiation. In particular, the CEP-dependent atomic coherence might be used, instead of the previous techniques of photoemission detection and measurement of the ionization yields, to extract information about and ultimately to measure the carrier-envelope phase.

II. CARRIER-ENVELOPE PHASE-DEPENDENT ATOMIC COHERENCE

In investigating the CEP-dependent atomic coherence in the weak-field regime where both tunneling and multiphoton ionization hardly take place, we only need to consider an atomic model of a few levels. Here we consider a three-level model interacting with a few-cycle optical pulse of the electric field [20] $E(t)=-\partial A(t)/\partial t$ with the vector potential $A(t)=A_0 e^{-(t-2\tau)^2/\tau^2}\sin(\omega t+\phi)$ as shown in Fig. 1. Here the atomic state is $|\Psi\rangle=c_0|0\rangle+c_1|1\rangle+c_2|2\rangle$, A_0 is the peak value of the vector potential, τ describes the pulse width, and ω and ϕ are the carrier-envelope frequency and phase, respectively. The quantum-mechanical interference due to the CEP-dependent bound-state atomic coherence should be easier to detect at low temperatures, where the deteriorating decoherence such as that due to Doppler broadening is greatly diminished. Consequently, the CEP-dependent atomic coherence should have some impact on and could play a role in versatile phenomena occurring in cold atomic media [30–38]. The three-level model of closely adjacent hyperfine excited levels $|j\rangle$ $(j=1,2)$ can be chosen to be the hyperfine-split levels for the D lines of cold alkali-metal atoms [39] confined in a magneto-optical trap (MOT), where there exist mature techniques to handle the interactions between the lasers and trapped alkali-metal atoms. For instance, in the case of a cesium atom, the levels $|0\rangle$, $|1\rangle$, and $|2\rangle$ can stand for its states $6\,^2S_{1/2}$, $6\,^2P_{1/2}$, and $6\,^2P_{3/2}$, respectively, and the transitions $|0\rangle\leftrightarrow|1\rangle$ and $|0\rangle\leftrightarrow|2\rangle$ correspond to the $D1$ and $D2$ lines with $\mu_{02}=3.8\times10^{-29}$ C m, $\mu_{01}=2.7\times10^{-29}$ C m, $\alpha=\mu_{02}/\mu_{01}\approx1.41$, and the transition frequencies (energies)

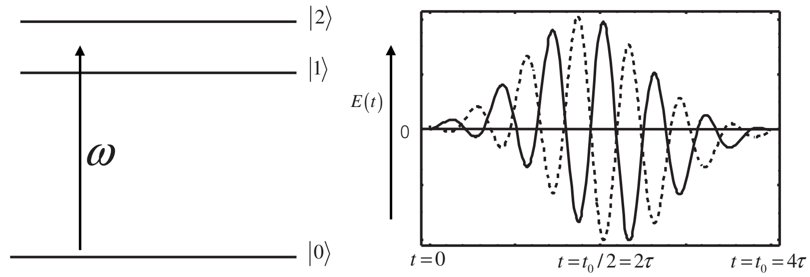

FIG. 1. Schematic diagram of a three-level atom illuminated by a few-cycle laser pulse with the carrier frequency ω and the width τ. The pulse shapes for the two values of the carrier-envelope phase $\phi=0$ (solid curve) and $\pi/2$ (dashed curve) have been plotted in the diagram for illustration.

$\omega_1 \approx 335.1$ THz, and $\omega_2 \approx 351.7$ THz, respectively [39].

The Schrödinger equation for the probability amplitudes c_j has the form

$$\dot{c}_0(t) = i\Omega\xi(t)[e^{-i\omega_1 t}c_1(t) + \alpha e^{-i\omega_2 t}c_2(t)], \tag{1a}$$

$$\dot{c}_1(t) = i\xi(t)\Omega e^{i\omega_1 t}c_0(t), \quad \dot{c}_2(t) = i\xi(t)\Omega\alpha e^{i\omega_2 t}c_0(t), \tag{1b}$$

where $\xi(t)=\omega^{-1}\partial[e^{-(t-2\tau)^2/\tau^2}\sin(\omega t+\phi)]/\partial t$, $\alpha=\mu_{02}/\mu_{01}$, $2\Omega=2\Omega^*=\mu_{01}\omega A_0/\hbar$ and $2\alpha\Omega$ are the Rabi frequencies for the transitions $|0\rangle \leftrightarrow |j\rangle$ $(j=1,2)$, respectively, ω_j $(j=1,2)$ are the atomic transition frequencies concerning the transition $|0\rangle \leftrightarrow |j\rangle$ with the corresponding dipole moments μ_{0j}, and μ_{0j} have been assumed to be real without loss of any generality.

Figure 2 plots the curves of the atomic coherence $\rho_{12}=\rho_{21}^*=c_1c_2^*$ versus the CEP ϕ under the initial conditions $c_{1,2}(0)=0$ and $c_0(0)=1$ at the time $t=t_0=4\tau$ elapsed since the ultrafast laser pulse, and it demonstrates that the atomic coherence has a sensitive dependence on the CEP of few-cycle laser pulses. The amplitude and the CEP dependences of the atomic coherence in Fig. 2 can readily be explained physically by perturbation theory in the small parameter $\epsilon=\Omega/\omega \ll 1$. Under the initial conditions $c_{1,2}(0)=0$ and $c_0(0)=1$, it is readily seen from Eq. (1) that $c_{1,2}(t)=O(\epsilon)$ and $c_0(t)=O(\epsilon^0)$, and hence $\rho_{12}=O(\epsilon^2)$, which clearly illustrates the feature that low Rabi frequencies induce less atomic coherence and hence are obviously nonfavorable from the viewpoint of the measurement. The lower limit for Rabi frequencies depends on the precision of the technique(s) in measuring the coherence. Cold alkali-metal atoms confined in a MOT instead of a single or a few hot atoms could increase the corresponding precision. Besides, noting that $\epsilon \sim E$ characterizes the electric field $E(t)$ having the period 2π for the CEP ϕ, the relation $\rho_{12}=O(\epsilon^2)$ implies that ρ_{12} should approximately have the period π for the CEP ϕ, just as clearly shown in Fig. 2.

On the other hand, we readily show that the three-level model is well justified in studying the atomic coherence $\rho_{12}=\rho_{21}^*=c_1c_2^*$ in the weak-field regime $\epsilon \ll 1$. The ultrashort pulse could couple more levels, say, levels $|j\rangle$ $(j=3,4,\ldots)$, besides the three considered levels $|j\rangle$ $(j=0,1,2)$. However, it can readily be shown that $c_{j\neq 0}(t)=O(\epsilon)$ and $c_0(t)=O(\epsilon^0)$ under the initial conditions $c_{j\neq 0}(0)=0$ and $c_0(0)=1$, and the $c_{j\geq 3}=O(\epsilon)$ introduce at most $O(\epsilon^3)$ contributions to ρ_{12}, already of the order $O(\epsilon^2)$ in the absence of the levels $|j\rangle$ $(j=3,4,\ldots)$, and hence these levels $|j\geq 3\rangle$ can be neglected so long as $\epsilon \ll 1$.

We would like to point out that, even for a periodically driven two-level system, any weak-field theories, so long as they are beyond the rotating-wave approximation, depend on the phase of the driving field under any fixed initial conditions for the amplitudes [41,42].

III. CARRIER-ENVELOPE PHASE-DEPENDENT QUANTUM BEATS

Let us now study the quantum beats due to the atomic coherence $\rho_{12}(t_0)=c_1(t_0)c_2^*(t_0)$ produced by a few-cycle ul-

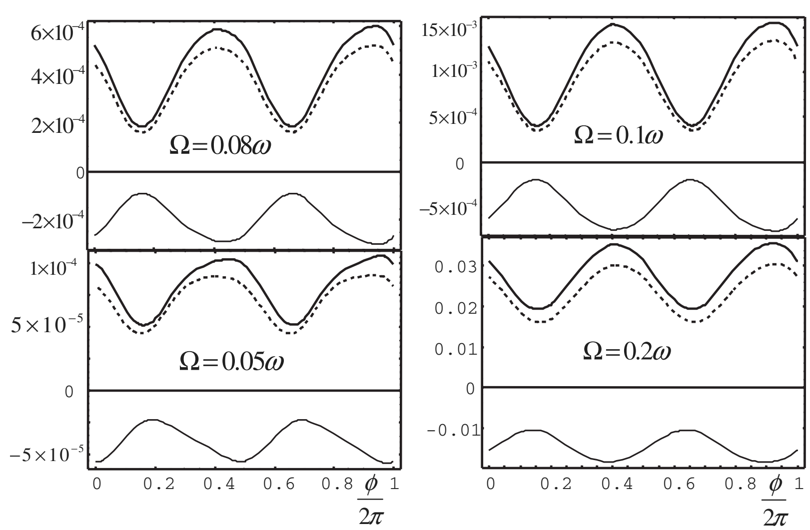

FIG. 2. Re $\rho_{12}(t_0)$ (dashed curves), Im $\rho_{12}(t_0)$ (thin solid curves), and $|\rho_{12}(t_0)|$ (thick solid curves) versus the scaled carrier-envelope phase $\phi/2\pi$ at the time $t=t_0=4\tau$, i.e., after the illumination with the few-cycle pulse of shape $\omega^{-1}\partial[e^{-(t-2\tau)^2/\tau^2}\sin(\omega t+\phi)]/\partial t$ with $\tau=5/\omega$ and $\omega=\omega_1$ for several values of the ratio Ω/ω designated on the diagrams, under the initial conditions $c_1(0)=c_2(0)=0$, and $c_0(0)=1$. Here $\rho_{jk}=c_jc_k^*$, and the other parameters are $\omega_2/\omega_1 \approx 1.05$ and $\alpha=\mu_{02}/\mu_{01}\approx 1.41$.

trafast pulse. Notice that the ultrafast pulse of width τ is applied to the three-level atom(s) in the time interval $t \in [0,t_0]$, and we have taken $t_0 \approx 4\tau$ throughout this paper. The phenomenon of quantum beats is investigated for the time interval $t > t_0$, i.e., its initial time is taken as $t=t_0$. Following the same procedure as Ref. [27], the quantum beat note $I_{qbn}=\langle\Psi(t)|\hat{E}_1^{(-)}(t)\hat{E}_2^{(+)}(t)|\Psi(t)\rangle$+c.c. can be calculated by the formula

$$I_{qbn} = \mathcal{E}_1\mathcal{E}_2[b_1(t)b_2^*(t)e^{i\omega_{21}(t-t_0)} + \text{c.c.}], \tag{2}$$

where $\omega_{21}=\omega_2-\omega_1$, $\hat{E}_1^{(-)}(t)=\mathcal{E}_1\hat{a}_1 e^{-i\omega_1(t-t_0)}$, $\hat{E}_2^{(+)}(t)=\mathcal{E}_2\hat{a}_2^\dagger e^{i\omega_2(t-t_0)}$, $\mathcal{E}_j=(\hbar\omega_j/2\varepsilon_0 V)^{1/2}$ is the electric field per photon for the mode j involving the transition $|0\rangle\leftrightarrow|j\rangle$ ($j=1,2$), and $\hat{a}_j$ and $\hat{a}_j^\dagger$ are the corresponding annihilation and creation operators, respectively. Here the system's state is taken as [27] $|\Psi(t)\rangle=\Sigma_{k=0,1,2}c_k(t)|k,0\rangle+b_1(t)|0,1_{\omega_1}\rangle+b_2(t)|0,1_{\omega_2}\rangle$. Here $|k,0\rangle$ denotes the atom in the atomic level $|k\rangle$ with no photon, while $|0,1_{\omega_j}\rangle$ describes the atom in its ground state $|0\rangle$ with one photon in the field mode j. Substituting the state $|\Psi(t)\rangle$ into the Schrödinger equation and using the Hamiltonian in the interaction picture $\hat{H}=\hbar\Sigma_{j=1,2}g_j(\hat{a}_j|j\rangle\langle 0|+\hat{a}_j^\dagger|0\rangle\langle j|)$ with the vacuum Rabi frequency $2g_j=\mu_{0j}\mathcal{E}_j/\hbar$ for the modes $j=1,2$ respectively, we readily obtain

$$i\left(\frac{d}{dt}+\gamma_j\right)c_j(t) = g_j b_j(t), \tag{3a}$$

$$i\frac{db_j(t)}{dt} = g_j c_j(t), \quad j=1,2, \tag{3b}$$

where the decay rate $2\gamma_j$ of the excited level j has been added phenomenologically [40]. The solution under the initial conditions of $b_1(t_0)=b_2(t_0)=0$ is $b_j(t)=c_j(t_0)s_j(t)$ with $s_j(t)=[g_j/\sqrt{\gamma_j^2-(2g_j)^2}](e^{\eta_{j+}(t-t_0)}-e^{\eta_{j-}(t-t_0)})$ and $2\eta_{j\pm}=-\gamma_j\pm\sqrt{\gamma_j^2-(2g_j)^2}$ ($j=1,2$). The conditions of $\gamma_j \gg 2g_j$ ($j=1,2$) are satisfied for the D lines of alkali-metal atoms [39] and hence $s_j(t)\approx(g_j/\gamma_j)(1-e^{-\gamma_j(t-t_0)})\approx g_j/\gamma_j$. Consequently, the quantum beat note in this case is of the form

$$I_{qbn} = I_0(\phi)\cos[\omega_{21}(t-t_0)+\theta(t_0)], \tag{4}$$

with $I_0(\phi)=(2\mathcal{E}_1\mathcal{E}_2 g_1 g_2/\gamma_1\gamma_2)|\rho_{12}(t_0)|$, and $\rho_{12}(t_0)=c_1(t_0)c_2^*(t_0)=|\rho_{12}(t_0)|e^{i\theta(t_0)}$. Here we have explicitly written the dependence of the amplitude $I_0(\phi)$ of the quantum beat on the CEP ϕ for convenience. $I_0(\phi)$ depends on the CEP ϕ through the sensitive CEP dependence of the coherence $|\rho_{12}(t_0)|$ as shown in Fig. 2.

The CEP dependence of the quantum beats revealed here is considerably greater than the CEP dependence of bound-state populations in the multiphoton ionization regime studied previously [20]. To see this point, let us define the depth of modulation in the amplitude signal of the quantum beats as $M=\{[I_0(\phi)]_{max}-[I_0(\phi)]_{min}\}/\{[I_0(\phi)]_{max}+[I_0(\phi)]_{min}\}/2$ or by using Eq. (4) $M=[|\rho_{12}(t_0)|_{max}-|\rho_{12}(t_0)|_{min}]/[|\rho_{12}(t_0)|_{max}+|\rho_{12}(t_0)|_{min}]/2$. It is seen from Fig. 2 that $M\sim 0.1-1$ ($M\approx 1/3$ for $\Omega=0.05\omega$) which is orders of magnitude larger than the depth of modulation in the total ionization signal [20].

IV. CONCLUSION

In summary, we have investigated the generation of bound-state atomic coherence by few-cycle laser pulses in the weak-field regime where ionization hardly takes place, and have mapped the atomic coherence thus produced onto the sensitive CEP-dependent signal of quantum beats. We believe that other quantum-interference phenomena besides the quantum beats due to the CEP-dependent atomic coherence would also show similar sensitive CEP dependence. It is pointed out that a relatively strong ultrashort pulse can be weakened, via passing through a strongly absorptive medium or in some other way, say, the beam splitting, to satisfy the weak-field condition, and hence its CEP information can also be extracted by the method described here.

ACKNOWLEDGMENTS

The work was supported by the NSF of China (Grant Nos. 10575040, 90503010, 60478029, and 10634060) and by the National Basic Research Program of China, Grant No. 2005CB724508.

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