Carrier-Envelope Phase Dependence of Few-Cycle Ultrashort Laser Pulse Propagation in a Polar Molecule Medium

Weifeng Yang,* Xiaohong Song, Shangqing Gong, Ya Cheng, and Zhizhan Xu†

State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China

(Received 12 March 2007; published 27 September 2007)

We theoretically investigate carrier-envelope phase dependence of few-cycle ultrashort laser pulse propagation in a polar molecule medium. Our results show that a soliton pulse can be generated during the two-photon resonant propagation of few-cycle pulse in the polar molecule medium. Moreover, the main features of the soliton pulse, such as pulse duration and intensity, depend crucially on the carrier-envelope phase of the incident pulse, which could be utilized to determine the carrier-envelope phase of a few-cycle ultrashort laser pulse from a mode-locked oscillator.

DOI: 10.1103/PhysRevLett.99.133602 PACS numbers: 42.50.Md, 33.80.Wz, 42.65.Re

Progress in femtosecond pulse generation has made it routine to generate optical pulses that are only a few cycles in duration [1–3]. The temporal variation of the electric field in such a pulse depends sensitively on the phase of the carrier frequency with respect to the envelope, i.e., the carrier-envelope phase (CEP). The CEP has significant influence on many nonlinear responses of matter to laser fields, ranging from the perturbative nonlinear optics regime [4] to the extreme strong-field nonlinear optics regime [5].

The abilities to measure and stabilize the CEP have therefore been a key point for laser-induced interactions. Using the powerful tools of frequency-domain laser stabilization (i.e., the so-called f-to-$2f$ spectral interference) method, a phase-stabilized and tunable few-cycle ultrashort laser pulse has been achieved in experiment [6]. However, direct measurement of the CEP is still very difficult so far. Usually, the CEP is extracted from the interaction of few-cycle laser pulses with atoms or other materials. In the strong-field regime, a well-known and state-of-the-art method is the stereo above-threshold ionization [7,8]. The strong left-right asymmetries of photoelectrons provide a very efficient means of measuring the CEP of few-cycle femtosecond laser pulses. Another promising candidate is the terahertz (THz) emission approach reported recently, which relies on the detection of the THz electromagnetic pulses emitted during photoionization of the air by the focused few-cycle pulses [9]. These measurements of CEP are in strong-field regime, and amplification will be necessary.

In fact, it is very meaningful to explore routines that are sensitive to the CEP and works with the direct output of a mode-locked oscillator (i.e., without amplification), because it may create a simpler technique for determining/controlling the comb offset frequency as has been stated in Ref. [10]. So far, the most promising pathway that has been reported with nonamplified few-cycle ultrashort laser pulses is the measurement of photocurrents from metal surfaces [11,12]. The strong dependence on the CEP is partly due to the fact that the surface breaks the inversion symmetry for laser-matter interaction. Phase-dependent effects in nonperturbative resonant extreme nonlinear optics regime, exemplified by carrier-wave Rabi flopping, have been predicted [13] and observed experimentally [14]. The carrier-wave Rabi flopping was first reported by Hughes [15], who discussed few-cycle ultrashort laser pulse propagating in an ensemble of identical two-level systems in vacuum, and demonstrated later experimentally by Mücke *et al.* [16]. Its phase dependence relies on the interference of different Rabi sidebands. However, this method is insensitive to absolute phase shift of π due to the inversion symmetry, which will result in the sign ambiguity of the electric field.

In this Letter, we present a new approach for CEP determination of nonamplified few-cycle ultrashort laser pulses on nonperturbative two-photon resonant extreme nonlinear optics in polar molecule media. This method is based on the detection of the duration or the intensity of the soliton pulse generated during the course of propagation. We show that, very interestingly, the information of the CEP of the initial incident pulse can be ''remembered'' by the generated soliton pulse, which provides a direct, time-domain access to the CEP related phenomena using two-photon process.

We employ the polar media in our simulation for several reasons. First, the enhancement of multiphoton excitations can be achieved due to the nonzero permanent dipole moments (PDM) in polar media as has been demonstrated in low intensity scheme in our previous work [17] and other experimental and theoretical works [18–20]. Thus, when the two-photon resonant excitation is considered, the macroscopic coherent polarization $P(t)$ built up in the medium will be enhanced correspondingly, and it acting as a source of a reemitted field $E_{\mathrm{em}}(t)$ may generate a soliton pulse during the propagation. Because of the transient character of the coherent polarization, the generated pulse will carry some information of the incident few-cycle laser pulse and the medium. Second, for polar media, the inversion sym-

metry is broken that may eliminate the $\pm\pi$ phase ambiguity [21,22] and allows one to determine the CEP.

The propagation properties of a few-cycle ultrashort pulse in the polar molecule medium can be modeled by the Maxwell-Bloch equations beyond the slowly varying-envelope approximation (SVEA) and rotating wave approximation (RWA). The approach we used for solving the full-wave Maxwell-Bloch equations is the finite-difference time-domain (FDTD) method which has been proven to be an accurate *ab initio* tool to simulate the interaction between few-cycle ultrashort pulse and matter [13,15,16,23]. The numerical analyses are based on molecular parameters characteristic of the $S_0 \rightarrow S_1$ electronic transition in 1-[*p*-(*N*, *N*-dimethylamino)phenyl]-4-(*p*-nitrophenyl)-1, 3-butadiene which was involved in previous experimental [24] and theoretical [25] investigations of the effects of PDM on pulse interaction. In atomic units, the molecular parameters are $\omega_0 = 8.59 \times 10^{-2}$, $\mu_{21} = 3.93$, and $d = 1.18 \times 10$, where ω_0 is the transition frequency between the ground and the excited states of the polar molecule medium, μ_{ij} are the dipole moment matrix elements, and $d = \mu_{22} - \mu_{11}$ is the difference in the PDM. The polar molecules are supposed to orient along the laser polarization [26]. The initial field with a hyperbolic secant shape is defined as $E_x(t) = E_0 \mathrm{sech}[(t - t_0)/\tau_0]\cos[\omega_l(t - t_0) + \phi]$, where ω_l is the central frequency and ϕ is the CEP of the incident pulse. $\tau_p = 2ar\cosh(1/\sqrt{0.5})\tau_0$ is the full width at half maximum (FWHM) of the pulse envelope, which is set as 5 fs here. The envelope area of the pulse is determined by $A(z, t = 0) = (\mu_{21}/\hbar)\int_{-\infty}^{\infty}\tilde{E}(z, t')dt' = \frac{\mu_{21}E_0\tau_0\pi}{1.76\hbar}$. A 4π pulse (corresponding to a peak intensity 2.64×10^{12} W/cm^2) with the condition of two-photon resonance ($\omega_l = \omega_0/2$) is considered in the numerical calculations. A macroscopic coherent polarization $P_x(t)$ builds up gradually in the medium driven by the incident field. The reemitted field $E_{\mathrm{em}}(t)$ which is proportional to the time derivative of the macroscopic polarization, i.e., $E_{\mathrm{em}}(t) \propto -\frac{\partial P_x}{\partial t}$ [27], reflects directly the response of the medium. Although we only present the simulation results with the above-mentioned parameters, the results shown below are also valid for other combinations of laser pulses and asymmetric media with nonzero PDM.

Figures 1(a) and 1(b) show the electric fields of the incident few-cycle ultrashort laser pulses with CEPs $\phi = 0$ and $\phi = \pi$, respectively. The temporal variation of the electric field depends sensitively on the CEP. After some propagation distance, a soliton pulse is generated in both cases [see Figs. 1(c) and 1(d)]. Moreover, the main features

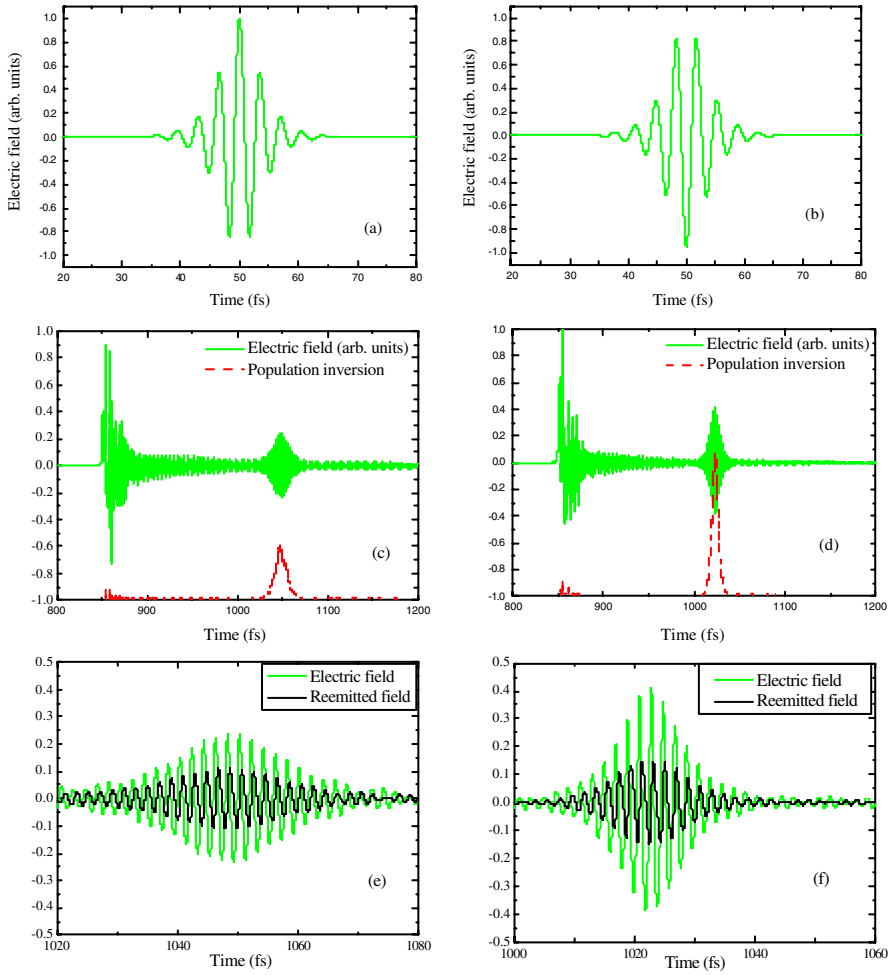

FIG. 1 (color online). The electric field of the incident 4π pulse with CEP (a) 0 and (b) π. (c), (d) The corresponding time dependent electric field and population inversion at the distance of 240 μm in the polar molecule medium. (e), (f) The comparison of electric field and corresponding reemitted field.

of the soliton pulse in each case are very different. The duration of the generated soliton pulse is longer while the intensity is lower for the incident pulse's CEP $\phi = 0$ than that for CEP $\phi = \pi$.

The basic physical mechanism of the soliton generation is that the medium absorbs and emits photons and redistributes energy in the pulse during the course of pulse propagation. The propagating pulse is altered in shape until it reaches a stable status, and a self-induced transparency (SIT) soliton pulse is generated [28]. By comparing the time dependent electric field of the generated soliton pulse and the reemitted field [see Figs. 1(e) and 1(f)], it can be seen that the characteristics of the soliton pulse, including the oscillation frequency of the electric field and CEP dependence of the duration and the intensity, are consistent with those of the reemitted field. This demonstrates that the soliton pulse generation is indeed a result of the response of the medium. The duration of the generated soliton pulse is determined not only by the incident pulse but also by the response time of the medium, which explains why the duration of the generated soliton pulse is longer than that of the incident pulse. Pulse retardation occurs because the velocity of SIT soliton pulse is much slower than that of the main pulse which can hardly induce any energy exchange between the pulse and the medium due to the weakened two-photon transition [see Figs. 1(c) and 1(d)]. Hence, the soliton pulse can break up from the main pulse.

The physical properties of PDM play an essential role in the whole process. First, media with permanent dipoles have energy levels with mixed parity and it is known that direct two-photon transitions can be allowed in these media [29]. Hence, when the condition of two-photon resonance is satisfied, the medium can absorb and emit photons and further induce the generation of the SIT solution pulse. Second, previous investigations have demonstrated that nonlinear interaction is stronger when the electric field at the peak of pulse is antiparallel to the PDM in asymmetric molecule systems [21,22]. In our case, the relationship between the peak direction of the electric field and the PDM affects directly the responses of the medium to the incident few-cycle laser pulse. In fact, it is a process of translating the CEP dependent two-photon transition into the CEP dependent soliton pulse generation in time domain. Hence, very interestingly, though the CEP of the pulse is changing during the evolution (see Fig. 1), the information of the initial CEP of the incident pulse can be remembered by the propagating soliton pulse.

Figures 2(a) and 2(b) display the FWHM and the intensity of the generated soliton pulse profile as a function of the initial CEP of the incident ultrashort laser pulse, respectively. It is found that the FWHM is directly related to the initial CEP of the incident ultrashort laser pulse with a nearly sinelike dependence, while the intensity is related with a nearly cosinelike dependence. Using this method, the CEP shift of π can be clearly determined. Moreover, because the slopes of the two sides of the curves around π are inverted, our approach can make a complete CEP determining without any ambiguity. Therefore, if the intensity (or pulse duration) is calibrated, these effects might have potential applications in determining the CEP of few-cycle ultrashort pulses directly from a mode-locked oscillator.

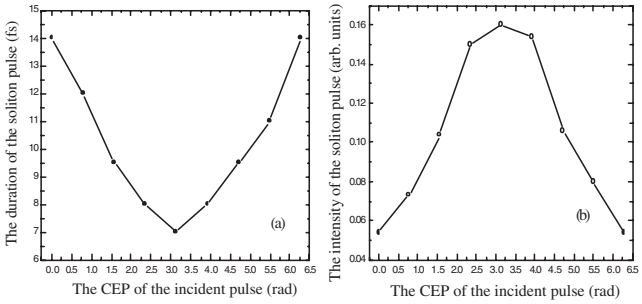

FIG. 2. (a) The FWHM and (b) the intensity of the soliton pulse as a function of the CEP of the incident pulse at $z = 240$ μm. The lines are a guide to the eye.

To make this approach feasible , the soliton pulse should be stabilized. Figure 3 shows the evolution of the generated soliton pulse propagating in the polar molecule medium when the CEP of the incident pulse is $\phi = 0$. It can be seen that the temporal shape of the soliton pulse is almost invariable after it was broken up from the main pulse. Similar results can be obtained for the cases of other CEPs. Moreover, the soliton pulse can be filtered out from the main pulse because of the frequency difference and the time delay between them. Furthermore, although the incident pulse duration is only 5 fs, the pulse duration of the generated soliton is much longer, and measuring the intensity of the generated soliton pulse can also provide the CEP information. More importantly, it should be pointed

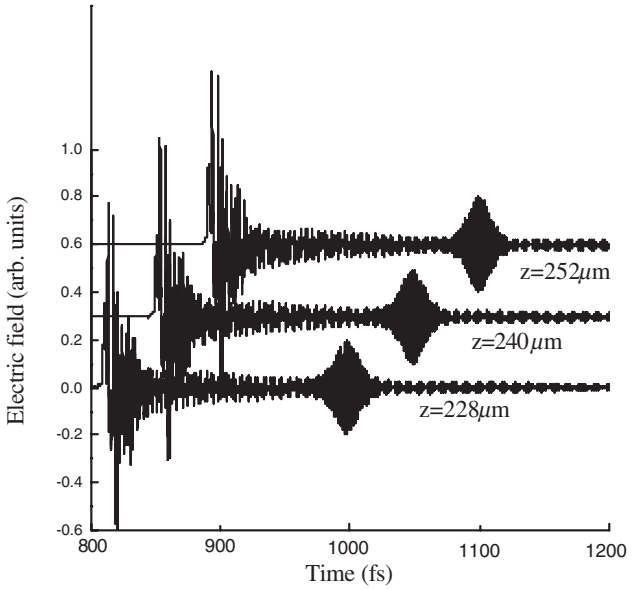

FIG. 3. The evolution of the few-cycle ultrashort pulse in the polar molecule medium at respective propagation distances of $z = 228$ μm, $z = 240$ μm, and $z = 252$ μm.

out that our result is not limited to the certain dipole molecule, but can be extended to other media which have nonzero PDM [18,30]. Based on these facts, our approach appears to be experimentally feasible.

In summary, we have theoretically analyzed the CEP dependence of the soliton pulse induced by few-cycle ultrashort pulse propagating in a polar molecule medium. The duration and the intensity of the soliton pulse depend sensitively on the CEP of incident excitation pulse. It is a very exciting phenomenon that depicts a new physical mechanism of the response of the medium to the incident few-cycle ultrashort pulse in the propagation process, i.e., translating the CEP dependent two-photon transition into the detectable time-domain pulse evolution under the moderate laser intensity, which would be beneficial to the CEP measurement and its applications.

We would like to thank Professor Ruxin Li and Dr. Zhinan Zeng for their valuable discussion of the physics. The work was supported by the National Basic Research Program of China (Grant No. 2006CB806000) and the National Natural Science Foundation of China (Grants No. 60608001 and No. 10523003).

*wfeng_yang@yahoo.com.cn
†zzxu@mail.shcnc.ac.cn

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