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Carrier–envelope phase effects for a dipolar molecule interacting with ultrashort laser pulse*

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In this paper the phase-dependent features of ultrashort laser pulse resonant propagation in a two-level dipolar molecule are demonstrated by solving full Maxwell–Bloch equations. The electronic properties of dipolar molecule 4-trans-[p-(N, N-Di-n-butylamino)-p -stilbenylvinyl] pyridine (DBASVP) molecule, one-dimensional asymmetric organic molecule, is calculated by density functional theory at *ab initio* level. The numerical results show that the carrier propagation and the spectrum evolution of the pulse are sensitive to its initial phase and the phase sensitivity is more obvious for larger area pulse. The phase-dependent feature is more evident in dipolar molecule because the permanent dipole moment makes the nonlinear effects stronger.

Keywords: carrier–envelope phase, self-phase modulation, Maxwell–Bloch equations
PACC: 4265, 3120A

1. Introduction

Recent progress in ultrafast optics has allowed the generation of ultra-intense light pulse comprising merely a few field oscillation cycles,[1,2] which brings about great challenges for experimentalists and theorists. For an ultrashort pulse, the value of the initial phase of the carrier wave with respect to the pulse envelope (the so-called carrier–envelope phase, CEP) becomes important in the interaction of intense few-cycle radiation with matter.[3,4] The reason is that all effects generated by means of laser pulses mainly depend on the time variation of their electromagnetic fields and thus on the absolute phase provided that the pulses are really short. Although it has been known for years (see, for example, Ref.[5]), the first CEP effect was only recently detected with the randomly fluctuating CEP.[4] It has been demonstrated that the CEP is important to many nonlinear progresses such as high harmonic generation,[6] atomic ionization,[7,8] final molecular state population[9] and generation of attosecond pulses.[10]. By exploiting the phase sensitivity of strong-field processes, it might become feasible to measure and stabilize the absolute phase of few-cycle light generated by a mode-locked laser, which opens up ways to full control of high-field interactions with never-before-achieved precision.[11]

In the last few years there has been considerable interest in the study of the interaction of ultrashort laser pulses with a two-level medium.[12−18] Study on the propagation of laser pulses was previously carried out under the assumption that the diagonal matrix elements of the dipole moment operator are equal to zero. This is the typical situation for many practical media. However, in recent years, one found that many organic molecules have strong nonlinear properties such as two-photon absorption.[19−23] Theoretical and experimental studies have shown that this kind of molecule has a few charge-transfer (CT) states in the visible or ultraviolet region; in the special case, the one-dimensional organic molecules normally has one CT state.[20] Therefore, the optical response of the molecules in the visible or ultraviolet region can be described by a few-state model. Furthermore, the one-dimensional asymmetric organic molecules have permanent dipole moments. The interaction between ultrashort laser pulses and a dipolar molecular medium is thus an interesting issue. When the interaction of continuous wave or laser pulses with dipolar molecule were considered, it was found that

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the permanent dipole moments have substantial effects on the molecular-state population since the selection rules and the molecular–laser coupling can be greatly modified.[24−28]

In the present work, we choose the dipolar molecule 4-trans-[p-(N, N-Di-n- butylamino)-p-stilbenylvinyl] pyridine (DBASVP) molecule (see Fig.1) studied in our previous work[19] as the model and simulate the resonant propagation of femtosecond pulse with different initial phases by using the full Maxwell–Bloch equations beyond the limits of approximations including slowly-varying-envelope approximation and the rotation-wave approximation. We will show that the propagation of ultrashort pulses in dipolar molecule is dependent on the initial phase and that the corresponding spectra show different properties. As field strength increases, the phase sensitivity becomes stronger.

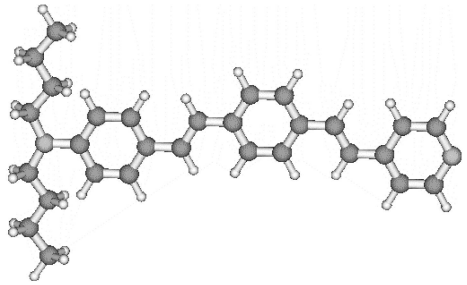


Fig.1. Structure of DBASVP molecule.

2. Theoretical model

Let the incident electromagnetic field to be polarized along the x axis and propagate along the z axis to an input interface of the medium at z=0. Maxwell's equations take the form of $\frac{\partial E_x}{\partial z} = -\mu_0 \frac{\partial H_y}{\partial t}$ and $\frac{\partial H_y}{\partial z} = -\frac{\partial P_x}{\partial t} - \varepsilon_0 \frac{\partial E_x}{\partial t}$, where μ_0 and ε_0 are the permeability and permittivity of free space.

Considering the one-dimensional structure of the DBASVP molecule, we can write the Bloch equations as follows:

$$\begin{aligned}
\frac{\partial u}{\partial t} &= -\gamma_{12} u - \omega_0 v + \frac{\Delta\mu}{\hbar} E_x v, \\
\frac{\partial v}{\partial t} &= -\gamma_{21} v + \omega_0 u + 2\frac{\mu E_x}{\hbar} d - \frac{\Delta\mu}{\hbar} E_x u, \\
\frac{\partial d}{\partial t} &= -\gamma_{22}(d+1) - 2\frac{\mu E_x}{\hbar} v,
\end{aligned} \tag{1}$$

where ω_0 equals the excited energy for molecule, and γ_{22} and $\gamma_{12}(\gamma_{21})$ are respective population and polarization relaxation constants. The difference of the permanent dipole moment of ground and excited states is denoted by $\Delta\mu$. μ is the transition dipole moment between them. u and v are related to the off-diagonal density matrix element of a two-level molecule by definition of $\rho_{12} = (u + \mathrm{i}v)/2$ and d represents the population difference of the two states.

The molecule is assumed to be rest in its ground state when there is no interaction between laser and medium. We choose the initial field with a hyperbolic-secant shape, $E_x(z, t = 0) = F\mathrm{sech}[1.76(z/c + z_0/c)/\tau_\mathrm{p}] \cos[\omega_\mathrm{p}(z + z_0)/c + \varphi]$. Where F is the peak of input electric field, φ is the initial phase and τ_p is the full width at half maximum of the pulse intensity envelope, here it is taken to be 2fs. The frequency of the electric field ω_p is equal to ω_0 in resonant case. The choice of z_0 ensures that the pulse penetrates negligibly into the medium at $t = 0$. The decay rates for molecular system can be chosen as $\gamma_{12} = \gamma_{22} = 1.0 \times 10^{12}/\mathrm{s}$.[29] The molecular density is taken as $N = 7.0 \times 10^{25}/\mathrm{m}^3$.

The electronic properties of the molecule are calculated by density functional theory.[19] There is only one CT state for the molecule in visible region. The molecule can then be modelled as a two-level medium when we deal with the propagation of pulse in it. The excited energy of the CT state is 3.05eV, the transition dipole moment $\mu = 4.18\times10^{-29}$Asm, and the difference of the moments of the two states is $\Delta\mu$=2.59×10^{-29}Asm. The envelope area of the pulse is determined by $A(z) = (\mu/\hbar)\int_{-\infty}^{\infty} E_0(z, t')\mathrm{d}t' = \frac{\mu F \tau_\mathrm{p} \pi}{1.76\hbar}$. The Maxwell–Bloch equation is solved by using the finite-difference time-domain method for the fields and the predictor-corrector method[15] for the material variables. The time and space increments Δt and Δz are chosen to ensure $c\Delta t \leq \Delta z$.

3. Results and discussion

Firstly we simulate the propagation of 2π pulses with initial phases of $\varphi = 0$ (solid line) and $\varphi = \pi/2$ (dashed line) in molecular medium (see Fig.2). At initial position $z = 0\mu$m, the two pulses are different (see Fig.2(a)). When the electric field of the pulse with $\varphi = 0$ reaches its maximum, the field with $\varphi = \pi/2$ is zero. If the pulse consists of much fewer field oscillation cycles such as single-cycle laser pulse, the difference will be more obvious.[30] This difference gives rise to different maximum intensity of initial two pulses, while the intensity determines the self-phase modulation (SPM) effects in the propagation of the pulse.

Figure 2(b) presents the carriers at propagation distance of z=16μm. It shows that the main pulses still maintain the difference except for the minor solitons with higher frequency ahead of the main pulses. As has been noted,[31] the small soliton propagates faster than the main pulses due to the self-induced transparency of the main pulse. More or less, the phase-dependent feature is trivial for the 2π pulse propagation.

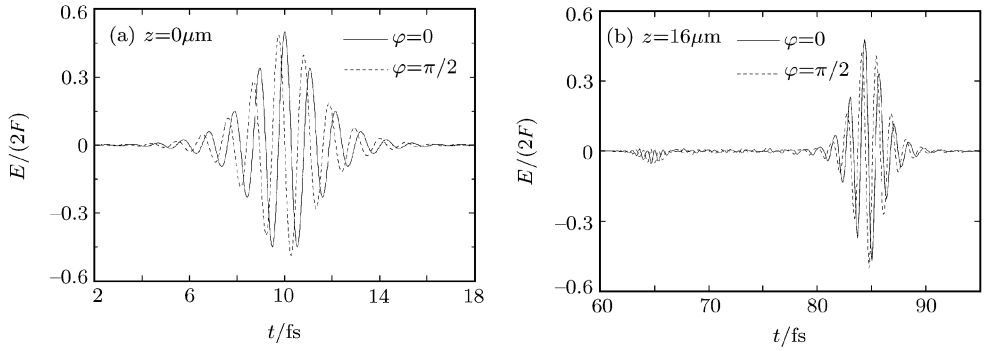


Fig.2. Carriers of the two 2π pulses with different initial phases of $\varphi = 0$ (solid line) and $\varphi = \pi/2$ (dashed line) at $z = 0\mu$m (a) and 16μm (b).

Figures 3(a) and (b) present the carriers of the 4π pulses with different initial phases in dipolar molecule at $z = 16\mu$m. In Fig.3(a), we can see that each pulse evolves to two 2π sub-pulses as a consequence of two Rabi floppings.[32] Figure 3(b) is an enlargement of the foregoing sub-pulses in Fig.3(a). It shows that the intensities of the two carrier fields are different, which results in different SPM of them. Hence, the phase difference of the two sub-pulses is not $\pi/2$, but it is larger than $\pi/2$ in the leading edges and less than $\pi/2$ in the trailing edges. Correspondingly, the carrier is reshaped and deviates from a cosine function. For the second sub-pulses, however, one can see that the phase difference is about π. In our previous work, it demonstrated that the permanent dipole moment $\Delta\mu$ causes the splitting of the two sub-pulses quicker and brings about stronger nonlinear effects compared with nondipolar medium.[26] In order to illustrate the dipole effects, we also plot the pulses propagating in pseudo-molecule medium by artificially setting $\Delta\mu = 0$ (see Fig.3(c)). It is interesting to note that the phase difference for the behind weaker sub-pulses still maintain nearly $\pi/2$. Therefore we can draw a conclusion that the phase dependence of pulses propagation in dipolar molecule is more evident than that in nondipolar molecular medium.

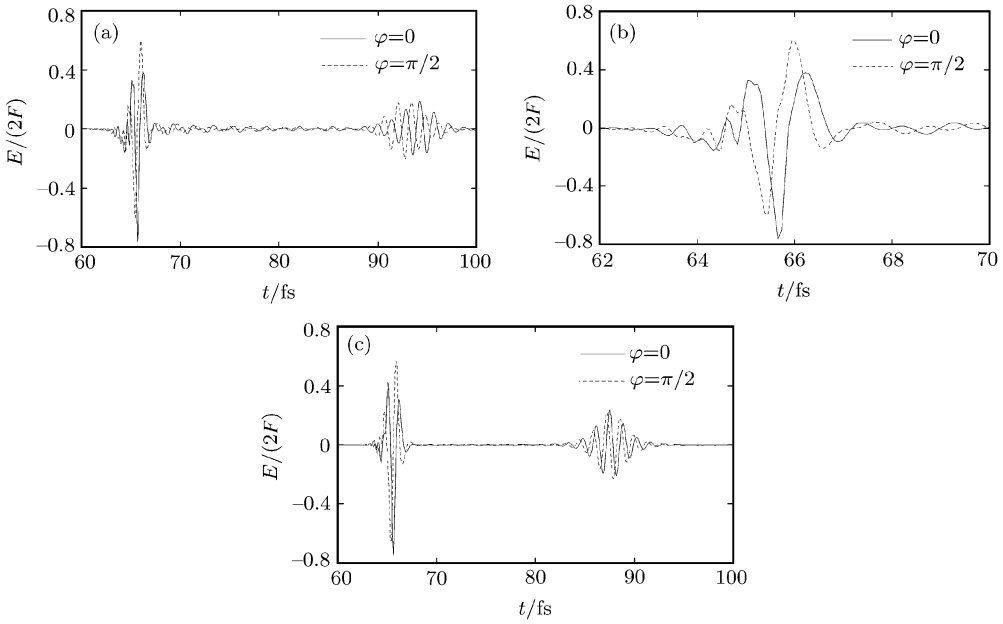


Fig.3. (a) Carriers of the 4π pulses with different initial phases of $\varphi = 0$ (solid line) and $\varphi = \pi/2$ (dashed line) at $z = 16\mu$m. (b) An enlargement of (a) from 62fs to 70fs. (c) The same as (a) only by taking $\Delta\mu = 0$ for molecule.

Figure 4 shows the corresponding spectra of 4π laser pulses with initial phases of 0 and $\pi/2$. Both the spectra of 4π pulses are broadened and lower and higher frequency appear mainly due to SPM. However, the lower and higher frequency spectra have different features for pulses with different initial phases. For instance, with initial phase of $\pi/2$, the second harmonic frequency disappears. The situation is understandable since the carriers with different initial phases are reshaped differently.

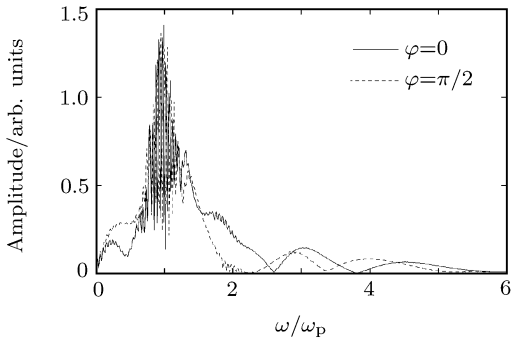


Fig.4. The spectra of 4π pulses with different initial phases of $\varphi = 0$ (solid line) and $\varphi = \pi/2$ (dashed line) at $z = 16\mu$m.

The difference for the evolution of the propagation pulses with different initial phases is more evident for the 8π pulses (see Fig.5). From Fig.5(a), one can see that the evolution of each pulse has specific profile, and the separation of some sub-pulses for pulse with $\varphi = 0$ is quite different from that for pulse with $\varphi = \pi/2$. If the molecule is assumed as nondipolar one, namely taking $\Delta\mu = 0$, the evolutions of pulses with 0 and $\pi/2$ shown in Fig.5(b) take the similar profile except for the existence of phase difference. The permanent dipole moment is demonstrated again to have strong effect on the phase dependence of the evolution of ultrashort pulse. The corresponding spectra of Fig.5(a) (see Fig.5(c)) are broadened and show oscillatory features. Besides this, the phase effect on the spectrum evolution is clearly displayed. For larger area pulses such as 8π, it has been found that the carrier-wave Rabi flopping (CWRF) will occur and result in highly reshaping.[27] The CWRF and SPM reshape the carrier together and the CWRF plays a more important role than SPM for 8π pulse. Since the carriers with different initial phases have different distribution, the reshaping of the pulses will then be different. As the intensity of field increases, the modulation of nonlinear effects become stronger and thus the phase dependence is enhanced. Because $\Delta\mu$ can bring about much more nonlinear properties, the phase sensitivity in dipolar molecule is more obvious than that in nondipolar molecular medium.

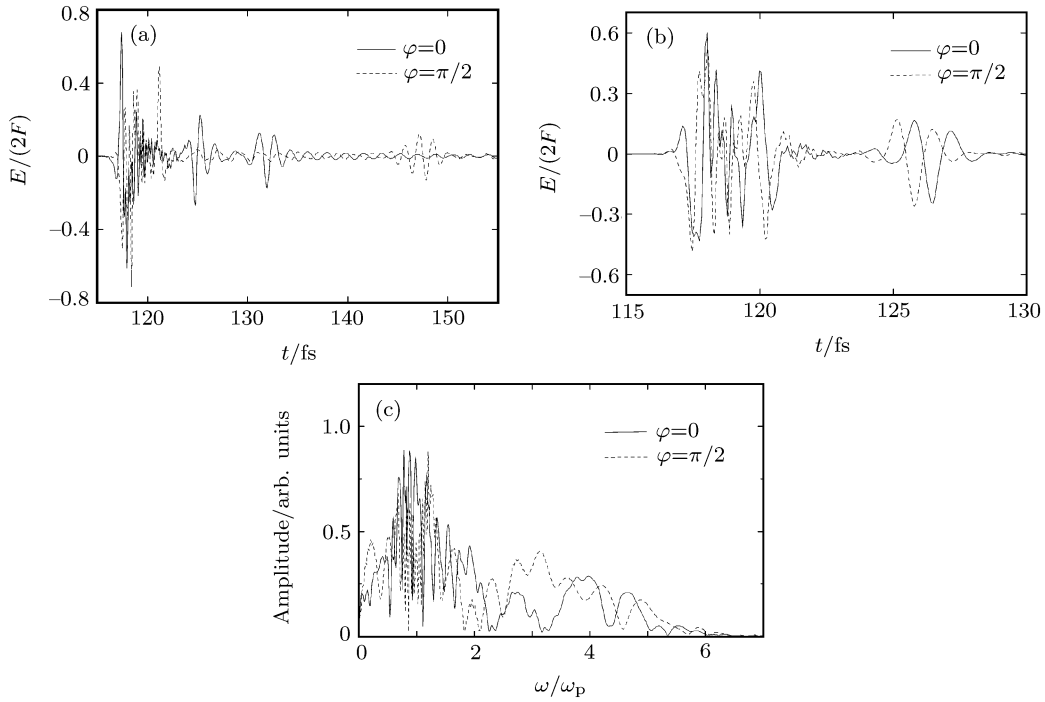


Fig.5. (a) Carriers of the 8π pulses with different initial phases of $\varphi = 0$ (solid line) and $\varphi = \pi/2$ (dashed line) at $z = 32\mu$m. (b) The same as (a) only by taking $\Delta\mu = 0$ for molecule. (c) The spectra corresponding to (a).

It is noted that the phase dependence of evolution of ultrashort pulses in dipolar molecular medium is more obvious than that in nondipolar molecular medium. This feature can be explained as follows. As shown before, the dipolar molecule has strong behaviour in both one-photon and two-photon absorption when it is excited from ground to CT states.[19] Therefore, the dipolar molecule shows stronger nonlinear optical properties compared with the corresponding nondipolar molecule. Accordingly, the CWRF and SPM caused by nonlinear optical properties result in more highly reshaping of pulse propagating in dipolar molecule, thus, the phase-dependent feature is enhanced.

4. Conclusion

In conclusion, by solving the full Maxwell–Bloch equations, we have studied the propagations of ultrashort laser pulses with different initial phases in DBASVP molecule modelled as a two-level dipolar molecular medium. The numerical results show that the carrier and the spectrum of the pulse are sensitive to its initial phase due to SPM and CWRF and the phase sensitivity is more obvious for larger area pulse. By comparing with propagation of the pulse in nondipolar molecule, it is shown that the phase-dependent feature is more evident in the dipolar molecule because the permanent dipole moment makes the nonlinear effects stronger.

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