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Resonant propagation of femtosecond laser pulse in DBASVP molecule: one-dimensional asymmetric organic molecule*

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We have investigated the resonant propagation of femtosecond laser pulse in 4-trans-[p-(N, N-Di-n-butylamino)-p-stilbenyl vinyl] pyridine medium with permanent dipole moments. The electronic structures and parameters for the compound have been calculated by using density functional theory. In the optical regime, there is one charge-transfer state, and the molecule can thus be simplified as a two-level system. Both the one- and two-photon transitions occur between the ground and charge-transfer states. The numerical results show that the permanent dipole moments have an obvious effect on the propagation of the ultrashort pulse laser. The ideal self-induced transparency disappears for 2π pulse, and second harmonic spectral components occur significantly due to the two-photon absorption process. For the 6π pulse, continuum frequency generation is produced and a shorter duration pulse in time domain with 465 as is obtained.

Keywords: Maxwell–Bloch equations, Rabi flopping, two-photon absorption
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1. Introduction

Currently, using the mode-locking (Kerr-lens-mode-locking) technique, one can obtain femtosecond laser pulse that consists of only a few optical cycles in the visible spectrum.[1] Combined with the chirped pulse amplification technique, the pulse energy has increased significantly. The femtosecond pulses are finding application in many diverse fields. The coherent control of chemical reactions, the acceleration of particles and the generation of coherent soft-x-ray radiation are a few examples mentioned here.

The availability of the ultrashort coherent lasers has led to many interesting and challenging problems involving short pulse propagation. The propagation of high intense laser in two-level medium gives rise to strong nonlinear effects such as self-phase modulation (SPM), carrier-wave Rabi flopping (CWRF) and high-order harmonic generation (HHG). Kasparian *et al* observed the infrared extension of the supercontinuum generated by femetosecond terawatt laser pulses propagating in the atmosphere.[2] It was demonstrated that the self-phase modulation plays an important role in it. Hughes stimulated the propagation of a 18fs pulse propagating through a two-level medium.[3] He proposed that the self-induced transparency (SIT) results are still recovered with minor modifications for small area laser pulse. However, for large area laser pulse, the area under individual carriers might themselves cause Rabi flopping, i.e., CWRF, which can lead to carrier-wave reshaping and significantly higher spectral components and even soft-x-ray generation.[4] Moreover, the femetosecond (fs) laser pulse that propagates in a medium with linear Lorentz response in a Kerr nonlinearity shows evident carrier shock, which is characterized by a strong third harmonic pulse.[5] Xiao *et al* studied theoretically the spectrum evolution of a 5fs pulse in a resonant two-level atom medium.

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They found that ideal SIT cannot occur for a 2π pulse, and for large pulse area continuum spectrum can be generated.[6]

Up to now, most studies in this area have been related to the two-level atom medium without permanent dipole moments.[7−9] When the interaction of the continuous wave laser and dipolar molecule was considered, it was found that 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.[10,11] Recently, Brown *et al* have derived analytical results with the rotation-wave approximation (RWA) for the interaction of a single pulsed laser with a two-level dipolar molecule.[12] However, the propagation properties of a few-cycle pulse laser in a two-level dipolar medium have rarely been investigated. The dipolar effects on the evolution of ultrashort pulse laser in both time and frequency domains are then hardly explored.

Recently, an organic compound, 4-trans-[p-(N, N-Di-n-butylamino)-p-stilbenyl vinyl] pyridine (DBASVP) (see Fig.1), has been designed.[13] The experimental and theoretical results[13,14] show that the molecule has strong nonlinear optical properties, in particular the high two-photon absorption cross section. As a one-dimensional (1D) asymmetric molecule, the compound has only one charge-transfer (CT) state in optical regime. Its optical and nonlinear optical properties can thus be well described by a two-state model with the ground and the CT states.[14] Furthermore, the permanent dipole moment difference of the ground and CT states is quite large. Therefore, when ultrashort pulse lasers propagate in the molecular medium, the medium can be simplified as a two-level system with permanent dipole moments.

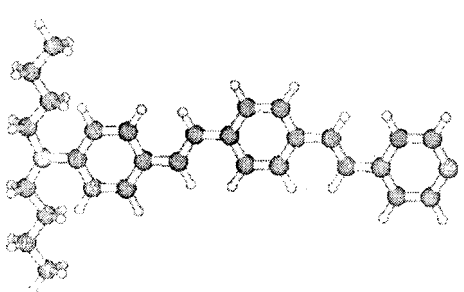


Fig.1. Structure of DBASVP molecule.

This paper is organized as follows. In Section 2, we present a theoretical model of the interaction of fs pulse with a two-level medium possessing permanent dipole moments. In Section 3, propagations of pulse and spectrum evolution for various input pulse areas are numerically investigated. Finally, conclusions are given in Section 4.

2. Theoretical model

We assume that the incident electromagnetic field is polarized along the x axis and propagates 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 1D structure of the DBASVP molecule, the Bloch equations can be written 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 denotes the excited energy for the molecule, and γ_{22} and $\gamma_{12}(\gamma_{21})$ are respectively population and polarization relaxation constants. The difference between the permanent dipole moments 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 $\rho_{12} = (u + \mathrm{i}v)/2$, and d represents the population difference of the two states.

The molecule is assumed to be at rest in its ground state when there is no interaction between laser and medium. We choose the initial field with a hyperbolic-secant shape, $E_x(z, t = 0) = F\mathrm{sech}[1.76(z/c + z_0/c)/\tau_\mathrm{p}] \cos[\omega_\mathrm{p}(z + z_0)/c]$, where F is the peak of the input electric field, τ_p is the full width at half maximum (FWHM) of the pulse intensity envelope, here taking it to be 2fs. The choice of z_0 should ensure that the pulse penetrates negligibly into the medium at $t = 0$. In Eq.(1) the decay rates for the molecular system can be chosen to be $\gamma_{12} = \gamma_{22} = 1.0 \times 10^{12}$/s. The molecular density is taken to be $N = 7.0 \times 10^{25}/\mathrm{m}^3$. By using density functional theory, we have calculated the electronic properties of the molecule.[14] The excited energy is 3.05eV , the transition dipole moment $\mu = 4.18\times10^{-29}$Asm, and the difference of dipole moments of the two states $\Delta\mu$=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 and

Bloch equations can be 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

We first model a 2π pulse propagating in the dipolar DBASVP molecule. The carrier waves and the corresponding spectra at several propagating distances are shown in Fig.2. The carrier wave is modulated clearly and a little sub-pulse with faster oscillation appears ahead of the main pulse (see Fig.2(a)). With further propagation, the separation of the sub-pulse from the main pulse becomes larger because the main pulse propagates slower due to SIT. Compared with the results for a 2π pulse propagating in a two-level atom medium,[3,6] one can find that the ideal SIT for a 2π pulse is further destroyed by the permanent dipole moment. Moreover, lower and higher spectral components appear for the pulse in the frequency domain shown in Fig.2(b). It is interesting to note that the second harmonic component occurs in the dipolar molecular medium besides the odd components, while only odd higher harmonic components appear in the non-dipolar atom medium. Since there exists strong two-photon absorption between the ground and CT states of the molecule, the second harmonic component is thus contributed by the two-photon absorption process. Even through the two-photon absorption is non-resonant, the second harmonic component is still obvious because of the strong intensity of the pulse and the strong two-photon absorption cross section of the molecule. Additionally, with the pulse propagating further, these additional spectral components tend to show more evident oscillation features.

Figure 3 presents a 4π pulse resonantly propagating in DBASVP molecule at distances of 4 and 12μm. Because the $2n\pi$ (n is an integer) pulse is unstable[16] due to multiple Rabi floppings (RF), the 4π pulse will evolve into two 2π pulses. The first sub-pulse is compressed and the second one is broadened. We can see that the separation between the two sub-pulses becomes larger and larger with the propagation going further because the group-velocity of pulse is a quadratic function of the pulse amplitude.[17] The faster oscillation terms occur in the middle of two separated pulses, and the second RF is incomplete compared with the case of non-dipolar medium.[18]

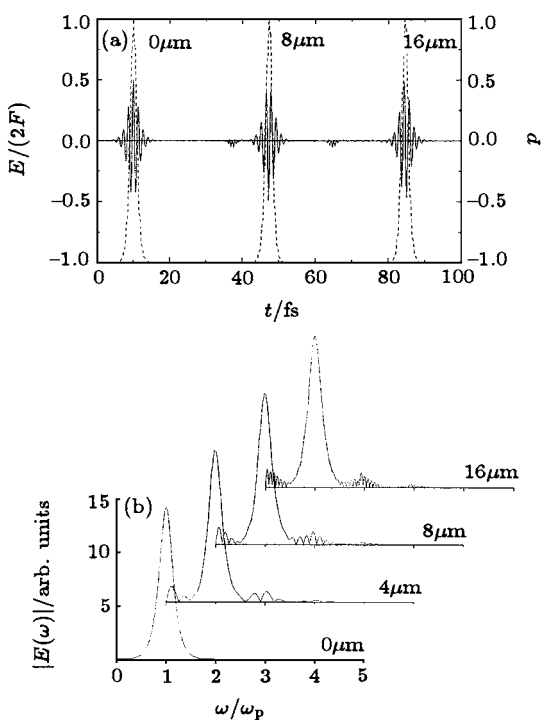


Fig.2. The 2π pulse propagation through the DBASVP medium. (a) The carrier wave of the pulse (solid line) and the population difference d (dashed line) at the respective propagation distances of 0, 8 and 16μm. (b) The corresponding spectrum evolution at z =0, 4, 8 and 16μm, respectively.

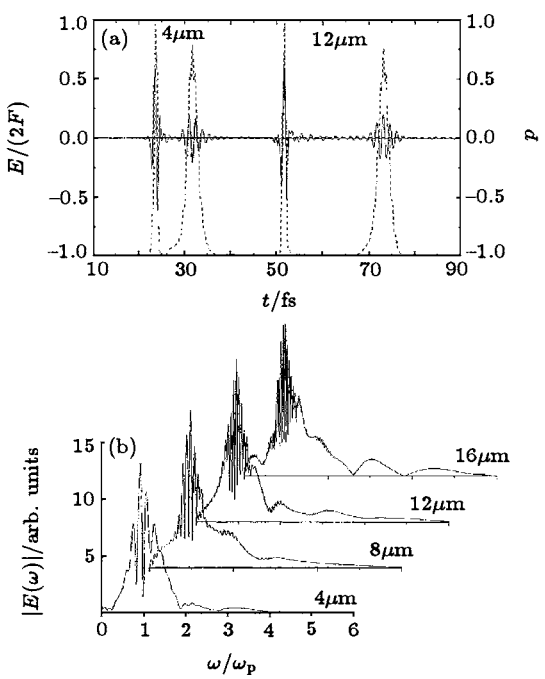


Fig.3. The 4π pulse propagation through the DBASVP medium. (a) The carrier wave (solid line) and the population difference d (dashed line) at the respective propagation distances of 4 and 12μm. (b) Evolution of the corresponding spectra at different propagation distances.

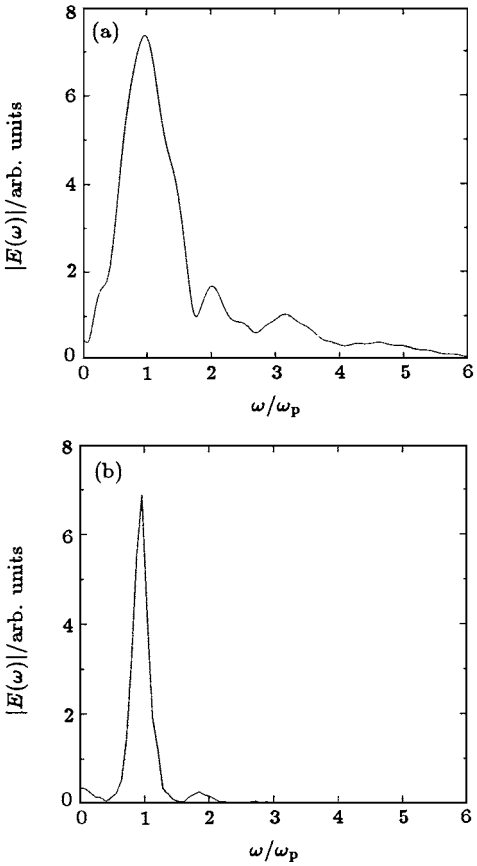


Fig.4. The two separated spectra at z=12μm for 4π pulse in DBASVP medium. (a) The spectrum related to the compressed sub-pulse. (b) That related to the broadened sub-pulse.

The corresponding spectra are shown in Fig.3(b). Since the carrier-wave shape is modulated by SPM resulting from the strong nonlinear optical properties of the medium, the spectra are broadened, and besides this, there are obvious oscillatory features appearing around the resonant and the second harmonic frequencies. The oscillatory spectrum becomes more evident with further propagation, which is supposed to be a result of the interference of two separated pulses.[6,19] In order to give further illustration, the spectra corresponding to each sub-pulse at a distance of 12μm are displayed in Fig.4. It can be seen that the spectrum related to the compressed sub-pulse (see Fig.4(a)) is broader than that given in Fig.4(b). Both the spectra contain the ω_p and $2\omega_p$ spectral components obviously, which demonstrates the mechanism for the oscillatory feature in the whole spectrum. Furthermore, the second-harmonic spectral component in Fig.4(a) is larger than that in 4(b), which is due to the stronger laser strength for the compressed sub-pulse.

For a larger area pulse, the carrier wave is more strongly modulated by SPM and CWRF. Figure 5(a) shows the carrier wave and population difference of the 6π pulse propagating at 12μm. The CWRFs are clearly observed, but they rarely occur in non-dipole medium.[6,3] In order to further illustrate the situation, we consider a pseudo-DBASVP molecule by artificially setting $\Delta\mu = 0$ in the real molecule (see

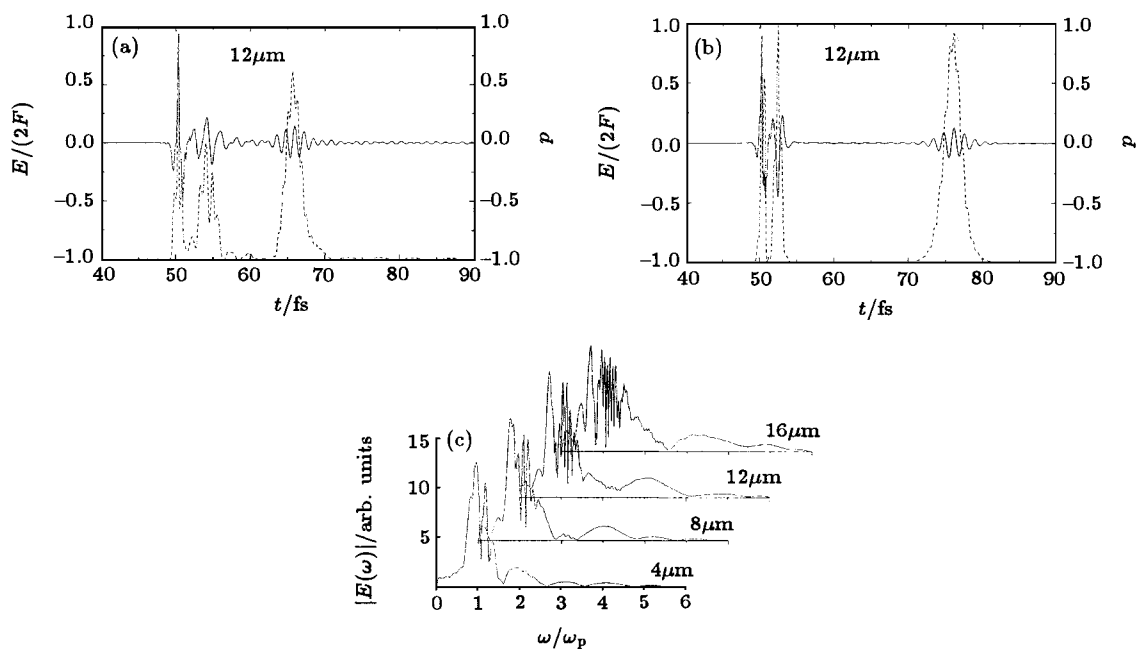


Fig.5. (a) The carrier wave (solid line) and population difference d (dashed line) at z=12μm for the 6π pulse propagating through the DBASVP medium. (b) The same as (a) only by taking $\Delta\mu = 0$. (c) Evolution of the corresponding spectra at different propagation distances.

Fig.5(b)). One can see that the effect of CWRF is quite a bit weaker. The existence of permanent dipole moment makes this kind of molecule have stronger nonlinear optical properties. Correspondingly, the permanent dipole moment can bring about stronger pulse reshaping[18] and makes the propagation of pulse deviate from area theorem more seriously. The effects induced by the permanent dipole moment become larger for stronger laser field,[20] and then one can expect that for larger area pulse, such as 8π pulse, the propagation of pulse in dipolar medium shows much different features from that in non-dipolar medium.

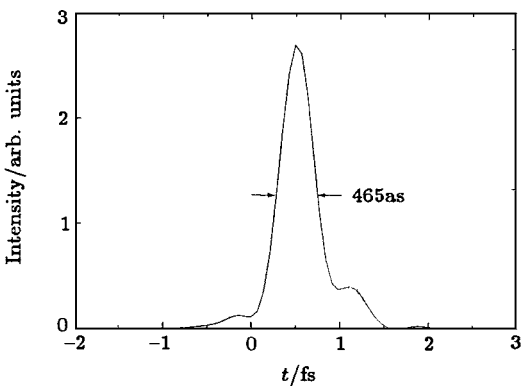


Fig.6. Single attosecond pulse corresponding to the high harmonic spectrum at z =16μm in Fig.5(c) between 2.5 and $6\omega_{\rm p}$.

From the corresponding spectrum evolution shown in Fig.5(c), one can see that the spectra are broadened and continuum generation appears with further propagation. If we take the Fourier transformation of the harmonic radiation at $z = 16\mu$m in Fig.5(c) between 2.5 and $6\omega_{\rm p}$, a shorter pulse in attosecond regime is generated (see Fig.6). Thus, we demonstrate a mechanism for obtaining attosecond pulse.

4. Conclusion

In conclusion, by studying the propagation and spectrum evolution behaviours of femtosecond laser pulse in DBASVP molecule with different areas of incoming pulses and propagation distances in the medium, we have found that the permanent dipole moment plays an important role in the propagation of the ultrashort pulse laser. In the dipolar medium, the ideal SIT disappears and significant second harmonic spectral components occur for 2π and 4π pulses. The permanent dipole moment of the medium makes the propagation of pulse deviate from the area theorem more seriously. For a larger pulse area, such as 6π pulse, continuum generation is produced. The shorter pulse in attosecond regime has also been demonstrated.

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