Effect of Lorentz local field correction on propagation of ultrashort laser pulse in one-dimensional para-nitroaniline (PNA) molecules*

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This paper investigates the effect of Lorentz local field correction (LFC) on the propagation of ultrashort laser pulses in a para-nitroaniline molecular medium under resonant and nonresonant conditions by solving numerically the full-wave Maxwell–Bloch equations beyond slowly-varying envelope approximation and rotating-wave approximation. The effect of the LFC is considerably obvious when pulses with large areas propagate in the dense molecular medium. In the case of resonance, the group velocity of the sub-pulses split from the incident pulse along propagation is severely decreased by the LFC, especially for the latest sub-pulse. However, in the case of nonresonance, the influence of the LFC on the temporal evolution of the pulse is less obvious and lacks homogeneity with an increase in incident pulse area, propagation distance and molecular density.

Keywords: Lorentz local field correction, ultra-short laser pulse, para-nitroaniline molecule, Maxwell–Bloch equations

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1. Introduction

The technological advances in ultraintense and ultrafast optics make it possible to generate femto- and atto-second laser pulses.[1−5] The propagation of an ultrashort laser pulse in a nonlinear molecular medium can produce many novel nonlinear phenomena. Recent studies of the interaction between intense fields and nonlinear materials have mainly focused on exploring the propagating features of ultrafast lasers,[6−15] and new mechanisms for generating shorter pulses have been demonstrated.[16−18] Usually, investigations of the interaction of laser fields with matter employ the semiclassical theory, i.e., the laser field is described by Maxwell equations, while the atomic or molecular medium is described by Bloch equations. For long pulses, the conventional slowly-varying envelope approximation (SVEA)[19] and rotating-wave approximation (RWA)[20] can be applied to the Maxwell–Bloch equations. However, for ultrashort pulses, the SVEA and RWA are proved to be invalid.[6−17] The propagation behaviours of few-cycle laser pulses in dilute two-level atomic or molecular mediums have been extensively studied by solving the full-wave Maxwell–Bloch equations without SVEA and RWA.[21−24]

In a dilute medium, the intermolecular distance is quite large, and the local field caused by the polarization of the nearby molecules is negligible. Therefore, the total electromagnetic field felt by an individual molecule in a dilute medium equals the incident external field. In contrast, the near dipole–dipole (NDD) interaction, which leads to LFC of the field imposed on the molecules,[25−27] must be considered in a dense medium. The NDD effects manifested in the form of LFC have been widely investigated by employing SVEA and/or RWA.[28,29] Recently, the local field effects on the propagation of few-cycle laser pulses by solving the full-wave Maxwell–Bloch equations without employing SVEA and RWA have been studied.[30,31]

In this paper, we study the evolution of the ultrashort laser pulses with the influence of the LFC in a dense polar molecular medium (para-nitroaniline (PNA)) by solving numerically the full-wave Maxwell–Bloch equations with the iterative predictor-corrector finite-difference time domain method.[8] The resonant and nonresonant interactions of the pulses with the

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dense medium will be discussed.

2. Formulation of the theory

The structure of the PNA molecule is schematically shown in Fig. 1. The PNA is a one-dimensional asymmetrical molecule with strong charge-transfer (CT) properties. The electronic properties of the molecule are calculated by use of the Gaussian package with density functional theory.[32] It shows that this molecule has only one CT state, which determines completely the nonlinear properties of the molecule in the optical region. Therefore, the two-level model made up of the ground state and the CT state is used to model the electronic structure of the molecule. The excitation energy of the CT state is $E_0 = \hbar\omega_0 = 3.85$ eV. The transition dipole moment between the two states is $\mu = 1.621 \times 10^{-29}$ C·m, and the permanent dipole moments of the two states are $\mu_{11} = 4.373 \times 10^{-29}$ C·m and $\mu_{22} = 2.655 \times 10^{-29}$ C·m, respectively.

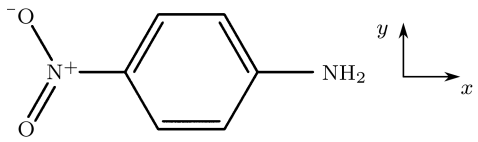


Fig. 1. Para-nitroaniline (PNA) molecule.

The evolution of the two-level system in an external electric field E_x is determined by the following Bloch equations,[33]

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

where u and v are the two real Bloch vectors related to the off-diagonal density matrix element of the two-level molecule by $\rho_{12} = (u + \mathrm{i}v)/2$, $d = \rho_{22} - \rho_{11}$ is the population difference between the upper state ρ_{22} and the ground state ρ_{11}. The molecules are assumed to stay in the ground state at the initial time when there is no interaction between the field and the medium, namely $\rho_{11}^0 = 1$, $\rho_{22}^0 = 0$ and $\Delta d(t = 0) = -1$. The $\Delta\mu = \mu_{22} - \mu_{11}$ is the difference between the permanent dipole moments. The γ_{22} and γ_{12} (γ_{21}) are the relaxation constants of the population of the upper state and the dipolar coherence, respectively. Usually, the decay rates can be chosen as $\gamma_{22} = 1.0 \times 10^9$ /s, $\gamma_{12} = 1.0 \times 10^{13}$ /s.[34]

Assume that the incident electromagnetic field is polarized along the x axis and propagates along the z axis. Hence, Maxwell's equations in the one-dimensional case are written as

$$\begin{cases} \dfrac{\partial E_x}{\partial z} = -\mu_0 \dfrac{\partial H_y}{\partial t}, \\ \dfrac{\partial H_y}{\partial z} = -\dfrac{\partial P_x}{\partial t} - \varepsilon_0 \dfrac{\partial E_x}{\partial t}, \end{cases} \quad (2)$$

where μ_0 and ε_0 are the permeability and permittivity of free space respectively. The P_x is the macroscopic polarization of the medium, which couples the Bloch and Maxwell's equations. In the two-level system,

$$P_x = N(\mu_{11}\rho_{11} + \mu_{22}\rho_{22} + \mu u), \quad (3)$$

where N is the molecular density.

In a dense medium, the microscopic local electric field E_{L} is related to the microscopic field E_x and the nonlinear volume polarization P_x according to the Lorentz–Lorentz relation

$$E_{\mathrm{L}} = E_x + P_x/3\varepsilon_0. \quad (4)$$

That is to say that, in the dense medium, the total local field E_{L} must be used to take the place of the external field E_x in the Maxwell–Bloch equations (1) and (2).

The initial pulse entering into the medium at $z = 0$ has a hyperbolicsecant shape, $E_x(z, t = 0) = F\,\mathrm{sech}[1.76(z/c + z_0/c)/\tau_{\mathrm{p}}]\cos[\omega_{\mathrm{p}}(z + z_0)/c + \phi]$, where F is the peak amplitude of the input electric field, ϕ is the initial phase and τ_{p} is the full width at half maximum of the pulse intensity envelope. Here, ϕ and τ_{p} are taken to be 0 and 2 fs, respectively. Two different carrier frequencies ω_{p} of the incident pulses are used in our study, one is the resonant frequency ω_0 and the other is $\omega_0/2$ in the nonresonant case. The choice of z_0 ensures that the pulse penetrates little distance into the medium at $t = 0$. For the hyperbolicsecant pulse, the area of the pulse is defined by

$$\theta(z) = \frac{\mu}{\hbar}\int_{-\infty}^{\infty} E_0(z, t')\,\mathrm{d}t' = \frac{\mu F \tau_{\mathrm{p}}\pi}{1.76\hbar}.$$

The time and space increments Δt and Δz are chosen to ensure $c\Delta t \leq \Delta z$.

3. Results and discussion

3.1. One-photon resonant propagation of ultrashort pulse

Figure 2 shows the time evolutions of the electric field E for the 6π laser pulse at the distances

of $z = 79.2$ μm under the one-photon condition at different concentrations. It shows that the propagation evolutions are quite different, even though the Lorentz local field correction (LFC) is not considered when the medium density increases. We can see clearly that, in the dilute medium, the light-matter interaction with the LFC makes little difference to the case without LFC. When the molecular density increases to $7 \times 10^{26}/\mathrm{m}^3$, there is an obvious difference in the two cases with and without LFC. Therefore, the large medium density has a great influence on the interaction between the laser pulses and the molecule medium. The pulses with LFC propagate faster than those without LFC, and the time interval between the sub-pulses with LFC is shorter than that without LFC. In other words, the NDD interaction makes the increase in group velocity of laser pulse.

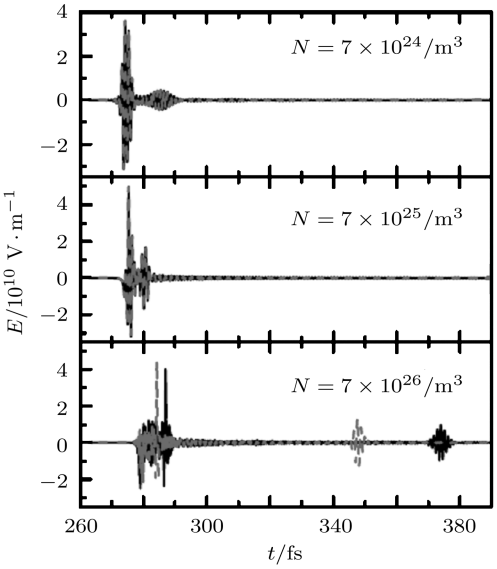


Fig. 2. The time evolutions of the 6π pulse at the distance of $z = 79.2$ μm for $N = 7 \times 10^{24}/\mathrm{m}^3$, $N = 7 \times 10^{25}/\mathrm{m}^3$ and $N = 7 \times 10^{26}/\mathrm{m}^3$, the solid line for the case without LFC and the dashed line for that with LFC. The other figures take the same illustration.

From Fig. 3, one can see clearly that the difference between the two cases with and without LFC is more remarkable as the pulse area increases in the dense media. With the increase in the initial pulse area, the sub-pulses with LFC transfer faster than that without LFC, and the time interval between the sub-pulses with LFC is shorter than that without LFC. This shows that the NDD interaction has an obvious effect on the reciprocity of the medium and the laser pulses especially when the initial pulses are strong, e.g. the 8π pulses. The cause is that in the case with a large area pulse, the strength of the electronic field is stronger, so the correction between the medium and the laser pulses becomes evident. Thus the result shows that the effect of LFC is strengthened in the case of a large area pulse.

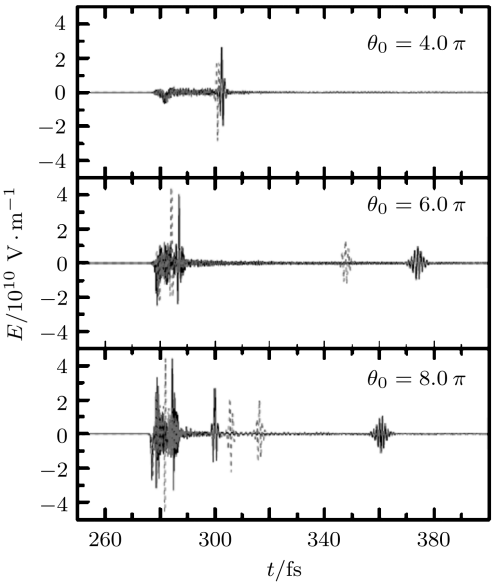


Fig. 3. The time evolutions of the $4\pi, 6\pi, 8\pi$ pulses at a distance of $z = 79.2$ μm for $N = 7 \times 10^{26}/\mathrm{m}^3$.

Figure 4 depicts the difference in the time evolutions of the electric field E for the 6π laser pulses at distances of $z = 26.4$ μm, $z = 79.2$ μm and $z = 132.0$ μm when the molecular density is $7 \times 10^{26}/\mathrm{m}^3$.

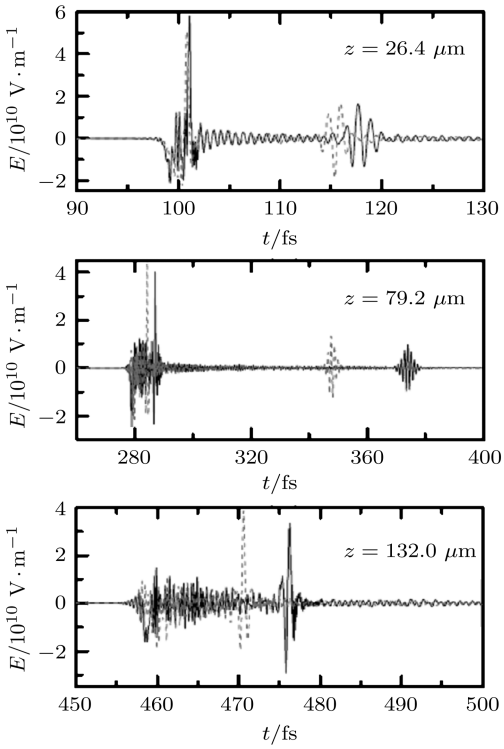


Fig. 4. The time evolutions of the 6π pulses at distances of $z = 26.4$ μm, $z = 79.2$ μm and $z = 132.0$ μm for $N = 7 \times 10^{26}/\mathrm{m}^3$.

One can see clearly that the pulses with LFC transfer faster than those without LFC pulses when the evolution distance becomes longer. The result discussed above shows that for the 6π pulse, the LFC has a critical effect in a dense medium. Considering the propagation distance, we find that the evolution of the pulses manifests difference in the two cases with and without LFC. As discussed above, the NDD interaction makes the increase in group velocity of the laser pulse. So the little transfer difference between the two cases at the short distance $z = 26.4$ μm gets larger as the pulses propagate farther. Therefore, when the laser pulse transfers over a long distance in a dense medium, the LFC cannot be ignored.

3.2. One-photon nonresonant propagation of ultrashort pulse

Figure 5 shows the time evolutions of the electric field E for the 6π laser pulses at a distance of $z = 79.2$ μm on the one-photon nonresonant condition. Due to one-photon nonresonant absorption, the pulse splits into fewer sub-pulses than the case of one-photon resonant absorption whether the LFC is considered or not. Being similar to the case on the one-photon resonant condition as depicted in Fig. 2, we find that the light-matter interaction in a dilute medium with the LFC shows little difference compared with the case without LFC. When the molecular density increases to $7 \times 10^{26}/\mathrm{m}^3$, there is an obvious difference between the cases with and without LFC. So the NDD interaction makes the increase in group velocity of the laser pulse under the one-photon nonresonant condition. Consequently, whether the condition is one-photon resonant absorption or not, the NDD interaction will enhance the interaction between the pulses and the medium, and speed up the propagation velocity of the sub-pulses.

Figure 6 shows the time evolutions of the $4\pi, 6\pi, 8\pi$ pulses at a distance of $z = 79.2$ μm on the one-photon nonresonant condition. As the initial pulse becomes stronger, the evolution of the pulse considering the LFC spreads faster than the case without the LFC. But unlike the one-photon resonant propagation, the time interval between cases with and without LFC does not increase as the initial pulse area rises. This indicates that the LFC has a different impact on the interaction between the laser pulse and the medium when the initial pulse area is different. The time evolutions of the electric field E for the 6π laser pulses at distances of $z = 26.4$ μm, $z = 79.2$ μm and $z = 132.0$ μm on the one-photon nonresonant condition, respectively, are shown in Fig. 7. One can see that the time interval between cases with and without LFC does not increase as the transporting distance increases.

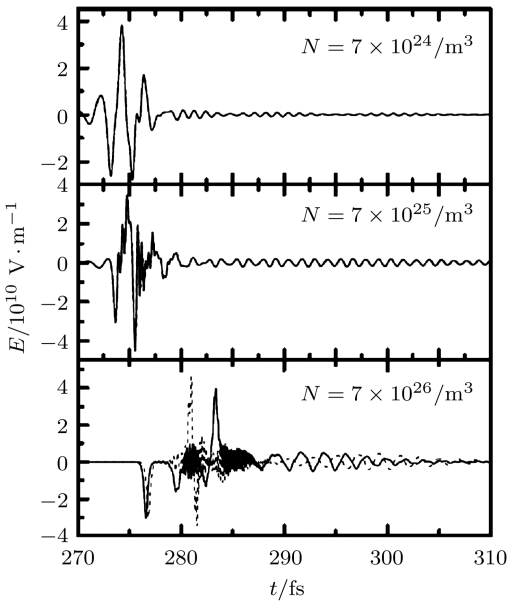


Fig. 5. The time evolutions of the 6π pulse at a distance of $z = 79.2$ μm for $N = 7 \times 10^{24}/\mathrm{m}^3$, $N = 7 \times 10^{25}/\mathrm{m}^3$ and $N = 7 \times 10^{26}/\mathrm{m}^3$.

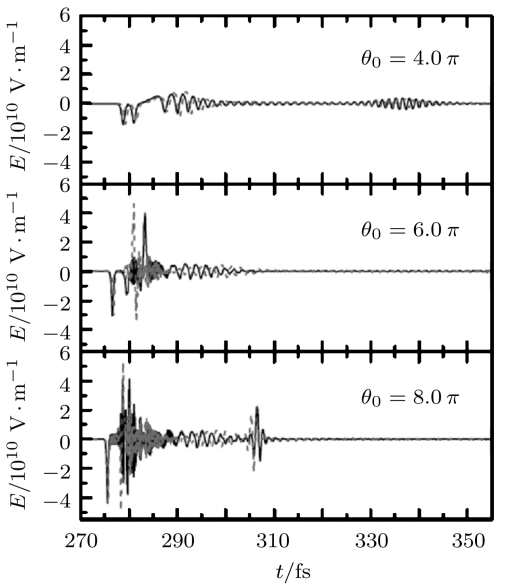
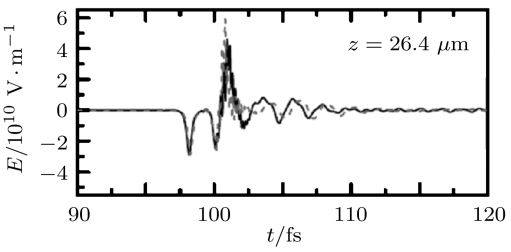


Fig. 6. The time evolutions of the $4\pi, 6\pi, 8\pi$ pulses at the distance of $z = 79.2$ μm for $N = 7 \times 10^{26}/\mathrm{m}^3$.



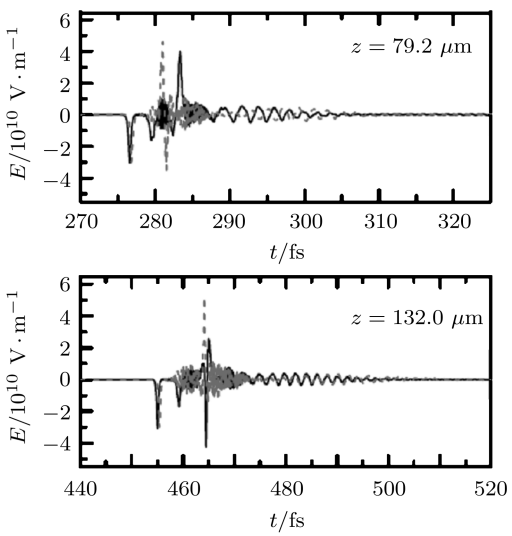


Fig. 7. The time evolutions of the 6π pulses at the distance of $z = 26.4$ μm, $z = 79.2$ μm and $z = 132.0$ μm for $N = 7 \times 10^{26}/\mathrm{m}^3$.

4. Conclusion

By solving the full Maxwell–Bloch equations, we have numerically investigated the propagation behaviours of ultrashort laser pulses in a dipolar molecule medium. In the dilute medium ($N = 7 \times 10^{24}/\mathrm{m}^3$), the effect of the Lorentz LFC is remarkably small. Under the one-photon resonant condition, the pulses with LFC transfer faster than those without LFC, and the time interval between the sub-pulses in the case with LFC is shorter than that without LFC. The phenomenon is not obvious under the one-photon nonresonant condition. When the initial pulse area is increased or the transporting distance becomes longer, the NDD interaction makes some pulses transfer faster than those without LFC for the one-photon nonresonant condition.

References

[1] Baltuška A, Wei Z Y, Pshenichnikov M S and Wiersma D A 1997 *Opt. Lett.* **22** 102

[2] Persson E, Schiessl K, Scrinzi A and Burgdörfer J 2006 *Phys. Rev.* A **74** 013818

[3] Leblond H, Sazonov S V, Mel'nikov I V, Mihalache D and Sanchez F 2006 *Phys. Rev.* A **74** 063815

[4] Scrinzi A, Yu Ivanov M, Kienberger R and Villeneuve D M 2006 *J. Phys.* B: *At. Mol. Opt. Phys.* **39** R1

[5] Brabec T and Krausz F 2000 *Rev. Mod. Phys.* **72** 545

[6] Xiao J, Wang Z Y and Xu Z Z 2001 *Chin. Phys.* **10** 941

[7] Ziolkowski R W, Arnold J M and Gogny D M 1995 *Phys. Rev.* A **52** 3082

[8] Hughes S 1998 *Phys. Rev. Lett.* **81** 3363

[9] Brown A, Meath W J and Kondo A E 2002 *Phys. Rev.* A **65** 060702

[10] Flesh R G, Pushkarev A and Moloney J V 1996 *Phys. Rev. Lett.* **76** 2488

[11] Yang W F, Song X H, Gong S Q, Cheng Y and Xu Z Z 2007 *Phys. Rev. Lett.* **99** 133602

[12] Song X H, Gong S Q, Jin S Q and Xu Z Z 2004 *Phys. Rev.* A **69** 015801

[13] Song X H, Gong S Q, Yang W F and Xu Z Z 2004 *Phys. Rev.* A **70** 013817

[14] Liu J C, Wang C K and Gel'mukhanov F 2007 *Phys. Rev.* A **76** 043422

[15] Liu J C, Wang C K and Gel'mukhanov F 2007 *Phys. Rev.* A **76** 053804

[16] Liu J C, Zhao K, Song Y Z and Wang C K 2006 *Acta Phys. Sin.* **55** 1803 (in Chinese)

[17] Zhao K, Li H Y, Liu J C and Wang C K 2006 *Chin. Phys.* **15** 2338

[18] Zhao K, Liu J C, Wang C K and Luo Y 2007 *J. Phys.* B: *At. Mol. Opt. Phys.* **40** 1523

[19] Born M and Wolf E 1975 *Principle of Optics* (Oxford: Pergamon)

[20] Lamb Jr W E 1964 *Phys. Rev.* A **134** 1429

[21] Mücke D, Tritschler T and Wegener M 2001 *Phys. Rev. Lett.* **87** 057401

[22] Xiao J, Wang Z Y and Xu Z Z 2002 *Phys. Rev.* A **65** 031402

[23] Patton B, Woggon U and Langbein W 2005 *Phys. Rev. Lett.* **95** 266401

[24] Song X H, Gong S Q, Li R X and Xu Z Z 2006 *Phys. Rev.* A **74** 015802

[25] Hopf F A, Bowden C M and Louisell W H 1984 *Phys. Rev.* A **29** 2591

[26] Ben-Aryeh Y, Bowden C M and Englund J C 1986 *Phys. Rev.* A **34** 3917

[27] Friedberg F, Hartmann S R and Manasseh J T 1989 *Phys. Rev.* A **40** 2446

[28] Wang N and Rabitz H 1995 *Phys. Rev.* A **51** 5029

[29] Calderón O G, Antón M A and Carreño F 2003 *Euro. Phys. J.* D **25** 77

[30] Xia K Y, Gong S Q, Liu C P, Song X H and Niu Y P 2005 *Opt. Express* **13** 5913

[31] Ma H, Fan X J, Tan X, Lu H W and Xu Z Z 2008 *Opt. Commun.* **281** 4493

[32] Wang C K, Zhao K, Su Y, Ren Y, Zhao X and Luo Y 2003 *J. Chem. Phys.* **119** 1208

[33] Boyd R W 2003 *Nonlinear Optics* (San Diego: Elsevier)

[34] Gel'mukhanov F, Baev A, Macák P, Luo Y and Ågren H 2002 *J. Opt. Soc. Am.* B **19** 937