Effect of the ratio of transition dipole moments on few-cycle pulse propagation

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Propagation of a few-cycle laser pulses in a dense V-type three-level atomic medium is investigated based on full-wave Maxwell-Bloch equations by taking the near dipole-dipole (NDD) interaction into account. We find that the ratio, γ, of the transition dipole moments has strong influence on the time evolution and split of the pulse: when $\gamma \leq 1$, the NDD interaction delays propagation and split of the pulse, and this phenomenon is more obvious when the value of γ is smaller; when $\gamma = \sqrt{2}$, the NDD interaction accelerates propagation and split of the pulse.

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The resonant interaction of ultrashort pulse laser with atoms or molecules is currently one of the hottest study subjects[1−4] and the propagation of ultrashort pulse laser in the atom or molecule mediums is an important part of the study[5,6]. However, when a few-cycle laser pulse is considered, the slowly varying envelope approximation (SVEA) and the rotating wave approximation (RWA) are invalid[7−12] and a lot of new characteristics are generated. Hughes[7] has shown that, for larger area pulse, Rabi flopping of carrier wave led to the area theorem[13] breakdown. Xiao *et al.*[8] studied area evolution of few-cycle pulse in a two-level medium and found that different from the long-pulse case, area variation of the few-cycle pulse is caused by the pulse splitting and not by the pulse broadening or compression. Netz *et al.*[14] investigated the influence of phase modulation on the reshaping of ultrashort laser pulses in resonant three-level systems and found that in small time region scales, the phase modulation affected the reshaping process during the pulse propagation. So far, the study of the interaction of ultrashort laser pulse with multilevel medium is few.

In a dense medium, where the atomic density is so high that there are, in the sense of average, so many atoms within a cubic resonance wavelength that the near-dipole-dipole interaction (NDD), which leads to the Lorentz local-field correction (LFC), must be considered[15−19]. Some papers have considered the NDD interaction by employing the SVEA and RWA. Recently, Xia *et al.*[20] investigated the NDD effects on the propagation of few-cycle pulse in a dense two-level medium without using the SVEA and RWA. In this paper, using the numerical solutions of the full Maxwell-Bloch (M-B) equation without the SVEA and RWA obtained by the iterative predictor-corrector method and the finite-difference time-domain method, we study the propagation of a few-cycle laser pulse in the dense V-type three-level medium. Some new important results are obtained.

Figure 1 depicts the levels of the Rb atom and the driving field. In a dense medium, according to the Lorentz-Lorenz relation[15], the microscopic local electric field E_{L}, which couples with atomic dipole moments, is related to the external field E and volume polarization P in the isotropic homogeneous medium,

$$E_{\mathrm{L}} = E + \frac{P}{3\varepsilon_0}, \tag{1}$$

where ε_0 is the electric permittivity in vacuum.

Considering the NDD interaction and the dissipative effects, the real form of the motion equations of the density matrix of the system, i.e., the one-dimensional (1D) Bloch equations, can be written as

$$\dot{u}_1 = -\omega_1 u_4 - \gamma\Omega_{\mathrm{L}} u_5 - \frac{1}{T_1} u_1, \tag{2a}$$

$$\dot{u}_2 = (\omega_1 - \omega_2) u_5 + \gamma\Omega_{\mathrm{L}} u_4 + \Omega_{\mathrm{L}} u_6 - \frac{1}{T_2} u_2, \tag{2b}$$

$$\dot{u}_3 = -\omega_2 u_6 + \Omega_{\mathrm{L}} u_5 - \frac{1}{T_3} u_3, \tag{2c}$$

$$\dot{u}_4 = \omega_1 u_1 - 2\Omega_{\mathrm{L}} u_7 - \gamma\Omega_{\mathrm{L}} u_2 - \frac{1}{T_4} u_4, \tag{2d}$$

$$\dot{u}_5 = u_2(\omega_2 - \omega_1) + \gamma\Omega_{\mathrm{L}} u_1 - \Omega_{\mathrm{L}} u_3 - \frac{1}{T_5} u_5, \tag{2e}$$

$$\dot{u}_6 = \omega_2 u_3 - \Omega_{\mathrm{L}} u_2 - 2\gamma\Omega_{\mathrm{L}} u_8 - \frac{1}{T_6} u_6, \tag{2f}$$

$$\dot{u}_7 = 2\Omega_{\mathrm{L}} u_4 + \gamma\Omega_{\mathrm{L}} u_6 - \frac{1}{T_7}(u_7 - u_{70}), \tag{2g}$$

$$\dot{u}_8 = 2\gamma\Omega_{\mathrm{L}} u_6 + \Omega_{\mathrm{L}} u_4 - \frac{1}{T_8}(u_8 - u_{80}), \tag{2h}$$

where T_i $(i = 1, 2, \cdots, 8)$ is the relation time corresponding to the delay of the real state vector component u_i $(i = 1, 2, \cdots, 8)$; u_{70} and u_{80} are the initial values of u_7 and u_8, respectively, ω_1 and ω_2 represent the transition frequencies from $|1\rangle$ to $|2\rangle$ and $|3\rangle$, respectively, $\gamma(= \mu_{13}/\mu_{12})$

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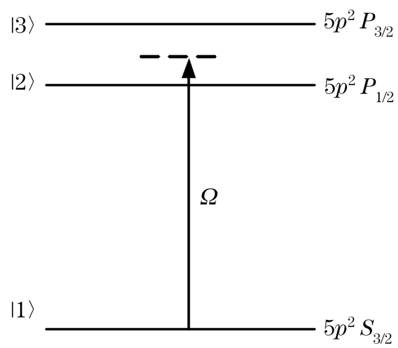

Fig. 1. Energy level of rubidium included in the field-atom interaction.

is the ratio of the transition dipole moments, where μ_{ij} are the dipole moment between the levels i and j $(i, j = 1, 2, 3$ and $i \neq j)$; $\Omega_{\mathrm{L}} = \mu_{12}E_{\mathrm{L}}/\hbar = \Omega_x - \varepsilon\,(u_1 + \gamma u_3)$ is the Rabi frequency of the microscopic local electric field, where $\Omega_x = \mu_{12}E_x/\hbar$ is the Rabi frequency of the pulse laser field, $\varepsilon = N\mu_{12}^2/3\varepsilon_0\hbar$ is the NDD parameter and has the unit of frequency, which presents the strength of the NDD interaction. u_i is related to the density matrix elements ρ_{ij} $(i, j = 1, 2, 3,)$ according to the following relations:

$$
\begin{aligned}
&u_1 = \rho_{12} + \rho_{21}, \quad u_2 = \rho_{23} + \rho_{32},\\
&u_3 = \rho_{13} + \rho_{31}, \quad u_4 = -i(\rho_{12} - \rho_{21}),\\
&u_5 = -i(\rho_{23} - \rho_{32}), \quad u_6 = -i(\rho_{13} - \rho_{31}),\\
&u_7 = \rho_{22} - \rho_{11}, \quad u_8 = \rho_{33} - \rho_{11}.
\end{aligned} \tag{3}
$$

Under the conditions that the electric field is linear polarization along the x-axis (E_x) and the magnetic field is along the y-axis (H_y), the Maxwell equations take the form:

$$\partial_t H_y = -\frac{1}{\mu_0}\partial_z E_x, \tag{4a}$$

$$\partial_t E_x = -\frac{1}{\varepsilon_0}\partial_z H_y - \frac{1}{\varepsilon_0}\partial_t P_x, \tag{4b}$$

where μ_0 is the magnetic permeability in vacuum, $P_x = -N\langle\vec{\mu}\rangle = N\mu_{12}\,(u_1 + \gamma u_3)$, N is the density of the medium.

In order to investigate the propagation properties of an ultrashort pulse in a dense V-type three-level atomic medium, we need to solve the full M-B Eqs. (2) and (4). But it is very difficult to solve analytically M-B equations, so we use the finite-difference time-domain (FDTD) method for the fields and the predictor-corrector method for the material variables to obtain their numerical solutions. The initial input fields are

$$
\begin{aligned}
E_x(t = 0, z) = {} & E_0 \mathrm{sech}\,[1.76(z/c - z_0/c)/\tau_{\mathrm{p}}]\\
& \times \cos\,[\omega_{\mathrm{p}}(z/c - z_0/c)]\,,
\end{aligned} \tag{5}
$$

$$H_y(t = 0, z) = \sqrt{\varepsilon_0/\mu_0}E_x(t = 0, z), \tag{6}$$

where E_0 is the maximal electric field amplitude, τ_{p} and ω_{p} are the full-width at half-maximum (FWHM) and the transition frequency of the pulse intensity envelope, respectively. In the following numerical analysis, we adopt the initial condition : $u_{70} = -1$, $u_{80} = -1$, and the following pulse and material parameters values: $\tau_{\mathrm{p}} = 5$ fs, $\omega_1 = 2.37$ fs^{-1}, $\omega_2 = 2.42$ fs^{-1}, $\omega_{\mathrm{p}} = 2.395$ fs^{-1}, $\mu_{12} = 1.48 \times 10^{-29}$ Asm. The relaxation times corresponding to the decay of the real state vector components are set to the uniform value $T_1 = \cdots = T_8 = 1 \times 10^{-10}$ s.

Now we present and analyze representative numerical solutions of the coupled M-B Eqs. (2) and (4). The ratio, γ, of the transition dipole moments μ_{13} and μ_{12} has strong influence on the propagation behavior of ultrashort pulse. We first consider the case of $\gamma = 1$. Figure 2 depicts the time evolution of the Rabi frequency at the respective distance of 24, 72, and 120 μm in a V-type three-level dense medium with equal transition dipole moments. As shown in Figs. 2(a) and (b), small oscillations occur at the leading and trailing edges due to the time derivative of the electric field. With the increase of propagation distance, the oscillation at the trailing edge becomes more obvious. In addition, the main pulse width with the NDD interaction is smaller than that without the NDD interaction as shown in Figs. 2(c) and (d), and this phenomenon also becomes more evident with the increase of propagation distance. The propagation of the pulse delays when the NDD interaction presents. In the case of with the NDD interaction, the pulse amplitude is smaller, so is the group velocity. Moreover, the delayed time becomes longer with propagation distance increasing.

For larger area pulses, for example, $A = 3.3\pi$, pulse splitting occurs and the NDD interaction also leads to the phenomena of pulse widened and time delayed during the pulse propagating as shown in Fig. 3(a). The phenomena of time delayed for the first sub-pulse are not obvious. But the phenomena of pulse widened and time delayed for the second sub-pulse are more obvious; moreover, time delayed is longer with the increase of propagation distance.

In most cases, the dipole moments of the allowed transitions are unequal. Figure 3(b) depicts the time evolution of the Rabi frequency at the respective distance of

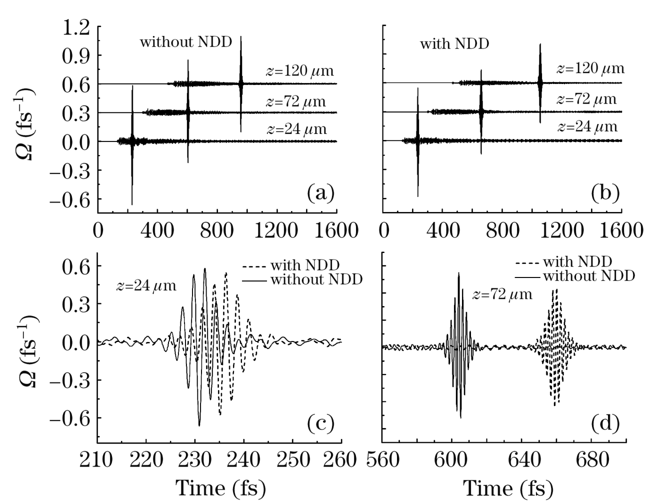

Fig. 2. Time evolution of Rabi frequency Ω at the respective distances of 24, 72, and 120 μm with $N = 6 \times 10^{26}$ m^{-3}, $A = 2\pi$, $\gamma = 1$. (a) Without NDD; (b) with NDD; (c) $z = 24$ μm; (d) $z = 72$ μm.

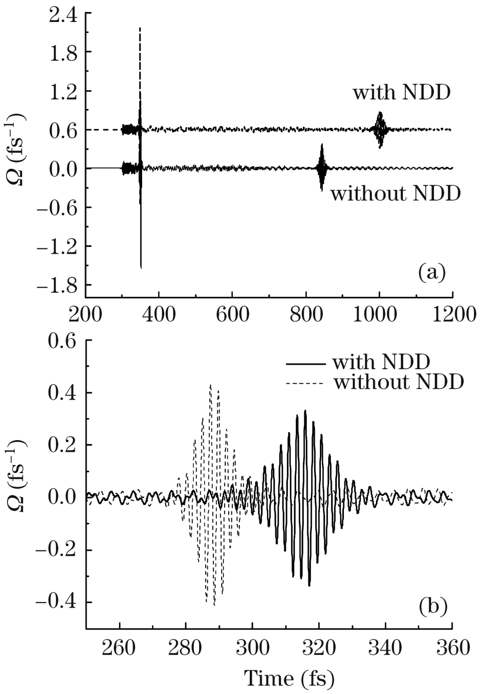

Fig. 3. Time evolution of Rabi frequency Ω with $N = 6\times10^{26}$ m^{-3} for (a) $\gamma = 1$, $A = 3.3\pi$, $z = 72$ μm; (b) $\gamma = 0.8$, $A = 2\pi$, $z = 24$ μm.

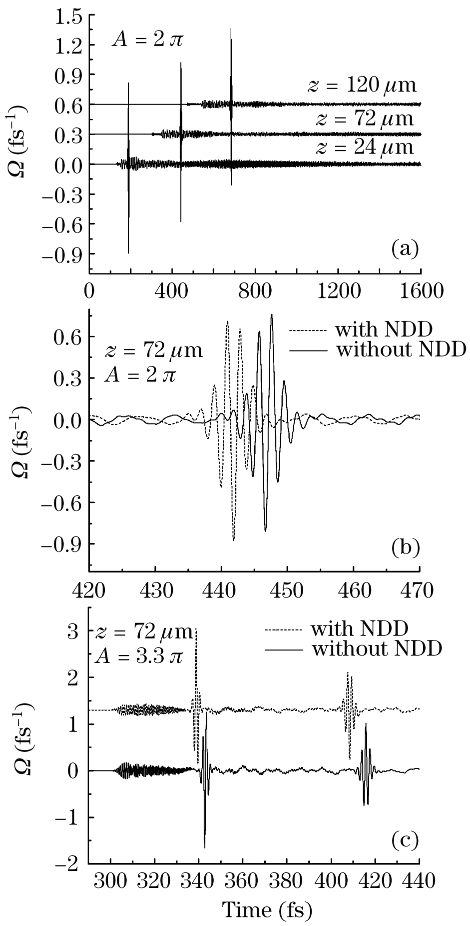

Fig. 4. Time evolution of Rabi frequency Ω with $N = 6\times10^{26}$ m^{-3} and $\gamma = \sqrt{2}$ for (a) $A = 2\pi$ with NDD; (b) $A = 2\pi$, $z = 72$ μm; (c) $A = 3.3\pi$, $z = 72$ μm.

24 μm in a V-type three-level dense medium with $\gamma = 0.8$. The phenomena of pulse widened and time delayed also appear during the pulse propagation when the NDD interaction presents. Comparing Fig. 3(b) with Fig. 2(c), we can also find that this phenomenon for $\gamma = 0.8$ is more obvious than that for $\gamma = 1$. The numerical result shows that when $\gamma \leq 1$, the smaller the value of γ is , the more obvious the phenomenon is.

Now we consider the case of $\gamma > 1$, for example, the transition dipole moments corresponding to the transitions $5s^2S_{1/2} \rightarrow 5p^2P_{1/2}$ and $5s^2S_{1/2} \rightarrow 5p^2P_{3/2}$ in a Rb atom are $\mu_{13} = 2.09\times10^{-29}$ Asm and $\mu_{12} = 1.48\times10^{-29}$ Asm, so $\gamma = \sqrt{2}$[21]. Figure 4 depicts the time evolution of the Rabi frequency at the respective distance of 24, 72, and 120 μm when $\gamma = \sqrt{2}$. Comparing Fig. 4(a) with Fig. 2(b), we can see that the pulse amplitude for $\gamma = \sqrt{2}$ is larger than that for $\gamma \leq 1$. Comparing Fig. 4(b) with Fig. 2(d), we can see that for the pulses with the same input area, the oscillation frequency is higher when $\gamma \leq 1$. Figure 4(b) shows that for 2π pulse, at the same distance, the time of pulse appears when the NDD interaction presents is earlier than that when the NDD interaction is absent, but this phenomenon is opposite to that when $\gamma = 1$ (Fig. 2). This phenomenon is also occurs for the larger area pulse (Fig. 4(d)), and more obvious for the second sub-pulses.

The phenomena above can be explained from the physical view as follows. Generaly speaking, the group velocity is usually strongly dependent on frequency. This effect is quantified by the group velocity dispersion (GVD)[22]. In fact, the group velocity of ultrashort pulse propagating in medium is determineded by many factors: the transition dipole moment, frequency of the pulse electric field, final pulse width, frequency spectrum, local field, and population[23]. In present paper, Rabi frequncies of the local fields with and without the NDD interaction are $\Omega_\mathrm{L} = \Omega_x - \varepsilon\,(u_1 + \gamma u_3)$ and $\Omega_\mathrm{L} = \Omega_x$, respectively. From M-B Eqs. (2) and (4) (considering simultaneously Eq. (3)), variation of Ω_L and γ will lead to the variety of solution of M-B Eqs. (2) and (4), i.e., the dispersions (u_1, u_2, u_3), absorptions (u_4, u_5, u_6), and populations $(\rho_{11}, \rho_{22}, \rho_{33})$. Variation of the dispersions will lead to the shift of frequency spectrum; variation of the dispersions and absorptions also affects the pulse width. These variations produced by the varying of Ω_L and γ are complex and nonlinear. Finally, the combinative effects of these variations lead to the change of the group velocity and the phenomena mentioned above.

In conclusion, our analysis shows that the ratio of the transition dipole moments has strong influence on the time evolution and split of the few-cycle pulse propagating in the dense V-type three-level atomic medium; when $\gamma \leq 1$, the NDD interaction delays propagation and split of the pulse, and this phenomenon is more obvious when the value of γ is smaller; for the case of $\gamma = \sqrt{2}$, the NDD interaction accelerates propagation and split of the pulse.

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