Influence of atomic densities on propagation property for ultrashort pulses in a two-level medium

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The influence of atomic densities on the propagation property for ultrashort pulses in a two-level atom (TLA) medium is investigated. With higher atomic densities, the self-induced transparency (SIT) cannot be recovered even for 2π ultrashort pulses. New features such as pulse splitting, red-shift and blue-shift of the corresponding spectra arise, and the component of central frequency gradually disappears.

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The interaction of laser light with a collection of two-level atom (TLA) medium has been extensively studied in the past years[1]. A number of fascinating nonlinear optical propagation properties, including Rabi oscillation (RO), self-induced transparency (SIT), photon echo, and optical shock formation, have been studied[1−5]. The interaction can be greatly affected by laser parameters such as the full-width at half-maximum (FWHM), pulse area, phase, etc.. For long-pulse cases, these propagation phenomena can be described adequately by the coupled matter Maxwell equations within the slowly varying envelope approximation (SVEA) and rotary wave approximation (RWA). However, the SVEA and RWA are invalid for extremely short and intense pulses with only few cycles, and many new features that are absent in the SVEA and RWA models arise[6,7]. For example, contrary to the long-pulse case, the variation of the few-cycle pulse area is caused by the pulse splitting but not by the pulse broadening or the pulse compression[8]. Moreover, it has been demonstrated that optical subcycle pulses can be generated due to pulse splitting and reshaping of a few-cycles time duration[9]. For small area pulses such as 2π and 4π, Ziolkowski and Hughes *et al.* have demonstrated that the SIT can still be recovered[10,11]. While for large area pulses, Hughes has found that due to carrier-wave RO, higher spectral components and even soft-X-ray generation can occur[5,12]. Phase control of spectral effects for ultrashort pulses and photoabsorption, which resonantly enhanced photoionization in an optical dense medium, has also been investigated widely[13−15].

Material parameters especially atomic densities also have great influence on the interaction between laser and material. For example, the impact of atomic density on high-order harmonic generation has been examined theoretically and experimentally. It is shown that both the adaptability of the reduced first-order wave equation and the maximum photon energy reached for the high-order harmonics depend on the atomic densities[16,17]. Shin *et al.* have studied the blue-shift of high-order harmonics and found that the fractional blue-shift increased linearly with increasing the gas density and the slopes of the density-dependent fractional blueshift for different harmonic orders were nearly same[18]. Ranka *et al.* have also investigated the spectrum of femtosecond laser pulse propagating through a TLA medium, and showed that the spectral feature develops oscillations that will become more numerous as the atomic density is increased[19]. Moreover, dark lines in the fluorescence spectrum and line width versus atomic densities have been studied, and dark lines inverted and line width narrowed in thick vapors were demonstrated[20]. Here we investigate the influence of atomic densities on the propagation property and present a quantitative full-wave Maxwell-Bloch (M-B) analysis to model pulses of only a few optical-cycles time duration. It is found that with higher atomic densities, the SIT cannot be recovered even for 2π ultrashort pulse propagating in the TLA medium, and several new features such as pulse splitting, red-shift and bule-shift of the corresponding spectra arise. This work is theoretically studied in part; what's more, our predictions employing realistic material and laser parameters are experimentally verified.

The propagation property of laser light fields in a resonant TLA medium with an atomic density N can be modelled using the full-wave M-B equations,

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

$$\partial_t E_x = -\frac{1}{\varepsilon_0}\partial_z H_y - \frac{1}{\varepsilon_0}\partial_t P_x, \tag{2}$$

$$\partial_t u = -\gamma_2 u - \omega_0 v, \tag{3}$$

$$\partial_t v = -\gamma_2 v + \omega_0 u + 2w\Omega, \tag{4}$$

$$\partial_t w = -\gamma_1 (w - w_0) - 2v\Omega, \tag{5}$$

where E_x and H_y are the electric and magnetic fields, γ_1 and γ_2 are the population and polarization relaxation constants, respectively. $\Omega = \mathrm{d}E_x/\hbar$ is the Rabi frequency, and ω_0 is the transition frequency of the TLA medium. The macroscopic nonlinear polarization $P_x = Ndu$ is related to the off-diagonal density matrix element $\rho_{12} = (u + iv)$ and the population difference $w = \rho_{22} - \rho_{11}$ between the upper and lower states; d is the dipole moment. The refractive index is determined by the real part of ρ_{12} and the gain coefficient is proportional to the imaginary part of ρ_{12}. We employ a standard finite-difference time-domain approach for solving the full-wave Maxwell equations, and predictor-corrector method to solve the Bloch equations. The time and space increments, Δt and Δz, are chosen to ensure $c\Delta t \le \Delta z$[10].

The condition for a single ultrashort laser pulse can be expressed by the equation $\Omega_0(t=0,z) = \Omega_\mathrm{m}\cos[\omega(z+z_0)/c]\sec h[1.76(z/c+z_0/c)/\tau_0]$, where the frequency ω is the fundamental laser field frequency, Ω_m is the maximum Rabi frequency of the fundamental field and $\tau = 2ar\cosh(1/\sqrt{0.5})\tau_0$ is the FWHM of the pulse intensity envelope of the single pulse laser. The single pulse area is $A = \Omega_\mathrm{m}\tau\pi/1.76$, corresponding to the electric field of $E_0 = 2.5\times10^7$ V/cm or an intensity of $I = 1.6\times10^{12}$ W/cm^2.

In the following numerical analysis, all the material and laser parameters we adopt are[11,12]: $\tau_1 = 18$ fs, $\omega_1 = 1.2\times10^{15}$ rad^{-1}, $\omega_{01} = 1.2\times10^{15}$ rad^{-1} (or $\tau_2 = 5$ fs, $\omega_2 = 2.3\times10^{15}$ rad^{-1}, $\omega_{02} = 2.3\times10^{15}$ rad^{-1}), $z_0 = 15$ μm, $d = 0.265e$ nm, $\gamma_1^{-1} = \gamma_2^{-1} = 1$ ns. The medium is initialized with $u = v = 0$, $\omega_0 = -1$ at $t = 0$, and the choice of z_0 ensures that the pulse penetrates negligibly into the medium at $t = 0$. For such laser intensity, recent experiments on semiconductors have shown that a description in terms of two-level systems has been able to reproduce the experimental results amazingly well[21,22]. Moreover, a few important publications that study this model have existed[1,8,11,12,17,23,24].

It is well known that the area theorem can be described by the equation $\mathrm{d}A/\mathrm{d}z = -(\alpha\cdot\sin A)/2$ $(\alpha \propto N)$[1]. For various long pulses with 2π area ($\sin A = 0$), atomic densities have no influence on the propagation property and the SIT can still be recovered. However, in the case of ultrashort pulses, the propagation behavior of 2π pulses can be greatly changed by atomic densities.

We first model the propagation of 2π pulse in resonant TLA mediums with relatively lower atomic densities. Figure 1(a) represents the electric field profile (solid line) of the 18-fs pulse and the population difference (dash-dot line) at the input surface and the propagating distance $z = 48$ μm of the nonlinear medium with $N = 4\times10^{17}$ cm^{-3}. The population difference indicates that the medium is entirely inverted and returned to its initial state. Hence, one complete Rabi flopping occurs. It is shown clearly that the SIT electric field (solid line) propagates as though it is unaffected by the presence of the nonlinear medium, i.e., as a solitary wave[10]. Figure 1(b) is the spectra corresponding to Fig. 1(a) and they are in full overlapping. In this case, we find when $N \le 8\times10^{18}$ cm^{-3}, the above results are still recovered. Similar results can be obtained for other ultrashort pulses with different time duration.

With the increase of the atomic densities, the propagation behavior is greatly changed. Figures 2(a) and (b) depict the spectra of the 18-fs pulse at the propagating distance $z = 48$ μm of the nonlinear medium with $N = 4\times10^{19}$ and 8×10^{19} cm^{-3}, respectively. It is obvious that the SIT cannot be recovered. When the densities increase, the spectral evolution dramatically changes. The spectra split and the spectral components exhibit shift. Both blueshift (higher spectral components) and redshift (lower spectral components) occur distinctly in the course of propagation and the amplification of central frequency gradually disappears.

In addition, we also study the spectra of the 5-fs pulse at the propagating distance $z = 48$ μm of the nonlinear medium with 8×10^{19} and 2×10^{20} cm^{-3}, as shown in Fig. 3. It can be seen that the propagation behavior of 5-fs pulse has the same tendency with 18-fs pulse. Moreover, similar results can be obtained for other 2π ultrashort pulse with different time duration. To interpret the influence of atomic densities on the propagation property, Fig. 4 presents the electric field profile (solid line) and RO (dash line) of the 18-fs pulse at the propagating distance $z = 48$ μm of the nonlinear mediums with $N = 4\times10^{18}$, 4×10^{19}, and 8×10^{19} cm^{-3}, respectively. It is shown clearly that the 2π pulse splits and intensity decreases with increasing the atomic densities. As for

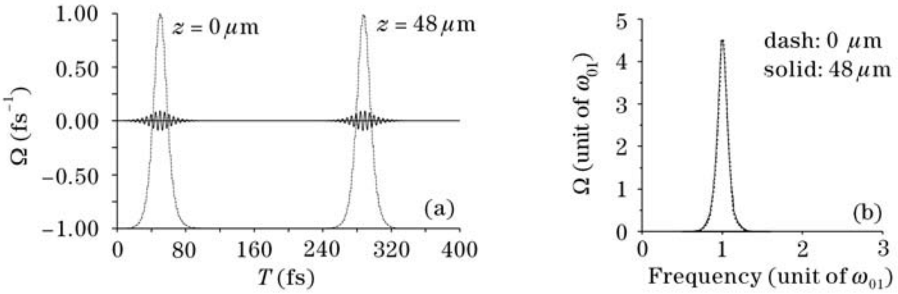

Fig. 1. (a) The electric field (solid line) and population difference (dash-dot line) of the 2π pulse at the input surface and the propagating distance of 48 μm of the nonlinear medium, (b) the spectra corresponding to (a).

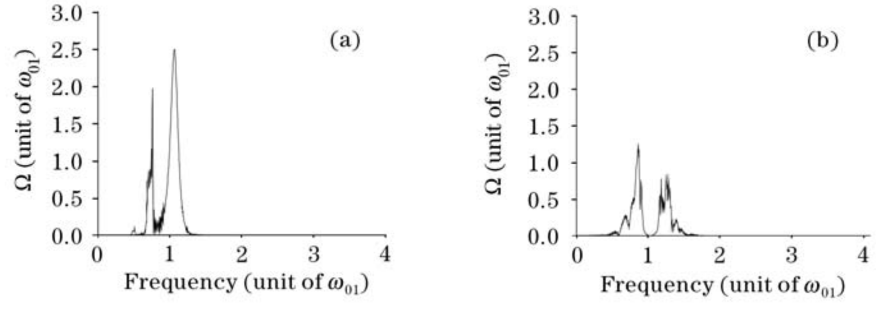

Fig. 2. The spectra of the 18-fs pulse at the propagating distance $z = 48$ μm of the nonlinear medium with $N = 4\times10^{19}$ cm^{-3} (a) and $N = 8\times10^{19}$ cm^{-3} (b).

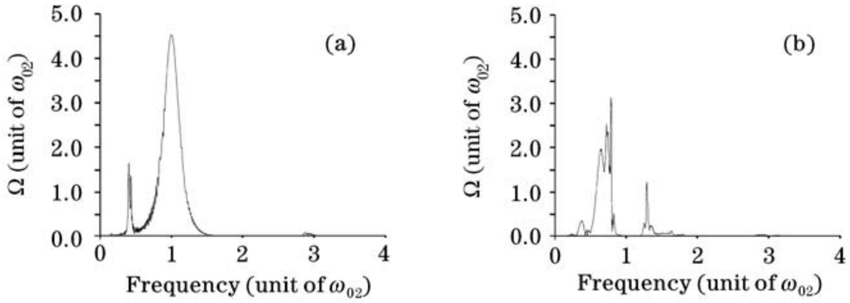

Fig. 3. The spectra of the 5-fs pulse at the propagating distance $z = 48$ μm of the nonlinear medium with $N = 8 \times 10^{19}$ cm^{-3} (a) and $N = 2 \times 10^{20}$ cm^{-3} (b).

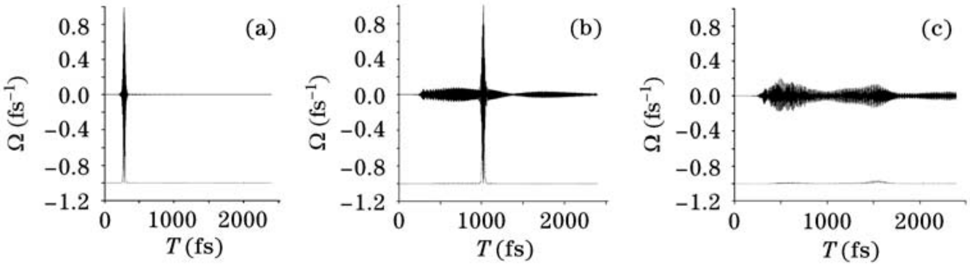

Fig. 4. The Rabi frequency (solid line) and population difference (dash line) of the 2π pulse at the propagating distance of 48 μm of the nonlinear mediums with $N = 4 \times 10^{18}$ cm^{-3} (a), $N = 4 \times 10^{19}$ cm^{-3} (b), and $N = 8 \times 10^{19}$ cm^{-3} (c).

the observed spectral transformation in Figs. 2 and 3, the physical interpretation can be given by intrapulse third-order four-wave mixing (FWM) of the type $2\omega \rightarrow \omega' + \omega''$, ω is the pulse component frequency within the bandwidth, ω' and ω'' are new resulting frequencies. Such a nonlinear process is ignored in the framework of RWA, but it is clearly presented in the exact Bloch equations and becomes significant for very short pulse[9].

In conclusion, we investigated the propagation behavior of 2π ultrashort pulse in the TLA medium with different densities by solving the full-wave M-B equations. It is demonstrated that when the atomic densities increase, the propagation behavior can be greatly changed, the SIT cannot be recovered, new features such as 2π pulse splitting, red-shift and blue-shift of the corresponding spectra arise, and the component of central frequency disappears.

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