Propagation of an arbitrary elliptically polarized few-cycle ultrashort laser pulse in resonant two-level quantum systems

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We investigate the propagation of an arbitrary elliptically polarized few-cycle ultrashort laser pulse in resonant two-level quantum systems using an iterative predictor-corrector finite-difference time-domain method. It is shown that when the initial effective area is equal to 2π, the effective area will remain invariant during the course of propagation, and a complete Rabi oscillation can be achieved. However, for an elliptically polarized few-cycle ultrashort laser pulse, polarization conversion can occur. Eventually, the laser pulse will evolve into two separate circularly polarized laser pulses with opposite helicities.

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The problem of laser pulse propagation in resonant two-level quantum systems has been a topic of research for many years. For linearly polarized laser pulses, it is well known that the propagation phenomenon can be described quite adequately by the Maxwell-Bloch equations. Under the rotating-wave approximation (RWA) [1] and the slowly varying-envelope approximation (SVEA) [2], eminent effects, such as the self-induced transparency (SIT) [3], Rabi oscillation [4], etc., have been exhibited. When the unidirectional wave propagation approximation is taken into account, the Maxwell-Bloch equations convert into the simpler and more accurate reduced Maxwell-Bloch (RMB) equations [5–7], which avoided SVEA. The RMB equations with linear polarization are integrable and are connected with a Zakharow-Shabat scattering problem. In fact, based on the finite-difference time-domain method [8], full-wave Maxwell-Bloch equations can be solved directly without invoking any of the standard approximations. A variety of new effects concerning the propagation of linearly polarized few-cycle and even subcycle ultrashort laser pulse in two-level quantum systems have been manifested [9–13] such as the time-derivative effects [9], carrier-wave Rabi oscillation [11], etc. Some of the theoretical predictions have been demonstrated experimentally. For example, carrier-wave Rabi flopping has been demonstrated experimentally in GaAs [14]. Moreover, in our previous paper, we have shown that the standard area theorem breaks down even for small-area subcycle attosecond pulses in a dense two-level medium [15].

Recently, the interaction of matter with arbitrary elliptically polarized laser pulses has attracted much interest. The selection rules for the exciting transitions in some matter require $\Delta J_z = \pm 1$. To excite these transitions, elliptically (or circularly) polarized laser pulses are necessary [16,17]. Moreover, elliptically polarized laser pulses can provide a new dimension of control parameters [18–21]. Hence, elliptically (or circularly) polarized optical pulses are highly important to achieve quantum coherent control in atoms, molecular, and semiconductor nanostructures [17,22–26].

However, in this rapidly evolving field, the nonlinear propagation of laser pulses, especially few-cycle laser pulses, with arbitrary polarization states has not been completely investigated. The vector nature of polarized light propagating in nonlinear dispersive or diffractive media has led to the discovery of a class of solitary-wave solution of the two coupled nonlinear Schrödinger equations [27–30]. Experimental observation of the elliptically polarized fundamental vector soliton of isotropic Kerr media was reported by Delqué *et al.* [31]. On the assumption that the laser pulse central frequency is far away from resonance, nonlinear guided propagation of few-cycle laser pulse with arbitrary polarization states is investigated by Stagira *et al.* [32]. Nevertheless, for resonant propagation, until recently, it has been found that the RMB equations for circularly polarized light or rotating RMB equations are indeed integrable by using the inverse scattering transform method and that they are connected with a Kaup-Newell scattering problem [33], the adopted crucial approximation works well as long as the density of atoms $N < 10^{18}$ cm^{-3} [5,34]. Very recently, Slavcheva *et al.* have found that the real-vector representation approach allows for coupling with the vectorial Maxwell equations for the optical wave propagation and thus the resulting Maxwell-pseudospin equations can be numerically solved in the time domain without any standard approximations [17]. Using this method, they demonstrated the selective excitation of atomic transitions with $\Delta J_z = 1$ or $\Delta J_z = -1$ by predefined helicity of the circularly polarized optical pulse and the formation of circularly polarized SIT solitons. As for the resonant propagation of arbitrary elliptically polarized few-cycle ultrashort laser pulses, to our knowledge, it has not yet been investigated in the literature.

In this paper, we investigate the propagation of arbitrary elliptically polarized few-cycle ultrashort laser pulses in resonant two-level systems. An effective area for arbitrary polarized laser pulse is derived, it is demonstrated that the selective excitation of atomic transitions with $\Delta J_z = 1$ or $\Delta J_z = -1$ by predefined helicity of the circularly polarized laser pulse can be explained from the point of view of the effective area. Moreover, we found that the polarization conversion of elliptical to circular polarization of few-cycle ul-

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trashort laser pulse can occur when propagating through the two-level quantum system with $\Delta J_z = \pm 1$ electric dipole transitions.

Consider an arbitrary elliptically polarized laser pulse propagating along the z direction in a two-level quantum system, elliptically polarized in a plane perpendicular to z, the interaction Hamiltonian in the case of $\Delta J_z = -1$ dipole transition can be written as follows (the case of $\Delta J_z = 1$ can be considered analogously) [17]:

$$\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} = \hbar \begin{pmatrix} 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) \\ -\frac{1}{2}(\Omega_x + i\Omega_y) & \omega_0 \end{pmatrix}. \tag{1}$$

Ω_x and Ω_y are the Rabi frequencies associated with the electric field components along the x and y axes, according to

$$\Omega_x(t) = \frac{\wp}{\hbar} E_x(t) = \frac{\wp}{\hbar} \tilde{E}_x(t)\cos(\omega t) = \tilde{\Omega}_x(t)\cos(\omega t), \tag{2a}$$

$$\Omega_y(t) = \frac{\wp}{\hbar} E_y(t) = \frac{\wp}{\hbar} \tilde{E}_y(t)\cos(\omega t + \phi) = \tilde{\Omega}_y(t)\cos(\omega t + \phi). \tag{2b}$$

$\tilde{E}_x$ and $\tilde{E}_y$ are the envelopes of the laser pulses.

The real coherence vectors $\vec{S}$ satisfy the following equation of motion [17,35,36]:

$$\dot{S}_i = f_{ijk}\gamma_j S_k, \quad i,j,k = 1,2,3, \tag{3}$$

which coincides with that derived by Feynman *et al.*: $\frac{d\vec{S}}{dt} = \gamma \nu \vec{S}$ [16]. γ_j can be calculated from $\gamma_j(t) = \frac{1}{\hbar}\text{Tr}[\hat{H}(t)\hat{\lambda}_j]$, $j = 1,2,3$, and $\hat{\lambda}_j$ are the Pauli matrices. This precession equation has the interpretation that the pseudospin vector $\vec{S}$ is precessing about the torque vector.

Consider the RWA and the resonance excitation ($\omega = \omega_0$), in the interaction picture, one can derive

$$\vec{\gamma} = \left(-\frac{1}{2}\tilde{\Omega}_x - \frac{1}{2}\tilde{\Omega}_y \sin\phi, -\frac{1}{2}\tilde{\Omega}_y \cos\phi, 0 \right). \tag{4}$$

Hence, the precession frequency is

$$\Omega_{\text{eff}}(t) = |\vec{\gamma}| = \sqrt{\frac{1}{4}(\tilde{\Omega}_x + \tilde{\Omega}_y \sin\phi)^2 + \frac{1}{4}\tilde{\Omega}_y^2 \cos^2\phi} = \frac{1}{2}\sqrt{\tilde{\Omega}_x^2 + \tilde{\Omega}_y^2 + 2\tilde{\Omega}_x\tilde{\Omega}_y \sin\phi}. \tag{5}$$

We assume that the input electric field components along the x and y axes have proportional time envelopes. Then, the effective area can be derived as

$$A_{\text{eff}} = \int_{-\infty}^{\infty} \tilde{\Omega}_{\text{eff}}(t)dt = \frac{1}{2}\sqrt{A_x^2 + A_y^2 + 2A_x A_y \sin\phi}, \tag{6}$$

where A_x and A_y are the areas of electric field components along the x and y axes, respectively. If $A_{\text{eff}} = 2\pi$, the population inversion in resonant two-level quantum system experiences a complete Rabi oscillation.

In what follows, the derived effective area will be applied to the numerical simulations of arbitrary elliptically polarized few-cycle ultrashort laser pulse propagating in resonant two-level quantum systems. The full-wave vectorial Maxwell pseudospin equations [17] are solved numerically by employing an iterative predictor-corrector finite-difference time-domain method, which avoids invoking any of the standard approximations. The initial laser pulse is given by

$$E_x(z=0,t) = \tilde{E}_x(t)\cos(\omega t) = E_{0x}\text{sec}\,h[1.76(t - t_0)/\tau_{px}]\cos(\omega t), \tag{7}$$

$$E_y(z=0,t) = \tilde{E}_y(t)\cos(\omega t) = E_{0y}\text{sec}\,h[1.76(t - t_0)/\tau_{py}]\cos(\omega t + \phi), \tag{8}$$

where ω is the laser pulse frequency, E_{0x} and E_{0y} are the initial field amplitudes, and τ_{px} and τ_{py} are the full width at half maximum of the laser pulse intensity envelopes. In the numerical analysis, the medium is initialized with $S_{30} = -1$ at $t = 0$. The parameters that we adopted are the following: $\wp = 2.65e$ Å, $N = 2 \times 10^{18}$ cm^{-3}, $\tau_{px} = \tau_{py} = 10$ fs, $\omega = \omega = 1.2$ fs^{-1}, corresponding to a wavelength $\lambda = 1.5$ μm. The results to follow can, of course, be scaled to various laser and material parameters.

First, we investigate the propagation of circularly polarized laser pulse in the resonant two-level system. From the derived effective area expression (6), we can draw the conclusion that for left circularly polarized laser pulse ($\phi = \pi/2$ and $A_x = A_y$), only when $A_x = A_y = 2\pi$, the effective area can be

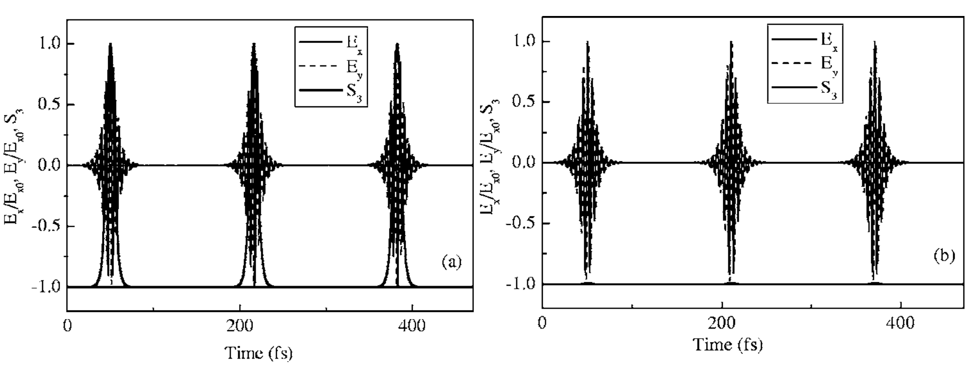

FIG. 1. The normalized electric field components ($E_{x,y}$) and the population inversion (S_3) at the simulation distance $z = 0$, 48, 96 μm. (a) represents the case of the left circularly polarized few-cycle laser pulse with $A_x = A_y = 2\pi$, $\phi = \pi/2$. (b) represents the case of the right circularly polarized few-cycle laser pulse with $A_x = A_y = 2\pi$, $\phi = 3\pi/2$.

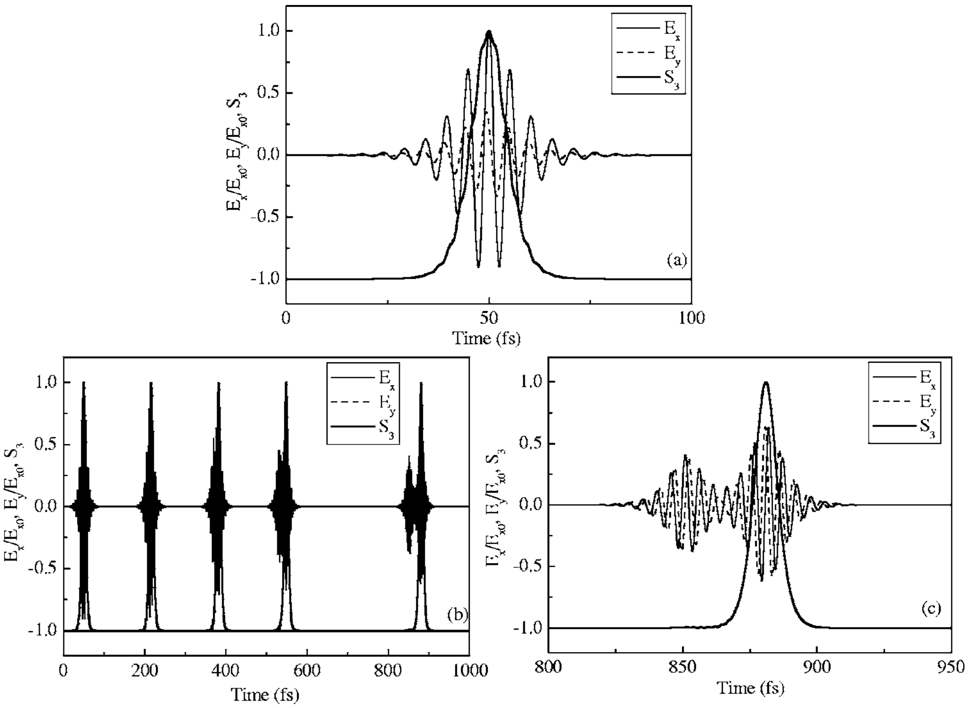

FIG. 2. (a) Normalized electric field components ($E_{x,y}$) of a 2π elliptically polarized few-cycle ultrashort laser pulse with $A_x = 3.15\pi$, $A_y = 1.10\pi$, $\phi = \pi/4$ and the population inversion (S_3) at the input surface of the nonlinear medium. (b) The time evolution of the laser pulse at the respective distances of 0, 48, 96, 144, and 240 μm. (c) As in (a) but at the simulation distance $z = 240$ μm.

equal to 2π, i.e., a complete Rabi oscillation can be achieved. While for right circularly polarized pulse ($\phi = 3\pi/2$ and $A_x = A_y$), the effective area is always equal to 0 whatever the areas of the two optical pulse components are, that means no excitation can occur. This prediction is confirmed by our numerical simulations (see Fig. 1) and that of Ref. [17]. In other word, the selective excitation of atomic transitions with $\Delta J_z = 1$ or $\Delta J_z = -1$ by predefined helicity of the circularly polarized laser pulse can be explained in the point of view of the effective area. The tiny discrepancy, for example, the small population inversion induced by the right circularly polarized pulse excitation [see Fig. 1(b)], comes from the counter-rotating terms, which are neglected in the RWA adopted during the process of the effective area derivation. For large area few-cycle laser pulse, this discrepancy will be increased.

The derived effective area is not only adapted to circularly polarized laser pulse, but also to arbitrary elliptically polarized laser pulse excitation. Figure 2 shows an example of an elliptically polarized few-cycle laser pulse with $\phi = \pi/4$ and the effective area $A_{eff} = 2\pi$ ($A_x = 3.15\pi, A_y = 1.10\pi$) propagating through the resonant two-level system. Figure 2(a) depicts the temporal development of the electric field E_x, E_y and the population inversion S_3 at the input surface of the nonlinear medium. It can be seen that the medium is completely inverted and returned to its initial state. Figure 2(b) presents the evolution of the electric fields at the respective propagation distances of 0, 48, 96, 144, and 240 μm. Complete Rabi flopping can be achieved during the course of propagation, which means the total effective area is invariant. However, the numerical simulation demonstrates that the elliptically polarized few-cycle laser pulse is not stable. During the course of propagation, amplitudes and the phase delay of the two electric components are varying. Eventually, it evolves into two separate circularly polarized laser pulses with opposite helicities [see Figs. 2(b) and 2(c)], which are stable. According to our previous analysis, the effective area of right circularly polarized laser pulse approximately equals 0 whatever the amplitude is. While for left circularly polarized laser pulse, a complete Rabi flopping can be achieved only when the effective area equals 2π. Hence, a 2π elliptically polarized few-cycle laser pulse will evolve into a 2π left circularly polarized SIT soliton. The residual energy is transferred to another 0π right circularly polarized laser pulse. For SIT soliton, the energy exchange between the system and the ultrashort laser pulse has the effect of producing

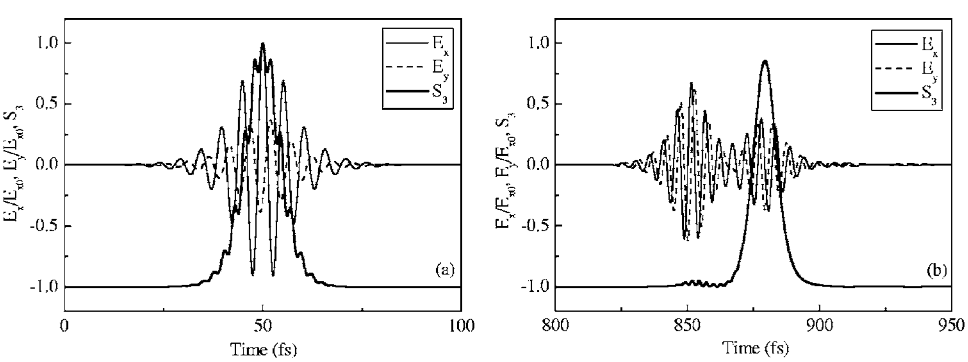

FIG. 3. (a) As in Fig. 2(a) but for a 2π elliptically polarized few-cycle laser pulse with $A_x = 5.48\pi$, $A_y = 3.12\pi$, $\phi = 5\pi/4$. (b) The corresponding normalized electric field components ($E_{x,y}$) and the population inversion (S_3) at the simulation distance $z = 240$ μm.

a pulse delay, making the pulse velocity slower [3]. While for right circularly polarized laser pulse, there is no energy exchange. It is the velocity discrepancy which results in the time delay between the generated right and left circularly polarized laser pulses.

Further consideration is given to the evolution of another elliptically polarized few-cycle laser pulse with $\phi=5\pi/4$, and $A_x=5.48\pi$, $A_y=3.12\pi$ (the effective area also equals 2π). Complete Rabi oscillation can also be achieved (see Fig. 3), which again confirmed the prediction of our derived effective area. In this case, more residual energy is transferred to the right circularly polarized laser pulse, and two separate circularly polarized laser pulses with opposite helicities can also be achieved [see Fig. 3(b)]. Similar results can be obtained for other elliptically polarized few-cycle laser pulses.

In conclusion, we have investigated the propagation of arbitrary elliptically polarized few-cycle laser pulse in resonant two-level quantum systems by numerically solving the full vector Maxwell pseudosin equations. An effective area has been derived, it is demonstrated that a complete Rabi oscillation can be achieved for arbitrary elliptically polarized ultrashort laser pulse whenever the effective area is equal to 2π. However, the elliptically polarized few-cycle laser pulses are not stable. During the course of propagation, they will evolve into two separate circularly polarized laser pulses with opposite helicities, which are more stable during the course of propagation.

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