Polarization evolution of a few-cycle ultrashort laser pulse propagating in a degenerate three-level medium

Xiaohong Song,* Weifeng Yang, Shangqing Gong, Ruxin Li, and Zhizhan Xu†
State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, People's Republic of China
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The propagation of an arbitrary polarized few-cycle ultrashort laser pulse in a degenerate three-level medium is investigated by using an iterative predictor-corrector finite-difference time-domain method. It is found that the polarization evolution of the ultrashort laser pulse is dependent not only on the initial atomic coherence of the medium but also on the polarization condition of the incident laser pulse. When the initial effective area is equal to 2π, complete linear-to-circular and circular-to-linear polarization conversion of few-cycle ultrashort laser pulses can be achieved due to the quantum interference effects between the two different transition paths.

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I. INTRODUCTION

Propagation of few-cycle ultrashort laser pulses in nonlinear media has attracted considerable interest during the past decade. When a light pulse is short enough, it interacts with atoms and molecules before they can be affected by their environment. The interaction is then described by simple quantum-mechanical rules [1]. Moreover, many features related to the propagation of ultrashort laser pulses through a nonlinear medium can contribute to ultrafast optical response measurements [2,3] and ultrashort pulse generation [4,5], etc. For example, the propagation of intense femtosecond pulses in a Raman-active medium is a strong attractor for the generation of single-cycle subfemtosecond pulses in the visible and uv spectral range [6,7]. Hence, studies of propagation dynamics of ultrashort laser pulses are of great importance.

If the temporal width of the laser pulse becomes comparable to the optical period, it is the electric field itself rather than the intensity envelope that drives the interaction. In order to analyze the nonlinear propagation effects of such laser pulses, a fully nonperturbative model is needed, which avoids invoking any of the standard approximations. Numerical techniques such as the finite-difference time-domain (FDTD) method have proven to be particularly useful and powerful tools for solving the Maxwell-Bloch equations in the time domain [8–13]. Using this technique, eminent new effects have been found [4,14–20]. For example, the formation of optical subcycle pulses has been demonstrated in dense media of two-level systems due to the pulse splitting and reshaping of few-cycle ultrashort laser pulses [4]. Another example is the notion of carrier-wave Rabi flopping, which was first introduced theoretically by Hughes [14], and subsequently demonstrated experimentally in GaAs [21].

Most of the investigations in this field consider linearly polarized few-cycle ultrashort laser pulse excitation. The electric field vector and dipole moment are confined to a single direction, and the selection rule $\Delta m=0$ is applied. In fact, the orientational distributions of the dipole moments must be considered in some transitions [22], and the vector character of the electric field can dramatically affect a number of nonlinear interactions [23–25]. The propagation of a vector electric field in resonant two-level atoms can be described by using the real-vector representation approach [26]. Based on this model, in our previous work, lossless propagation and polarization evolution of elliptically polarized few-cycle ultrashort laser pulse have been demonstrated [27]. Moreover, it has been found recently that circular-to-linear and linear-to-circular conversion of optical polarization can occur due to the natural anisotropic shape of self-assembled dots. Conversion values up to 50% can be achieved, which may have practical applications in information processing [28].

In this work, we investigate the propagation dynamics process of polarized few-cycle ultrashort laser pulses in a degenerate Λ-type three-level atomic medium. The two electric dipole transitions are between the states with $\Delta m=\pm 1$, respectively. Our results show that, due to the quantum interference effects between the two different transition paths, highly efficient circular-to-linear and linear-to-circular polarization conversion of few-cycle ultrashort laser pulses can be realized in this medium during the course of propagation. The conversion efficiency can be close to unity.

II. THEORETICAL MODEL

The system under consideration is shown in Fig. 1. The full Hamiltonian is

$$\hat{H}=\hat{H}_0+\hat{H}_{\text{int}}=\begin{pmatrix} 0 & -\vec{\wp}_{12}\cdot\vec{E} & 0 \\ -\vec{\wp}_{21}\cdot\vec{E} & \hbar\omega_0 & -\vec{\wp}_{23}\cdot\vec{E} \\ 0 & -\vec{\wp}_{32}\cdot\vec{E} & 0 \end{pmatrix}. \quad (1)$$

$\vec{\wp}_{12}=\vec{\wp}^{*}_{21}=(\wp_x-i\wp_y)/2$, $\vec{\wp}_{32}=\vec{\wp}^{*}_{23}=(\wp_x+i\wp_y)/2$ are the matrix elements of the electric dipole moments. Here we consider $\wp_x=\wp_y=\wp$. The electric field is given by

*Corresponding author. Email address: sxhywf@yahoo.com.cn
†Corresponding author. Email address: zzxu@mail.shcnc.ac.cn

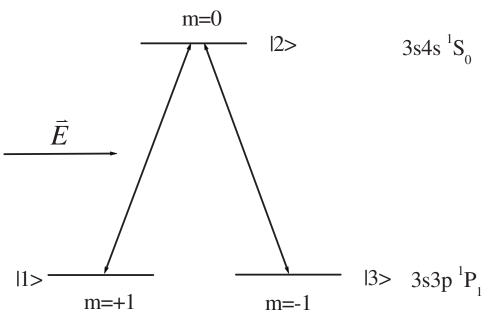

FIG. 1. An example of a degenerate Λ-type three-level system formed by the sublevels in the $3s3p\ {}^1P_1 \rightarrow 3s4s\ {}^1S_0$ transition for atomic magnesium.

$$E(z=0,t) = \hat{x}E_x(t) + \hat{y}E_y(t) = \hat{x}\tilde{E}_x(t)\cos(\omega t) + \hat{y}\tilde{E}_y(t)\cos(\omega t + \phi). \tag{2}$$

$\tilde{E}_x$ and $\tilde{E}_y$ are the pulse envelopes of the two electric components,

$$\tilde{E}_{x,y}(t) = E_{0x,0y}\,\mathrm{sech}[1.76(t-t_0)/\tau_{x,y}]. \tag{3}$$

E_{0x} and E_{0y} are the initial field amplitudes, and τ_x and τ_y are the full widths at half maximum (FWHMs) of the ultrashort laser pulse intensity envelopes. The corresponding Rabi frequencies are defined as $\Omega_x(t)=\wp E_x(t)/\hbar$ and $\Omega_y(t)=\wp E_y(t)/\hbar$, respectively. ϕ is the phase difference between the two electric components. When $\tilde{E}_x=\tilde{E}_y$, $\phi=\pi/2$ is associated with right-circularly-polarized light, while $\phi=3\pi/2$ is associated with left-circularly-polarized light.

Exploiting the symmetry of the rotations under the SU(N) group, it has been shown [13,29,30] that the density matrix $\rho(t)$ and the Hamiltonian $H(t)$ can be expressed in terms of the N^2-1 generators λ_j of the SU(N) Lie algebra. The time evolution of the density matrix can then be expressed in terms of the evolution of an (N^2-1)-dimensional real coherence vector **S**. In the degenerate Λ-type three-level medium, the time-evolution real coherence (pseudospin) vector equations are derived as follows (the deduction process is shown in Appendix A):

$$\dot{S}_1 = \frac{1}{2}\Omega_y S_3 - \omega_0 S_4 + \frac{1}{2}\Omega_x S_6 + \Omega_y S_7 - \frac{1}{T_2}S_1, \tag{4}$$

$$\dot{S}_2 = -\frac{1}{2}\Omega_y S_3 + \omega_0 S_5 - \frac{1}{2}\Omega_x S_6 - \frac{1}{2}\Omega_y S_7 + \frac{\sqrt{3}}{2}\Omega_y S_8 - \frac{1}{T_2}S_2, \tag{5}$$

$$\dot{S}_3 = -\frac{1}{2}\Omega_y S_1 + \frac{1}{2}\Omega_y S_2 + \frac{1}{2}\Omega_x S_4 - \frac{1}{2}\Omega_x S_5 - \frac{1}{T_2}S_3, \tag{6}$$

$$\dot{S}_4 = \omega_0 S_1 - \frac{1}{2}\Omega_x S_3 + \frac{1}{2}\Omega_y S_6 + \Omega_x S_7 - \frac{1}{T_2}S_4, \tag{7}$$

$$\dot{S}_5 = -\omega_0 S_2 + \frac{1}{2}\Omega_x S_3 - \frac{1}{2}\Omega_y S_6 - \frac{1}{2}\Omega_x S_7 + \frac{\sqrt{3}}{2}\Omega_x S_8 - \frac{1}{T_2}S_5, \tag{8}$$

$$\dot{S}_6 = -\frac{1}{2}\Omega_x S_1 + \frac{1}{2}\Omega_x S_2 - \frac{1}{2}\Omega_y S_4 + \frac{1}{2}\Omega_y S_5 - \frac{1}{T_2}S_6, \tag{9}$$

$$\dot{S}_7 = -\Omega_y S_1 + \frac{1}{2}\Omega_y S_2 - \Omega_x S_4 + \frac{1}{2}\Omega_x S_5 - \frac{1}{T_1}(S_7 - S_{70}), \tag{10}$$

$$\dot{S}_8 = -\frac{\sqrt{3}}{2}\Omega_y S_2 - \frac{\sqrt{3}}{2}\Omega_x S_5 - \frac{1}{T_1}(S_8 - S_{80}). \tag{11}$$

The population relaxation and dephasing times are given by T_1 and T_2, respectively. Using the same method described in our previous work [27], the effective area for an arbitrary polarized laser pulse in this medium can be written as

$$A_{eff} = \int_{-\infty}^{+\infty} \tilde{\Omega}_{eff}(t)dt = \frac{1}{\sqrt{2}}\sqrt{A_x^2 + A_y^2}, \tag{12}$$

where $A_{x,y}=(\wp/\hbar)\int_{-\infty}^{+\infty}\tilde{E}_{x,y}(t)dt$ are the areas of the electric field components along the x and y axes, respectively.

Consider the propagation of an ultrashort laser pulse along the z axis in a degenerate three-level medium, polarized in a plane perpendicular to z. The Maxwell equations take the form

$$\frac{\partial H_x}{\partial t} = \frac{1}{\mu}\frac{\partial E_y}{\partial z},$$

$$\frac{\partial H_y}{\partial t} = -\frac{1}{\mu}\frac{\partial E_x}{\partial z},$$

$$\frac{\partial E_x}{\partial t} = -\frac{1}{\varepsilon}\frac{\partial H_y}{\partial z} - \frac{1}{\varepsilon}\frac{\partial P_x}{\partial t},$$

$$\frac{\partial E_y}{\partial t} = \frac{1}{\varepsilon}\frac{\partial H_x}{\partial z} - \frac{1}{\varepsilon}\frac{\partial P_y}{\partial t}. \tag{13}$$

The Maxwell equations are coupled to the time-evolution equations of the quantum system via the macroscopic polarization $\vec{P}$. This macroscopic polarization acts as a source for the radiation field and can dramatically affect the polarization evolution of the ultrashort laser pulse during the course of propagation.

The macroscopic polarization induced in the medium is given by the expectation value of the dipole moment operator:

$$\vec{P}(t) = -Ne\langle\hat{Q}\rangle = -Ne\,\mathrm{Tr}(\hat{\rho}\hat{r}) = -N\,\mathrm{Tr}(\hat{\rho}\hat{\wp}), \tag{14}$$

where N is the density of the medium. Using Eqs. (A3a) and the expression for the electric dipole moments given above, the polarization components along x and y can be obtained as follows:

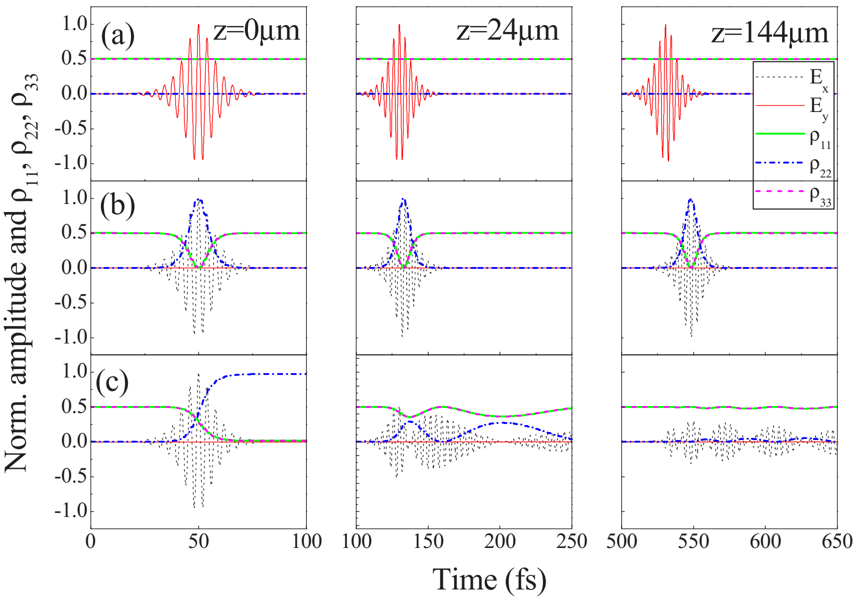

FIG. 2. (Color online) Evolution of a few-cycle laser pulse and the time-dependent populations ρ_{11}, ρ_{22}, and ρ_{33} at the simulation distances $z=0$, 24, and 144 μm. The medium is initially prepared in a coherent superposition of the lower states $c_1(z,0)=1/\sqrt{2}$, $c_2(z,0)=0$, $c_3(z,0)=1/\sqrt{2}$. (a) y-polarized ultrashort laser pulse propagation with the effective area equal to 2π ($A_x=0$, $A_y=2.828\pi$, i.e., $A_{eff}=2\pi$); (b) x-polarized ultrashort laser pulse propagation with effective area equal to 2π ($A_x=2.828\pi$, $A_y=0$, i.e., $A_{eff}=2\pi$). (c) 0.9π effective area ($A_x=1.273\pi$, $A_y=0$) x-polarized ultrashort laser pulse propagation.

$$P_x=\frac{1}{2}N\wp S_1+\frac{1}{2}N\wp S_2,\quad P_y=-\frac{1}{2}N\wp S_4-\frac{1}{2}N\wp S_5, \quad (15)$$

where N is the density of the medium. Thus, the vector Maxwell equation Eq. (13) is coupled successfully to the time-evolution equations (4)–(11).

In the numerical simulation, we employ a standard FDTD approach [8] for solving the full-wave Maxwell equations, and a predictor-corrector method to solve the pseudospin equations, which avoids invoking the slowly varying envelope approximation and the rotating wave approximation (RWA). The specific parameters are $\wp=2.08\times10^{-29}$ C m, $N=4\times10^{18}$ cm^{-3}, $\tau_x=\tau_y=10$ fs, $T_1=T_2=1$ ns, and $\omega=\omega_0=1.6$ fs^{-1}. The atomic dipole moment data and transition frequency of atomic magnesium are deduced from the NIST tables [31]. The results to follow can, of course, be scaled to various optical and material parameters.

III. RESULTS AND DISCUSSION

We first investigate the case in which the two lower states $|1\rangle$ and $|3\rangle$ are coherently prepared: $c_1(0)=1/\sqrt{2}$, $c_2(0)=0$, $c_3(0)=(1/\sqrt{2})e^{-i\psi}$ (ψ is an adjustable phase). As has been shown in Refs. [32,33], such a coherent superposition can be created with the use of stimulated Raman adiabatic passage [34]. Propagation of linearly polarized long laser pulses in a coherently prepared medium has been widely studied and exhibits a variety of interesting effects [35–41]. In what follows, we shall be interested in the effect of various initial polarization conditions and atomic coherence on the propagation dynamics process in a degenerate Λ-type three-level medium with the $\Delta m=\pm1$ transition when the pulse duration is a few optical cycles.

Consider the initial coherently prepared medium with $\psi=0$, i.e., $c_1(0)=c_3(0)=1/\sqrt{2}$. Figure 2(a) shows the propagation of a few-cycle ultrashort laser pulse with polarization direction along the y axis ($A_x=0$, $A_y=2.828\pi$, i.e., $A_{eff}=2\pi$) through the medium. It can be found that there is no population variation during the course of propagation, and the medium is then effectively transparent to the incident laser pulse. This phenomenon is the so-called electromagnetically induced transparency (EIT) [36]. In contrast, for the incident laser pulse polarized along the x axis ($A_x=2.828\pi$, $A_y=0$, i.e., $A_{eff}=2\pi$) [see Fig. 2(b)], the populations in levels $|1\rangle$ and $|3\rangle$ remain equal during the interaction, and the medium is completely inverted and returned to its initial state. No polarization evolution occurs during the course of propagation. Self-induced transparency (SIT) [42] is essentially reproduced.

The physical mechanism of the above phenomena can be understood as follows. First, the macroscopic polarization is determined by both the initial atomic coherence of the medium and the polarization condition of the initial incident laser pulse [see Eqs. (4)–(11) and (15)]. Then it acts as a source for the radiation field and will further influence the pulse evolution when propagating through the medium [see Eq. (13)]. In the first case [Fig. 2(a)], the macroscopic polarizations along the two axes induced in the medium are both equal to 0 because of the destructive quantum interference effect between the two different transition paths. Thus, the medium is totally transparent to the incident laser pulse. In the second case [Fig. 2(b)], the direction of macroscopic polarizations induced in the medium is consistent with that of the incident laser pulse. Hence, the polarization property of the laser pulse remains invariant when propagating through the medium. When the effective area is equal to 2π, SIT is essentially recovered.

The above analysis can be totally demonstrated by analytic deduction through the equations of motion for the probability amplitudes. The deduction process is shown in the Appendix B. Moreover, it should be pointed out that no RWA is involved in the deduction process. As a result, our analysis holds true even when the pulse duration is down to a few optical cycles.

When the initial effective area is less than π (for example, $A_x=1.273\pi$, $A_y=0$, i.e., $A_{eff}=0.9\pi$), contrary to the long-pulse case, in which the pulse is absorbed by the medium

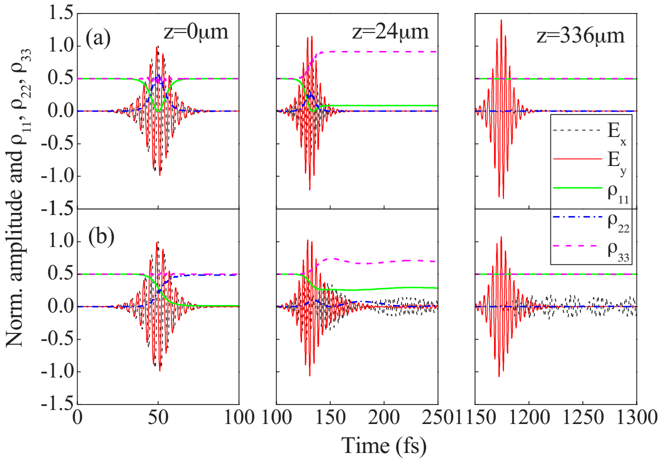

FIG. 3. (Color online) Polarization evolution of few-cycle ultrashort laser pulse propagating in a medium that is initially coherent prepared in the same way as that in Fig. 2. (a) 2π right-circularly-polarized few-cycle ultrashort laser pulse (A_x=A_y=2π, ϕ=$\pi/2$) propagation at the simulation distances z=0, 24, and 336 μm; (b) as in (a) but for the effective area equal to 0.9π (A_x=A_y=0.9π, ϕ=$\pi/2$).

[42], the x-polarized few-cycle ultrashort laser pulse will evolve into several nonvanishing 0π pulses [see Fig. 2(c)]. Thereafter, hardly any population exchanges can be seen in the medium. It should be noticed in this case that, although the pulse shape changes, the polarization property remains invariant during the course of pulse evolution, which is consistent with our analysis above.

For ultrashort laser pulses with other initial polarization conditions beyond the two special cases mentioned above, what form will they eventually evolve to? From the point of view of the macroscopic polarization, if the direction of the macroscopic polarization induced in the medium is not consistent with that of the initial incident laser pulse, it will result in variation of the polarization property of the propagating ultrashort laser pulse. Figure 3(a) shows a right-circularly-polarized few-cycle ultrashort laser pulse propagating in the same initial coherent preparation of the medium as that in Fig. 2, and the initial effective area is 2π (A_x=A_y=2π, ϕ=$\pi/2$, i.e., A_{eff}=2π). Obvious polarization conversion can be seen during the course of propagation, and the initial population exchange between the three states gradually dies out. Eventually, an EIT-type few-cycle ultrashort laser pulse linearly polarized along the y axis is formed, and thereafter the laser pulse propagates unaltered. This evolution phenomenon can be utilized to achieve efficient circular-to-linear polarization conversion of few-cycle ultrashort laser pulses.

Our inspection also shows that, for few-cycle ultrashort laser pulse, the efficiency of polarization conversion depends on the effective area of the initial incident ultrashort laser pulse. Figure 3(b) shows a right-circularly-polarized few-cycle ultrashort laser pulse with initial effective area equal to 0.9π (A_x=0.9π, A_y=0.9π, ϕ=$\pi/2$, i.e., A_{eff}=0.9π). The evolution is much different from that of Fig. 3(a). During the course of propagation, pulse splitting accompanies the polarization conversion. Though there are hardly any population

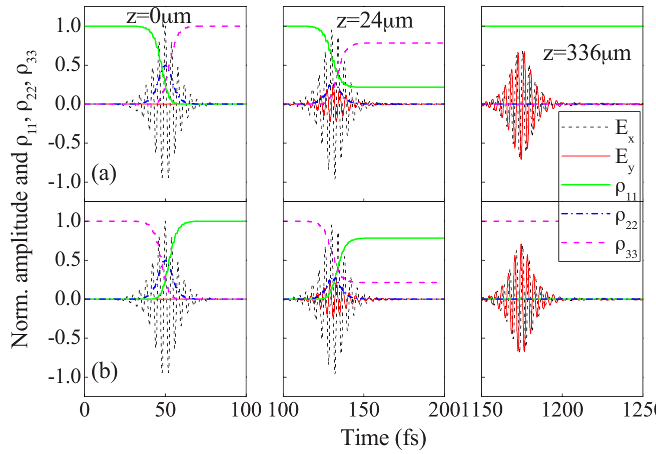

FIG. 4. (Color online) Evolution of a few-cycle ultrashort laser pulse initially linearly polarized along the x axis (A_x=2.828π, A_y=0) in a medium that is initially prepared in a single state. (a) Medium is initially prepared in the level 1>; a left-circularly-polarized few-cycle ultrashort laser pulse [$\tilde{E}_x(t)$=$\tilde{E}_y(t)$, ϕ=$3\pi/2$] is eventually formed; (b) the medium is initially prepared in the level 3>; a right-circularly-polarized few-cycle ultrashort laser pulse [$\tilde{E}_x(t)$=$\tilde{E}_y(t)$, ϕ=$\pi/2$] is eventually achieved.

exchanges after propagating 144 μm, both EIT-type and 0π SIT-type pulses coexist eventually. Hence, to achieve complete circular-to-linear polarization conversion of few-cycle ultrashort laser pulses, the effective pulse area should be equal to 2π.

In fact, polarization conversion occurs not only in an initial coherent superposition of states, but also in a single initial state. When the population is initially in the level $|1\rangle$, a left-circularly-polarized ultrashort laser pulse is prohibited according to the selection rules, which is similar to the EIT form. As a result, if a linearly polarized few-cycle ultrashort laser pulse with effective area equal to 2π (for example, A_x=2.82π, A_y=0, i.e., A_{eff}=2π) is injected into this medium, the pulse will evolve into a left-circularly-polarized few-cycle ultrashort laser pulse [$\tilde{E}_x(t)$=$\tilde{E}_y(t)$, ϕ=$3\pi/2$] [see Fig. 4(a)]. On the contrary, if the medium is initially in the level $|3\rangle$, a right-circularly-polarized few-cycle ultrashort laser pulse [$\tilde{E}_x(t)$=$\tilde{E}_y(t)$, ϕ=$\pi/2$] [see Fig. 4(b)] is eventually formed. Hence, both left- and right-circularly-polarized few-cycle ultrashort laser pulses can be achieved by controlling the initial population distribution of the medium.

IV. CONCLUSIONS

In summary, we have investigated the polarization evolution of few-cycle ultrashort laser pulse propagating in a degenerate Λ-type three-level medium with Δm=± 1. It has been found that both the initial atomic coherence of the medium and the polarization condition of the incident laser pulse can affect the macroscopic polarization of the medium, which plays an essential role in the polarization evolution of ultrashort laser pulses during the course of propagation. When the vector macroscopic polarization induced in the medium is equal to 0 or consistent with that of the incident ultrashort laser pulse, the polarization property of the ultrashort laser pulse will remain invariant during the course of propagation. Otherwise, polarization property of the laser pulse will change. By controlling the initial coherence preparation of the medium and the effective area of the incident ultrashort laser pulse, efficient linear-to-circular and circular-to-linear polarization conversion of few-cycle ultrashort laser pulses can be achieved, which might be helpful in achieving ultrafast polarization switching.

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APPENDIX A: DEDUCTION PROCESS OF THE TIME-EVOLUTION REAL COHERENCE (PSEUDOSPIN) VECTOR EQUATIONS

The dynamical evolution of an N-level atomic system is determined by the Liouville equation for the density operator:

$$i\hbar\frac{\partial\hat{\rho}}{\partial t}=[\hat{H},\hat{\rho}]. \tag{A1}$$

It has been shown [13,29,30] that the density matrix $\rho(t)$ and the Hamiltonian $H(t)$ can be expanded in terms of the N^2-1 generators of the SU(N) Lie algebra λ_j according to

$$\hat{\rho}(t)=\frac{1}{N}\hat{I}+\frac{1}{2}\sum_{j=1}^{N^2-1}S_j(t)\hat{\lambda}_j, \tag{A2a}$$

$$\hat{H}(t)=\frac{1}{2}\hbar\left[\frac{2}{N}\left(\sum_{k=1}^{N}\omega_k\right)\hat{I}+\sum_{j=1}^{N^2-1}\gamma_j(t)\hat{\lambda}_j\right], \tag{A2b}$$

where $\hbar\omega_k$ is the energy of level k, and $\hat{I}$ is a unit operator. The coefficients S_j(t) and γ_j(t) are given by

$$S_j(t)=\mathrm{Tr}[\hat{\rho}(t)\hat{\lambda}_j], \tag{A3a}$$

$$\hbar\gamma_j(t)=\mathrm{Tr}[\hat{H}(t)\hat{\lambda}_j]. \tag{A3b}$$

If the generators $\hat{\lambda}_j$ are chosen to satisfy

$$\mathrm{Tr}(\hat{\lambda}_j\hat{\lambda}_k)=2d_{jk}, \tag{A4}$$

the evolution of the density matrix [Eq. (A1)] can be expressed in terms of the evolution of an (N^2-1)-dimensional real coherent vector $\vec{S}=(S_1,S_2,\dots,S_{N^2-1})$:

$$\frac{dS_j}{dt}=\sum_{k=1}^{N^2-1}\Lambda_{jk}(t)S_k(t),\quad j=1,2,\dots,N^2-1, \tag{A5}$$

where

$$\Lambda_{jk}=-\frac{1}{2i\hbar}\mathrm{Tr}(\hat{H}[\hat{\lambda}_j,\hat{\lambda}_k]). \tag{A6}$$

When a three-level system is considered, a possible choice of the Gell-Mann SU(3) generators satisfying Eq. (A4) is given by [13,30]

$$\lambda_1=\begin{pmatrix}0&1&0\\1&0&0\\0&0&0\end{pmatrix},\quad \lambda_2=\begin{pmatrix}0&0&0\\0&0&1\\0&1&0\end{pmatrix},\quad \lambda_3=\begin{pmatrix}0&0&1\\0&0&0\\1&0&0\end{pmatrix},$$

$$\lambda_4=\begin{pmatrix}0&i&0\\-i&0&0\\0&0&0\end{pmatrix},$$

$$\lambda_5=\begin{pmatrix}0&0&0\\0&0&i\\0&-i&0\end{pmatrix},\quad \lambda_6=\begin{pmatrix}0&1&i\\0&0&0\\-i&0&0\end{pmatrix}, \tag{A7}$$

$$\lambda_7=\begin{pmatrix}-1&0&0\\0&1&0\\0&0&0\end{pmatrix},\quad \lambda_8=\frac{1}{\sqrt{3}}\begin{pmatrix}-1&0&0\\1&-1&0\\0&0&2\end{pmatrix}.$$

Because of the relation

$$[\hat{\lambda}_j,\hat{\lambda}_k]=2if_{jki}\hat{\lambda}_i. \tag{A8}$$

the equation of motion (A5) takes the form

$$\dot{S}_i=f_{ijk}\gamma_jS_k,\quad i,j,k=1,\dots,8, \tag{A9}$$

where summation over j,k is assumed, f_{ijk} is the completely antisymmetric structure constant of the SU(3) group that can be derived from Eq. (A8), and γ_j can be calculated from Eqs. (1), (A3b), and (A7).

Now we obtain the following equations for the time-evolution real coherence (pseudospin) vector:

$$\dot{S}_1=\frac{1}{2}\Omega_yS_3-\omega_0S_4+\frac{1}{2}\Omega_xS_6+\Omega_yS_7, \tag{A10}$$

$$\dot{S}_2=-\frac{1}{2}\Omega_yS_3+\omega_0S_5-\frac{1}{2}\Omega_xS_6-\frac{1}{2}\Omega_yS_7+\frac{\sqrt{3}}{2}\Omega_yS_8, \tag{A11}$$

$$\dot{S}_3=-\frac{1}{2}\Omega_yS_1+\frac{1}{2}\Omega_yS_2+\frac{1}{2}\Omega_xS_4-\frac{1}{2}\Omega_xS_5, \tag{A12}$$

$$\dot{S}_4=\omega_0S_1-\frac{1}{2}\Omega_xS_3+\frac{1}{2}\Omega_yS_6+\Omega_xS_7, \tag{A13}$$

$$\dot{S}_5=-\omega_0S_2+\frac{1}{2}\Omega_xS_3-\frac{1}{2}\Omega_yS_6-\frac{1}{2}\Omega_xS_7+\frac{\sqrt{3}}{2}\Omega_xS_8, \tag{A14}$$

$$\dot{S}_6=-\frac{1}{2}\Omega_xS_1+\frac{1}{2}\Omega_xS_2-\frac{1}{2}\Omega_yS_4+\frac{1}{2}\Omega_yS_5, \tag{A15}$$

$$\dot{S}_7 = -\Omega_y S_1 + \frac{1}{2}\Omega_y S_2 - \Omega_x S_4 + \frac{1}{2}\Omega_x S_5, \quad \text{(A16)}$$

$$\dot{S}_8 = -\frac{\sqrt{3}}{2}\Omega_y S_2 - \frac{\sqrt{3}}{2}\Omega_x S_5. \quad \text{(A17)}$$

By introducing the phenomenological decay times T_1 and T_2 [8,13,26], one can finally obtain the equations for the time-evolution real coherence (pseudospin) vector in the presence of relaxation effects [see Eqs. (4)–(11)].

APPENDIX B: POLARIZATION-INVARIANT PULSE PROPAGATION

According to the analysis above, there are two special cases in which the polarization property of the laser pulse stays invariant: the first case is that the macroscopic polarizations along the two axes are both equal to 0, then EIT-type pulse propagation can achieved; the second case is that the direction of the macroscopic polarizations induced in the medium is consistent with that of the incident laser pulse, which corresponds to SIT-type pulses. To demonstrate these analyses, we substitute Eq. (A3a) into Eq. (15), and obtain

$$P_x(t) = \frac{1}{2}N\wp S_1 + \frac{1}{2}N\wp S_2 = \frac{1}{2}N\wp(\rho_{12} + \rho_{21} + \rho_{23} + \rho_{32}) = \frac{1}{2}N\wp[c_1(t)c_2^*(t) + c_2(t)c_1^*(t) + c_2(t)c_3^*(t) + c_3(t)c_2^*(t)], \quad \text{(B1)}$$

$$P_y(t) = -\frac{1}{2}N\wp S_4 - \frac{1}{2}N\wp S_5 = \frac{i}{2}N\wp(\rho_{12} - \rho_{21} + \rho_{23} - \rho_{32}) = \frac{i}{2}N\wp[c_1(t)c_2^*(t) - c_2(t)c_1^*(t) + c_2(t)c_3^*(t) - c_3(t)c_2^*(t)]. \quad \text{(B2)}$$

We also derive the equations of motion for the probability amplitudes near the input surface of the medium. From Eq. (1) one gets (without involving the RWA)

$$i\frac{\partial}{\partial t}c_1(t) = -\frac{1}{2}[\Omega_x(z=0,t) - i\Omega_y(z=0,t)]c_2(t), \quad \text{(B3)}$$

$$i\frac{\partial}{\partial t}c_2(t) = -\frac{1}{2}[\Omega_x(z=0,t) + i\Omega_y(z=0,t)]c_1(t) + \omega_0 c_2(t) - \frac{1}{2}[\Omega_x(z=0,t) - i\Omega_y(z=0,t)]c_3(t), \quad \text{(B4)}$$

$$i\frac{\partial}{\partial t}c_3(t) = -\frac{1}{2}[\Omega_x(z=0,t) + i\Omega_y(z=0,t)]c_2(t). \quad \text{(B5)}$$

If the initial coherent preparation $c_1(0)=1/\sqrt{2}$, $c_2(0)=0$, $c_3(0)=(1/\sqrt{2})$ is considered, to achieve EIT-type pulse propagation, both $P_x(t)$ and $P_y(t)$ should be equal to 0, which means $c_2(t)=c_2(0)=0$ [see Eqs. (B1) and (B2)] is necessary. Then Eqs. (B3)–(B5) reduce to

$$\Omega_x(z=0,t) + i\Omega_y(z=0,t) + [\Omega_x(z=0,t) - i\Omega_y(z=0,t)] = 0, \quad \text{(B6)}$$

a solution of Eq. (B6) is given by

$$\Omega_x(z=0,t) = 0. \quad \text{(B7)}$$

Then the derived initial polarization condition is consistent with that in Fig. 2(a).

To achieve SIT-type pulse propagation, the system must be kept in stabilization; thus $c_3(t)/c_1(t)=c_3(0)/c_1(0)=1$ is needed. Substituting $c_3(t)=c_1(t)$ into Eqs. (B3)–(B5), one gets the necessary initial pulse polarization condition as

$$\Omega_y(z=0,t) = 0. \quad \text{(B8)}$$

Under these situations, one can derive from Eqs. (B1) and (B2)

$$P_y(z=0,t) = 0. \quad \text{(B9)}$$

As a result, the macroscopic polarization induced in the medium is consistent with that of the incident laser pulse, which does not affect the polarization property of the incident laser pulse, and SIT-type ultrashort laser pulse propagation is achieved. This again demonstrates our analysis above.

For other initial population distributions, the necessary initial polarization conditions of the incident ultrashort laser pulse for both EIT-type and SIT-type pulse propagation can also be derived using the same method.

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