Coherent and phase-sensitive phenomena of ultrashort laser pulses propagating in three-level Λ-type systems studied with the finite-difference time-domain method

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Propagation of single- and two-color hyperbolic secant femtosecond laser pulses in a three-level Λ-type quantum system is investigated by solving the Maxwell and density matrix equations with the finite-difference time-domain and Runge-Kutta methods. As a first study of our modeling, we simulate pulse self-induced transparency (SIT) in two-level systems and see how this phenomenon can be controlled by manipulating the initial relative phase between the SIT pulse and a second control pulse, provided the ratio between both pulse frequencies obeys the relation $\omega_1/\omega_2=3$. We then examine frequency down-conversion processes that are observed with single- and two-color pulses the envelope area of which is equal to or a multiple of 2π, for pulse frequencies close to resonance with the transitions of a three-level Λ medium. Also, phase-sensitive phenomena are discussed in the case of two-color ω-3ω pulses propagating resonantly in the three-level system. In particular, possibilities for such coherent control are found for frequency down-conversion processes when the ratio of the frequencies of optical transitions is $\omega_{13}/\omega_{12}=3$. The conditions for quantum control of four-wave mixing processes are also examined when the pulse frequencies of two-color ω-3ω pulses are far from any resonance of the three-level system. We demonstrate the possibility to cancel the phase sensitivity of the four-wave coupling in a Λ-type system by competition effects between optical transitions.

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I. INTRODUCTION

Noticeable achievements in modern optics, such as, for instance, generation of few-cycle pulses [1], experimental demonstration of light slowdown [2,3], its application for enhancement of nonlinear processes [3] and quantum information processing [3,4], coherent population transfer phenomena [5], or nonlinear ultrafast spectroscopy techniques and coherent control [6], among others, provide important challenges for theoretical modeling, since, e.g., in some cases the theory must go beyond the most widely used approximations, such as the rotating-wave (RWA) or the slowly varying envelope (SVEA) approximations. Therefore, the development of models which treat Maxwell equations without any approximations and which consider the dynamics of the density matrix or the Schrödinger equations for more than two levels, with account for quantum-state coherences and level populations, is of great importance.

During the past decade, the finite-difference time-domain (FDTD) method [7] has become widely explored in the optical community for solving the full set of Maxwell equations. The optical properties of the medium have been examined mainly on the basis of simple two-level models [8–14]. An extension of the two-level model has recently been used for modeling light interactions with inhomogeneously broadened materials [15]. In some papers, the dynamics for more than two levels have been taken into account, but considering an adiabatic elimination of the medium polarizations (rate equations models) to investigate light interacting with semiconductor [16] and solid-state [17] materials. The FDTD model for light propagation in a three-level quantum system (with two degenerate levels and in two spatial dimensions) has been introduced in Ref. [18] through the Gell-Mann representation of the density matrix equations. In that work, the propagation of 2π pulses through the absorbing medium has been considered in a system with a twice-degenerate upper level and a nondegenerate lower level. The models mentioned above omit all or some of the coherences between different states. It is well known, however, that optical coherences play a key role in many optical processes and phenomena [19]. Only recently, interaction of laser pulses with a V-type three-level system without such simplification has been investigated with a FDTD predictor-corrector method [20].

In this paper we introduce a model to study the interaction of ultrashort pulses with a three-level quantum system either of a Λ, *V*, or a *cascade* configuration in one spatial dimension. We solve the full Maxwell and the density matrix equations by combining the FDTD and Runge-Kutta methods. In particular, the interaction of a three-level Λ system with two initially synchronized femtosecond pulses is considered in two cases: when the pulse frequencies are close to resonance with the allowed optical transitions and when they are tuned far from resonance of any of the two optical transitions. The former case is of interest for absorption-emission resonance spectroscopy, coherent population trapping (CPT), electromagnetically induced transparency (EIT), and related phenomena. The latter case is important, for instance, in ultrafast four-wave mixing or Raman spectroscopy techniques. We next further detail these different topics.

A. Near to resonance coherent pulse propagation

In the broad area of matter interactions with light pulses, there is considerable interest in investigations of solitarylike optical pulses that propagate resonantly in a medium without substantial changes of their wave forms. It is known that pulses, the envelope area of which is equal to 2π, propagate

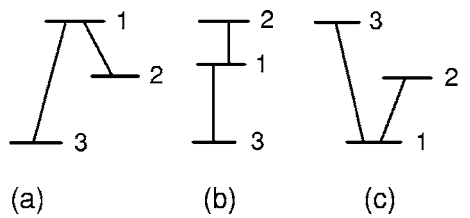

FIG. 1. Three-level quantum system of Λ- (a), *cascade*- (b), and *V*- (c) type configurations.

without substantial changes of their wave forms when their central frequency is in resonance with a two-level transition. This phenomenon was first considered for two-level media in Ref. [21] under the RWA and SVEA (see also Ref. [22]) and in Refs. [8,9] by a direct integration of the full Maxwell-Bloch equations in one spatial dimension by using a FDTD method. Such form-stable pulse propagation is well known to be caused by self-induced transparency (SIT) phenomena (in the frame of the *area theorem* [21]). It is also well known that any pulse, the area of which is larger than 2π, is split into a chain of 2π pulses during propagation [21], and therefore only pulses with an area of 2π propagate in a two-level medium with preserving wave form.

In Refs. [23,24], studies were made under the RWA and SVEA. It was shown that in a three-level system two types of form-stable pulse propagation are possible and that they can be distinguished on the basis of dynamics in the *dressed-state* representation. Such unperturbed propagation of pulses can be caused either by EIT (EIT pulses) or by SIT (SIT pulses) [24]. A large population in the dark state indicates dominance of EIT effects, while a depopulated dark state is a signature of SIT. In Ref. [24], it was shown how SIT-pulse propagation is unstable and that it is reduced to EIT-pulse propagation. One type of form-stable pulses, for which an analytical solution has been obtained [21], is the so-called hyperbolic secant (HS) pulses. In the present work we will consider propagation of such pulses by numerical methods. In particular, we will investigate the effects of two-color ultrashort pulses propagating coherently in a three-level Λ-type medium [see Fig. 1(a)] beyond the RWA and SVEA. These approximations have been disregarded in order to properly account for nonlinear effects of the emission for any detuning, which in our calculations is considered from close to resonance up to far from the one-photon resonance condition.

A theorem similar to the area theorem (called the *dark area* theorem) has been formulated in Ref. [25] for propagation of pulses in a three-level Λ-type system with strong inhomogeneous broadening of atomic transitions. This theorem describes the evolution of the envelope area of a dark field which governs the dynamics of the two lower dressed states. From this theorem it follows that under appropriate conditions form-stable propagation of two-color phase-matched pulses should be possible in a three-level system. In the present study, we numerically address the question of form-stable propagation of single and phase-matched two-color HS pulses with envelope area of the order of 2π in the case of homogeneous broadening.

B. Far from resonance coherent pulse propagation

The studies presented in this article are also devoted to the investigation of coherent control schemes by multicomponent (multicolor) ultrashort laser pulses when their frequencies are far from any resonance of the quantum system. The relative phase between the pulses can strongly modify pulse propagation in some cases and can be the origin for new spectroscopy methods, as has been recently reported in the case of ω-3ω femtosecond pulses propagating in two-level systems [13,14]. Indeed, in Ref. [13], it was discussed how the amplitude of the four-wave mixing signal at frequency 5ω (i.e., the anti-Stokes component) can be controlled by the relative phase between the ω-3ω intensity- and phase-matched pulses driving the interaction. It was then shown that such a phase-sensitive effect can be canceled by an appropriate choice of the frequency of the fields, for a given two-level transition. Such a cancellation takes place when the ac Stark frequency shifts of the energy levels caused by each pulse are compensated [14]. In the present study, we extend these investigations to the case of a three-level Λ system by examining the effect of an additional (third) level on phase-sensitive coherent phenomena. The calculations are performed beyond the RWA and SVEA since we consider ultrashort pulses and, as has been shown in Ref. [14], counterrotating terms play a key role in the cancellation of phase-sensitive four wave mixing. We will show numerically that the phase sensitivity of four-wave mixing remains in a three-level system and that one can find a field frequency ω at which the phase sensitivity is canceled due to competition effects between the different transitions of the Λ scheme.

The paper is organized as follows. In Sec. II we introduce the Maxwell and density matrix equations for the three-level system in one spatial dimension and describe the method of their solution. Phase-sensitive pulse propagation in a two-level medium is reported in Sec. III. Results of propagation of single- and two-color HS pulses which envelope area is a multiple of 2π and the central frequencies of which are in resonance with the optical transitions of a three-level Λ system are presented in Sec. IV. Phase control of four-wave mixing of two-color ω-3ω pulses detuned from the optical resonances of the Λ system is discussed in Sec. V. A summary of the results and conclusions are given in Sec. VI.

II. MODEL

Let us consider propagation of electromagnetic linearly polarized plane waves in a medium filled with three-level quantum systems (atoms or molecules) either in a Λ, *V*, or *cascade* configuration (see Fig. 1). When propagation in one spatial dimension (1D) is considered—for instance, along the x axis—only two nonzero electromagnetic field components are retained in the Maxwell equations. In this case, in a Cartesian basis, the 1D Maxwell equations can be expressed as follows:

$$\frac{\partial}{\partial t}\begin{pmatrix} B_y \\ D_z \end{pmatrix} = \frac{\partial}{\partial x}\begin{pmatrix} E_z \\ H_y \end{pmatrix}, \tag{1}$$

where E_z and D_z are the electric field and electric displacement vector components, and H_y and B_y are the magnetic field and magnetic induction vector components, respectively.

On the other hand, the evolution of a three-level quantum system in the electric-dipole approximation can be described by the equations for the density matrix elements as

$$\partial_t\sigma_{11} = (\partial_t\sigma_{11})_r - 2\Omega_E \,\mathrm{Im}\, \sigma_{13} - 2\Omega_E\beta \,\mathrm{Im}\, \sigma_{12},$$

$$\partial_t\sigma_{22} = (\partial_t\sigma_{22})_r + 2\Omega_E\beta \,\mathrm{Im}\, \sigma_{12},$$

$$\partial_t\sigma_{33} = (\partial_t\sigma_{33})_r + 2\Omega_E \,\mathrm{Im}\, \sigma_{13},$$

$$\partial_t\sigma_{12} = -(\Gamma_{12} + iq_{12}\omega_{12})\sigma_{12} + i\Omega_E[\beta(\sigma_{11} - \sigma_{22}) - \sigma_{23}^*],$$

$$\partial_t\sigma_{13} = -(\Gamma_{13} + iq_{13}\omega_{13})\sigma_{13} + i\Omega_E[(\sigma_{11} - \sigma_{33}) - \beta\sigma_{23}],$$

$$\partial_t\sigma_{23} = -(\Gamma_{23} + iq_{23}\omega_{23})\sigma_{23} + i\Omega_E(\sigma_{12}^* - \beta\sigma_{13}), \qquad (2)$$

where σ_{11}, σ_{22}, and σ_{33} represent the populations of levels 1, 2, and 3, respectively (see Fig. 1 and normalization below). The terms $(\partial_t\sigma_{ii})_r$ describe the relaxation of the population of level i in the absence of incident electromagnetic waves. These terms should be specified for each particular type of three-level system. For the Λ-type system that we have considered in the present simulations they can be expressed as $(\partial_t\sigma_{11})_r = -\gamma_1\sigma_{11}$, $(\partial_t\sigma_{22})_r = -\gamma_2\sigma_{22} + \gamma_{12}\sigma_{11}$, and $(\partial_t\sigma_{22})_r = +\gamma_{13}\sigma_{11} + \gamma_{23}\sigma_{22}$. $\gamma_l = \Sigma_m\gamma_{lm}$, with γ_{lm} being the decay rate of the lth-level population to the mth-level one; Γ_{lm} are the decay rates of the off-diagonal density matrix elements σ_{lm} (medium polarizations); and $\omega_{lm} = |E_m - E_l|/\hbar$ are the angular frequencies with respect to the optical transitions $l-m$, with $E_{l,m}$ being the energies of the quantum states l and m. The parameters q_{lm} describe the particular configuration of the three-level system as follows: $q_{lm}=1$ is for a Λ-type configuration, $q_{lm}=-1$ is for a V-type configuration, and $q_{13}=1$, $q_{12}=-1$, and $q_{23}=1$ stay for a *cascade*-type configuration (for the numbering of the quantum states, see Fig. 1). In deriving Eqs. (2) it was assumed that only the 1-2 and 1-3 transitions are allowed in the electric-dipole approximation ($d_{23}=0$). Note also that for each type of system particular initial conditions must be considered.

In Eqs. (2) and in what follows, the normalization of the variables that has been adopted is

$$\Omega_{\{E,D,B,H\}} = \{E_z, D_z, B_y, H_y\} d_{13}/\hbar,$$

$$\sigma_{lm} = \rho_{lm} N_a 2 d_{13}^2/\hbar,$$

$$\beta = d_{12}/d_{13}, \qquad (3)$$

where ρ_{lm} are the density matrix elements, d_{lm} is the effective dipole coupling coefficient (electric-dipole moment) of the l-m optical transition, and N_a is the density of polarizable atoms. For a closed quantum system, which is the case that we have considered, the probability conservation condition $\rho_{11}+\rho_{22}+\rho_{33}=1$ applies. In this case Eqs. (2) can be slightly simplified by excluding one equation of the level populations. Also, for completeness, the constitutive relations have to be added. When only the 1-2 and the 1-3 transitions are allowed in the electric-dipole approximation and for nonmagnetic materials, the constitutive expressions read as follows:

$$\Omega_E = (\Omega_D + \mathrm{Re}[\sigma_{13} + \beta\sigma_{12}])/\varepsilon_0, \qquad (4)$$

$$\Omega_B = \Omega_H \mu_0, \qquad (5)$$

where ε_0 and μ_0 are the electric and magnetic constants, respectively. Equations (1) and (2) and the constitutive relations (4) and (5) are investigated along this article with a FDTD scheme that is detailed below.

Yee's discretization (staggered in space and in time) [26] for the electric and magnetic fields has been used in our modeling. For the termination of the spatial grid and for the reduction of back reflection of light, perfectly matched layers [7,27] have been introduced on both sides of the spatial grid. In our simulation the reflection from the boundaries does not exceed 0.001%. The density matrix elements are calculated in the space points at which electric components of the field are defined. The algorithm for the solution of the Maxwell and the density matrix equations (1) and (2) can be described as follows. At the time interval $(t, t+dt)$, the Maxwell equations (1) are solved by the FDTD technique [7] and the value of the electric displacement $\Omega_D(x, t+dt)$ is obtained at the end of this time interval. Then, at time $\tau \in (t, t+dt)$, the evolution of the electric displacement is linearly approximated in time at each spatial point:

$$\Omega_D(x,\tau) = \Omega_D(x,t) + [\Omega_D(x,t+dt) - \Omega_D(x,t)]\tau/dt. \qquad (6)$$

Therefore, by substitution of Eq. (6) into Eq. (4), the density matrix equations (2) are reduced to ordinary differential equations (at each spatial point) with time-dependent coefficients. Such equations can be solved by any method for ordinary differential equations. We solve them by using a Runge-Kutta-Fehlberg method of seven-eight orders with time step control, which is used for tracking more precisely any sharp variations in the density matrix elements. The general features of the results reported below are also verified by a FDTD predictor-corrector method [8].

In this paper, in order to be consistent with previous work [13,14], we will consider the propagation of single- or two-color phase-matched HS pulses; i.e., we will assume that the input pulse has one or two spectral components at central frequencies ω_1 and/or ω_2 with initial amplitudes E_{ω_1} and/or E_{ω_2}, respectively. The wave form of such pulses at the input point ($x=0$) can be described as follows:

$$E_z(x=0,t) = E_{\omega_1}(t) + E_{\omega_2}(t) = E_{\omega_1}\frac{\cos[\omega_1(t-t_0)+\phi]}{\cosh[(t-t_0)/t_p]} + E_{\omega_2}\frac{\cos[\omega_2(t-t_0)]}{\cosh[(t-t_0)/t_p]}, \qquad (7)$$

where $\tau_p = 2\,\mathrm{arccosh}(\sqrt{2})t_p$ is the full width at half maximum (FWHM) of the pulse intensity envelope and t_0 is the time at which the pulse envelope maximum enters the grid. ϕ is the initial phase difference between the E_{ω_1} and E_{ω_2} pulses.

Two types of three-level Λ systems will be considered: (i) with arbitrary ratio of optical transition frequencies $\omega_{13}/\omega_{12} \neq 3$ and (ii) with the particular ratio of optical transition frequencies $\omega_{13}/\omega_{12}=3$. As commented above, pulse propagation in these Λ systems will be examined for two cases of tuning of the central pulse frequencies: (i) the frequencies of the E_{ω_1} and E_{ω_2} pulses being in resonance with the optical transitions—i.e., $\omega_1=\omega_{13}$ and $\omega_2=\omega_{12}$—and (ii) the frequencies of the E_{ω_1} and E_{ω_2} pulses being tuned far from any one-photon resonance of the system ($\omega_1\neq\omega_{13(12)}$ and $\omega_2\neq\omega_{13(12)}$). For the calculations presented below, the following values of the parameters have been adopted: $N_a=2\times10^{24}$ m^{-3} and $d_{13}=d_{12}=4.2\times10^{-29}$ Cm. Since we consider femtosecond pulses, the relaxation of the density matrix elements is neglected: $\gamma_1=\gamma_2=\gamma_{12}=\gamma_{13}=0$, $\Gamma_{12}=(\gamma_1+\gamma_2)/2$, $\Gamma_{13}=\gamma_1/2$, and $\Gamma_{23}=\gamma_2/2$ (i.e., $\Gamma_{lm}=0$). The initial populations of the excited states and the initial coherences between all states are set to zero $[\sigma_{11}(x,t=0)=\sigma_{22}(x,t=0)=\sigma_{12}(x,t=0)=\sigma_{13}(x,t=0)=\sigma_{23}(x,t=0)=0]$, if it is not indicated otherwise in the text. We assume that the dipole moments of both optical transitions are equal to each other—i.e., $\beta=1$. Only in Sec. III, where a two-level system is considered, we use $\beta=0$. In what follows, we will use the notation $(m\pi,n\pi)$ for pulses with $m\pi$ area of the pulse envelope at ω_1 and $n\pi$ area of the pulse envelope at ω_2.

III. TWO-COLOR PULSE PROPAGATION IN A TWO-LEVEL MEDIUM: PHASE-SENSITIVE SEPARATION OF PULSES

Recently, it has been shown theoretically that SIT-pulse propagation in a two-level medium can be modified by the presence of a second (control) field [28]. Here, we numerically show that modification of SIT-pulse propagation can be controlled not only by the amplitude of the control field (in our case the control field is another pulse), but also by the initial phase difference ϕ between SIT and control pulses, provided the frequency of the SIT pulse ($\omega_1=3\omega$) is the third harmonic of the control pulse frequency ($\omega_2=\omega$)—i.e., $\omega_1/\omega_2=3$. We consider the case of initially synchronized ω-3ω pulses [see Eq. (7)] with a pulse width such as $\tau_p=10$ fs, central wavelengths $\lambda_{3\omega}=0.8$ μm and $\lambda_\omega=2.4$ μm, and envelope area of 2π. Propagation of pulses in a two-level system is modeled by Eqs. (1) and (2) with $\beta=0$; in this case, only the dynamics of the 1-3 transition (see Fig. 1) is affected by pulse propagation. The resonance frequency of the transition is taken as $\omega_{13}=3\omega$.

In our simulations we observe that a single-color 3ω pulse propagates resonantly in the two-level medium without substantial changes of its wave form, as is expected [8,9,21,22]. The presence of the control ω pulse, however, modifies the propagation of the 3ω pulse. Due to the SIT effect [22], the 3ω resonant pulse propagates with smaller group velocity than the ω pulse (the frequency of which is far from the ω_{13} resonance). For the parameters adopted in our calculation, the group velocity of the 3ω pulse is approximately 10% of the velocity of light in vacuum. Therefore, although being initially synchronized, the ω and 3ω pulses will separate from each other during propagation. Importantly, however, in the region where the pulses overlap—i.e., for short propagation distances—the pulses still strongly interact through

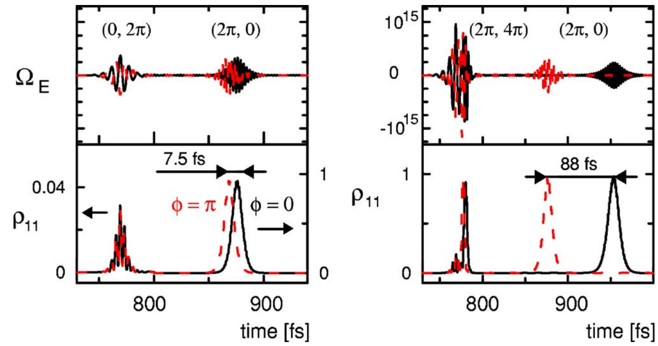

FIG. 2. (Color online) Phase-sensitive separation of $(2\pi,2\pi)$ pulse (left column) and $(4\pi,4\pi)$ pulse (right column) in a two-level system at propagation distance 220 μm. Dashed (red online) and solid (black online) lines represent results for initial phase difference $\phi=\pi$ and $\phi=0$, respectively.

phase-sensitive four-wave mixing, as is explained in Refs. [13,14]. In the left column of Fig. 2, the separation of ω-3ω pulses with envelope area of 2π is shown for two different values of the initial phase difference, $\phi=\pi$ and $\phi=0$. One can see that the separation of the pulses at $x=220$ μm is sensitive to the initial phase difference ϕ between the two incident pulses. We attribute this difference to the phase-sensitive interaction of ω and 3ω input pulses in the region where they overlap and where four-wave mixing of ω and 3ω pulses produces a phase-sensitive signal at 5ω [13,14]. Indeed, the amplitude of four-wave mixing at 5ω is smaller at $\phi=\pi$ than at $\phi=0$. The ω and 3ω pulses are therefore reshaped by four-wave mixing in such a way that the envelope amplitude of the 3ω pulse is larger in the former case (i.e., for $\phi=\pi$). As a result, after separation, for an initial phase difference such as $\phi=\pi$, the 3ω pulse has the larger envelope amplitude and, consequently (see Ref. [22]), a larger group velocity compared to the case of the initial phase difference $\phi=0$.

In the right column of Fig. 2, the phase-sensitive separation of ω-3ω pulses is presented in a case where the envelope area of the pulses is equal to 4π. Our results can be compared and are complementary to those obtained in Ref. [11]. One can see that the difference between the group velocities of the 3ω pulses (with $\phi=\pi$ and $\phi=0$) is larger for a larger envelope area. This can be understood as follows: with an increase of the initial pulse amplitudes, the phase-sensitive difference between the four-wave mixing signals at 5ω frequency in the region where the pulses are synchronized becomes larger. Therefore, the difference between the resulting envelope amplitudes after a short propagation and between the group velocities increases too compared to the case of initial 2π pulses, as one can see in Fig. 2 (right column). We hence conclude that the origin of this phase-sensitive separation is the same as described above for pulses with 2π envelope areas; namely, it is the phase-sensitive four-wave mixing effect of synchronized ω-3ω pulses [13,14].

IV. IN-RESONANCE PULSE PROPAGATION IN A THREE-LEVEL MEDIUM: PHASE-SENSITIVE FREQUENCY DOWN-CONVERSION

In this section, we consider the propagation of pulses that belong to the visible or the near-infrared spectral range and the central frequencies of which are in resonance with the optical transitions $\omega_1=\omega_{13}$ and $\omega_2=\omega_{12}$ of a three-level Λ-system [see Fig. 1(a)]. We will assume that the duration of the pulses is $\tau_p=80$ fs. The spectral width of such pulses is much smaller than the separation in frequency of the two optical transitions, and therefore any propagation effect caused by the interaction of the pulses with the adjacent transition is strongly reduced. On the other hand, since the temporal width is much shorter than the relaxation times of the medium, the relaxation of the density matrix elements can be omitted; i.e., we deal with the transitory coherent interactions. Especially, we will consider the propagation of pulses the initial envelope area of which is equal to or a multiple of 2π. The angular frequency of the 1-3 transition is taken as $\omega_{13}=2.36\times10^{15}\ \mathrm{s}^{-1}$ (which corresponds to a wavelength such as $\lambda_{13}=0.8\ \mu$m). We investigate the propagation of single-color and two-color pulses and focus our study on two particular cases: first, the ratio of the transition frequencies does not meet a special relation, and then we consider the interesting case where the relation $\omega_{13}/\omega_{12}=3$ is met.

A. Single-color $(2\pi,0)$ and $(0,2\pi)$ pulses

Let us consider the propagation of single-color $(2\pi,0)$ and $(0,2\pi)$ pulses which frequency is in resonance with either the 1-3 or the 1-2 optical transition [see Fig. 1(a)].

We show first the results of the propagation of a $(2\pi,0)$ pulse (i.e., a pulse with an area of 2π in resonance with the 1-3 transition) in the three-level Λ medium as is simulated by the theory presented in Sec. II. We consider to begin a three-level Λ system which optical transitions satisfy $\omega_{12}/\omega_{13}=0.7$. The wavelengths of resonant pulses are in this case $\lambda_1=0.8\ \mu$m and $\lambda_2=1.14\ \mu$m. Since the RWA has not been invoked in the theory, frequencies of the system that are far from the material resonances are properly taken into consideration. In these conditions, we find that the presence of a third level has a crucial effect on the propagation of the single-color $(2\pi,0)$ pulse. In Fig. 3 we show an almost total conversion of the energy from the pulse at $\omega_1=\omega_{13}$ (ω_1 pulse) into a pulse with $\omega_2=\omega_{12}$ (ω_2 pulse), at a propagation distance such as $x\approx46\ \mu$m in this case. Therefore, the ω_1 pulse is converted into a ω_2 pulse with approximately hyperbolic secant shape. We find that the envelope area of the output ω_2 pulse is $\approx97\%$ of the area of the input ω_1 pulse. We attribute this pulse frequency-conversion effect to the Raman process which induces absorption at ω_1 and emission at ω_2. We have verified this conclusion by simulating Eqs. (1) and (2) with pulse frequencies detuned from the one-photon resonance and keeping the two-photon resonance condition.

The conversion process can be enhanced by an additional input pulse at ω_2. The addition of an input wave at ω_2 with small amplitude decreases the distance at which the switching takes place. This is shown in the inset in Fig. 3. This decrease is caused by the stimulated emission in the 1-2 transition and by the stimulated Raman process in the 3-1-2 transitions in the presence of the ω_2 pulse.

We now consider a three-level Λ-type system with the particular ratio of optical transition frequencies $\omega_{13}/\omega_{12}=3$.

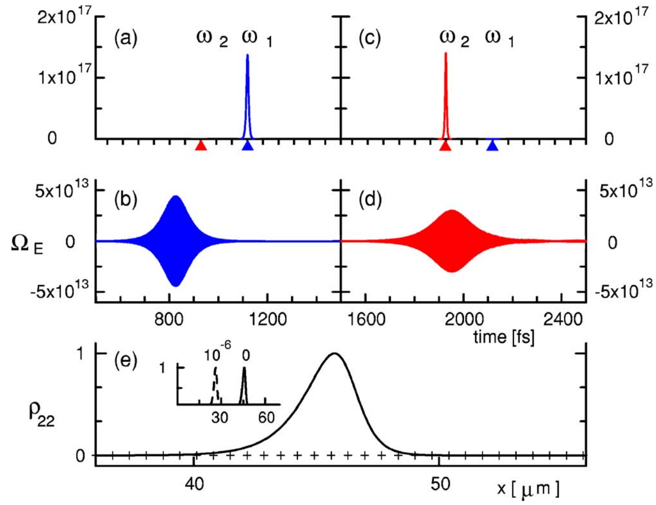

FIG. 3. (Color online) Frequency down-conversion effect observed during propagation of a $(2\pi,0)$ pulse in a three-level system with $\omega_{12}/\omega_{13}=0.7$: optical spectra (a), (c) and time evolution of the electric field (b), (d) of the pulse before (left column) and after (right column) switching into a ω_2 pulse. Distribution of population of level 2 (e) after the pulse has passed throughout the system. Crosses in (e) denote the distribution of population of level 1. In the inset in (e) the solid line indicates the distribution of population of level 2 after $(2\pi,0)$ pulse propagation (ω_1 pulse propagation in the absence of ω_2 pulse) and the dashed line indicates the distribution of population of level 2 after $(2\pi,\varepsilon)$ pulse propagation—i.e., ω_1 pulse propagation in the presence of a weak ω_2 pulse with $\varepsilon=2\pi\times10^{-6}$ being the area of the additional weak ω_2 pulse.

The wavelengths of two resonant pulses are $\lambda_1=0.8\ \mu$m and $\lambda_2=2.4\ \mu$m. In this case ($\omega_{13}/\omega_{12}=3$), the propagation effects of a $(2\pi,0)$ pulse (with frequency $\omega_1=\omega_{13}=3\omega$) are similar as those observed above for an arbitrary ratio of optical transition frequencies. Therefore, frequency down-conversion of the pulse will take place at some propagation distance; see Fig. 4(a). The difference in the present case is that the conversion from $\omega_1=3\omega$ to $\omega_2=\omega$ takes place at a longer distance ($x=193\ \mu$m) than in the case where $\omega_{13}\neq3\omega_{12}$ ($x=46\ \mu$m), as shown in Figs. 4(a) and 3(e), respectively. We think that this retardation is due to the fact that in the case $\omega_{13}=3\omega_{12}$ there is a larger difference in frequency between the transitions 3-1 and 1-2 with respect to the case that we have considered with $\omega_{13}\neq3\omega_{12}$. The retardation may also benefit from several higher-order processes that occur in this particular level configuration (e.g., two- and three-photon absorption in the 3-1 and 3-2 transitions), which may slightly inhibit the frequency conversion process. It is also worth noting that the envelope area of the output pulse with $\omega_2=\omega$ is in this case approximately 71% of the area of the input pulse with $\omega_1=3\omega$ for the parameters that we have considered in the simulation. Therefore, we find that the amplitude of the converted pulse is smaller than in the previous case where $\omega_{13}\neq3\omega_{12}$.

In Sec. III we have shown that propagation of a pulse with frequency $\omega_1=3\omega$ being in resonance with the transition of a two-level system ($\omega_{13}=3\omega$) can be modified by an additional pulse with frequency $\omega_2=\omega$ and that this effect is sensitive to the initial relative phase between the two pulses. We now

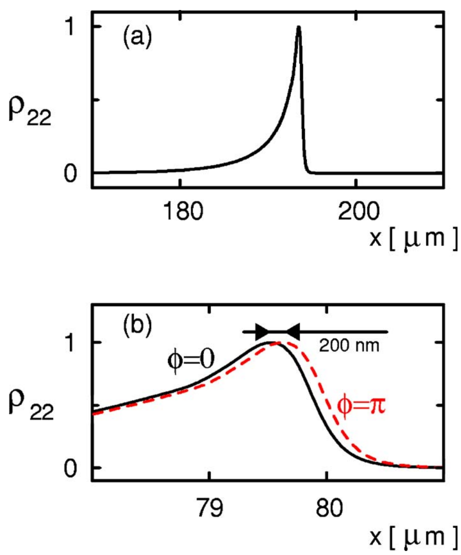

FIG. 4. (Color online) Distributions of the population of level 2 in a three-level system with $\omega_{13}/\omega_{12}=3$ after the propagation of a $(2\pi,0)$ 3ω pulse (a) and $(2\pi,\varepsilon)$ 3ω-ω pulses (b) with $\varepsilon=2\pi \times 10^{-4}$ being the area of additional weak pulse at frequency $\omega_2=\omega$. In (b) the dashed line (red online) represents the result for the initial difference between the pulse phases $\phi=\pi$ and solid line (black online) for the case $\phi=0$.

aim to investigate the possibilities of phase control of the frequency down-conversion phenomenon by manipulating the initial relative phase between a ω_1 pulse (with pulse area of 2π) and an additional weak ω_2 pulse (with small pulse area $\varepsilon \ll 2\pi$) in the three-level Λ medium with $\omega_{13}=3\omega_{12}$; i.e., we consider propagation of $(2\pi,\varepsilon)$ pulses. We find that the distance at which the conversion takes place depends on the initial phase difference between the input ω_1 and the weak ω_2 pulse, as is shown in Fig. 4(b). For the parameters used in this simulation the difference in the switching distance is approximately 200 nm. Finally, we have also found that the distance at which the switching [frequency down-conversion of the $(2\pi,\varepsilon)$ pulse] occurs decreases with an increase of the amplitude ε of the weak ω_2 pulse, in both the cases $\omega_{13}/\omega_{12}=3$ and $\omega_{13}/\omega_{12}\neq 3$.

We next report on the results of the study of the propagation of a $(0,2\pi)$ pulse (i.e., a pulse with an area of 2π in resonance with the 1-2 transition) in a three-level Λ system with distinct configurations. Figure 5 shows the dynamics of the electric field, excited-level populations, and real parts of the coherences for three separate positions (x=55, 110, and 165 μm) in the propagation of the pulse. In the left column of Fig. 5, we show that propagation of a $(0,2\pi)$ pulse when $\omega_{13}/\omega_{12}\neq 3$ is almost wave form preserving. We also show in the left column of Fig. 5 that a $(0,2\pi)$ pulse induces transient changes and oscillations in all medium polarizations ρ_{ij} and all level populations. In our simulations these transient changes are rather small, because the initial populations of excited levels (levels 1 and 2) are equal to zero and the frequency ω_2 of the incident pulse is far from resonance of the 1-2 transition. After the pulse propagation, all medium variables (polarizations and level populations) return to their initial values for $\omega_{13}/\omega_{12}\neq 3$ [see Figs. 5(b)–5(f)].

In the right column of Fig. 5, we show the results of the propagation of a $(0,2\pi)$ pulse when $\omega_{13}/\omega_{12}=3$. Due to the particular relation between the two transitions of the system in this case, the propagation of a $(0,2\pi)$ pulse with frequency $\omega_2=\omega_{12}$ is affected by nonlinear multiphoton processes, such as hyper-Raman [30] and also two- and three-photon absorption in the 3-2 and 3-1 transitions, respectively. These processes lead to a weak decay of the ω_2 pulse. As a consequence, we find that during propagation the wave form of a $(0,2\pi)$ pulse is not completely preserved when $\omega_{13}/\omega_{12}=3$. On the other hand, we also observe that when the $(0,2\pi)$ pulse passes through the medium, it increases the population of level 2 [see Fig. 5(i)], a phenomenon that will be further commented on below, and excites all the coherences of the system [see Figs. 5(j)–5(l)] by means of the multiphoton processes commented on above.

B. Two-color strong pulses

We now consider propagation of two-color HS $(m2\pi,n2\pi)$ pulses the frequencies of which (ω_1 and ω_2) are in resonance with the two frequencies ($\omega_{13}=\omega_1$ and $\omega_{12}=\omega_2$) of the optical transitions of a three-level Λ-type medium [see Fig. 1(a)]. To emphasize the propagation effects in this study we consider pulses with a time pulse width as short as τ_p=10 fs, which consequently have a strong intensity. In this case, we find that form-stable propagation does not exist due to different nonlinear processes that effect pulse propagation and lead to perturbation of the pulse wave form. We show next that the modeling considered in our study, which considers pulse propagation beyond RWA and SVEA on the basis of the Maxwell and density matrix equations, is appropriate for a description of such nonlinear processes. Indeed, one of such processes is Raman scattering, which affects form-preserving propagation of pulses in the system. In Fig. 6 the spectrum of two-color $(2\pi,2\pi)$ pulses with wavelengths $\lambda_1=0.8$ μm and $\lambda_2=1.14$ μm is shown after a propagation distance in the medium such as 12.5 μm. We clearly observe contributions from Stokes and anti-Stokes processes, which are also shown schematically in the insets. The pulse with the larger frequency (ω_1) induces an anti-Stokes field at a higher frequency through the process shown schematically by solid lines in the right inset of Fig. 6, while the pulse with the lower frequency (ω_2) produces Stokes emission at a lower frequency, as is shown by solid lines in the inset on the left. Such Raman processes have been experimentally observed and are reported in Ref. [29]. It is worth noting that the Stokes field produced by the pulse with the lower frequency (ω_2) cannot be present for frequencies $\omega_{12}<\omega_{13}/2$, which is the case in the particular case of our study where $\omega_{12}=\omega_{13}/3$.

Another process that has a strong effect on the two-color pulse propagation is stimulated emission. In Fig. 7 we consider the dynamics of the excited-level populations [1 and 2 in Fig. 1(a)] at a given spatial point in the case of the propagation of $(m2\pi,n2\pi)$ resonant pulses in a $\omega_{13}=3\omega_{12}$ three-level Λ-type configuration, also illustrating the single-color cases. The more general case with $\omega_{13}\neq 3\omega_{12}$ presents a similar behavior and has been omitted here. From Fig. 7 we

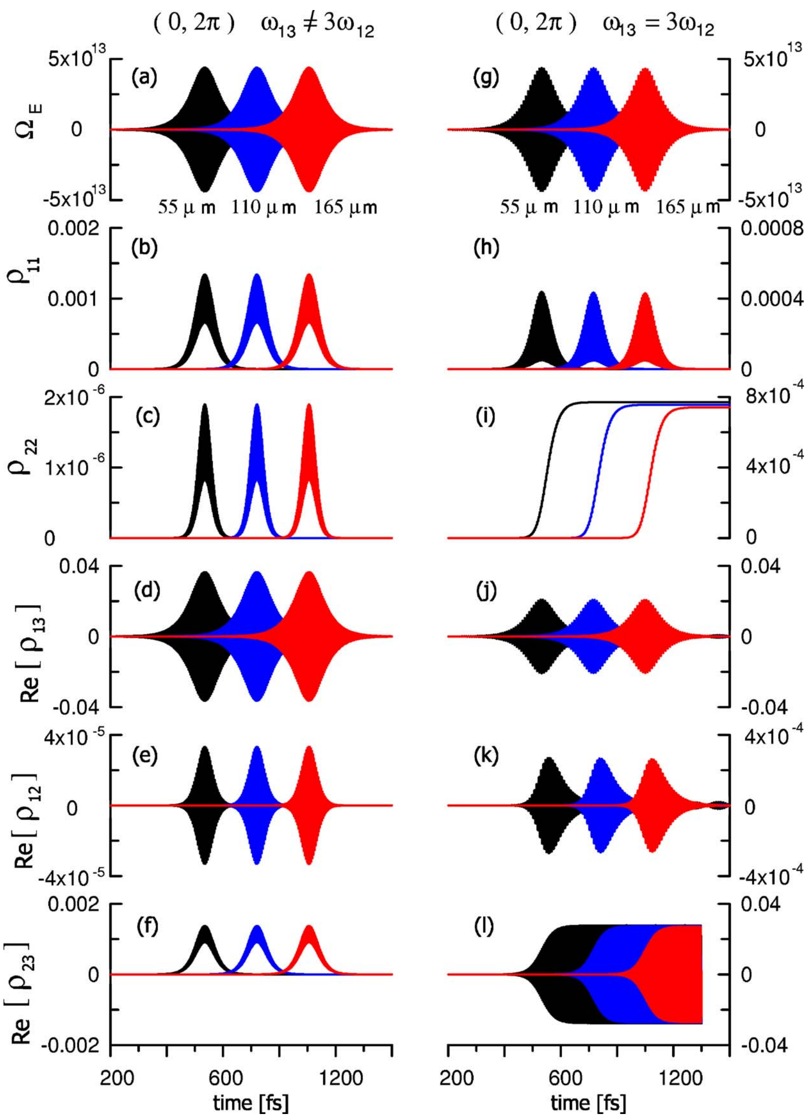

FIG. 5. (Color online) Dynamics of the electric field (a), (g), excited-level populations (b), (c), (h), (i), and real parts of the coherences (d)–(f), (j)–(l) during $(0,2\pi)$ pulse propagation in a three-level Λ-type medium with the ratio of optical transition frequencies $\omega_{12}=0.7\times\omega_{13}$ (left column) and $\omega_{13}=3\omega_{12}$ (right column). The plots are shown at three separate points in the medium, as indicated.

see that any incident pulse initiates an increase in the population of level 1. If the initial population of level 2 is small, as is the case in our simulations, population inversion in the transition 1-2 will be created at the pulse front, which will result in emission of light at ω_{12} provided some light at this frequency is present to induce stimulated emission [see Figs. 7(b)–7(d)]. This process together with Raman scattering will result in population of level 2, as has been commented on above. The process can be boosted now by the presence of both strong waves at $\omega_1=\omega_{13}$ and $\omega_2=\omega_{12}$, as can be observed in Figs. 7(c) and 7(d)). We therefore remark on the observation of efficient population transfer from level 3 to level 2 in the case of the propagation of two-color pulses. Since the initial population of level 2 is equal to zero, the SIT effect is mainly present in the 1-3 transition, and therefore the group velocity of the ω_2 pulse is larger than that of the ω_1 pulse. Hence, the ω_2 pulse goes ahead of the ω_1 one during propagation through the three-level Λ system. This counterintuitive sequence results in an effective stimulated Raman adiabatic passage (STIRAP) mechanism [5].

On the other hand, in Fig. 8 the spatial distributions of the population of level 2 are shown at different ratios of ampli-

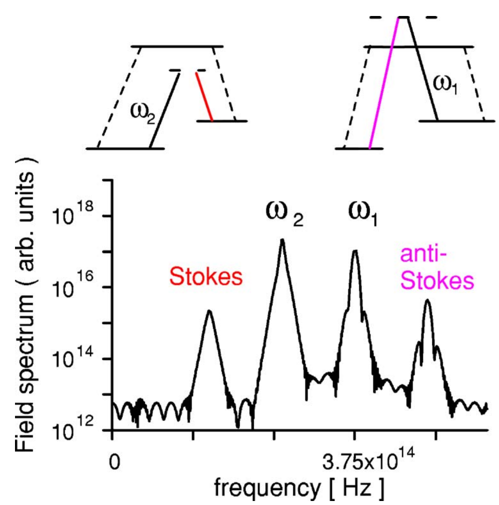

FIG. 6. (Color online) Spectrum in logarithmic scale of the Raman scattering of two-color $(2\pi,2\pi)$ resonant pulses in a three-level Λ system with $\omega_{12}=0.7\omega_{13}$ at a propagation distance such as 12.5 μm. Solid lines in the insets show schematically the processes leading to the generation of the lowest- and highest-frequency Raman contributions in this case.

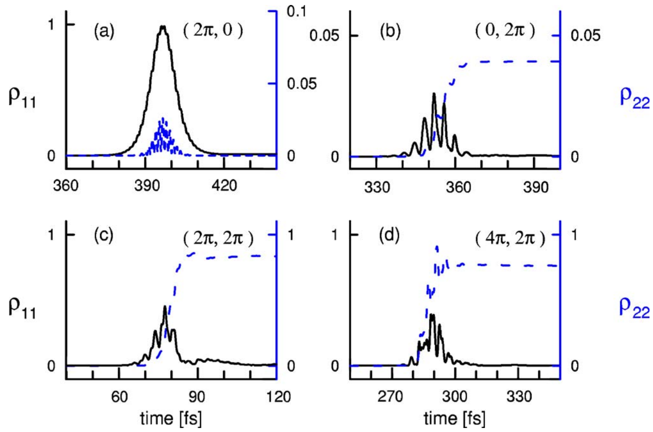

FIG. 7. (Color online) Dynamics of the populations of levels 1 (black solid line) and 2 (blue dashed line) induced by single-color (a), (b) and two-color (c), (d) resonant pulses at the propagation distances 145 μm (a), (b), 25 μm (c), and 72.5 μm (d), in a three-level Λ system with $\omega_{13}=3\omega_{13}$.

tudes $(m2\pi, n2\pi)$ and for two values of the initial phase difference of the input $\omega_1=3\omega_2$ resonant pulses, always with $\omega_{13}=3\omega_{12}$. From Fig. 8 it follows that the initial phase difference between the two pulses has a strong influence on the spatial distribution of the coherent population transfer effect. The maxima of the population distributions show the space region at which there is efficient frequency down-conversion (transfer of energy from the ω_1 pulse into the ω_2 pulse, which implies coherent population transfer from level 3 to level 2). We see that the position of these regions (peaks in the population of level 2) is governed by the relative amplitudes and also by the relative phase between the pulses. Interestingly, it is clear that at fixed pulse amplitudes the population transfer can be controlled solely by the relative phase between the pulses. Calculations at different values of the relaxation rates γ_1 and γ_2 in the range $[0, 1\ \text{ps}^{-1}]$ have been also performed in order to test the effect of decoherence in the system, and it has been found that the phase-sensitive population transfer effect remains. However, we find that the maximum value of the resulting population in level 2 decreases with increasing the value of the relaxation rates γ_1 and γ_2. As is expected, we have not found such phase-sensitive population transfer in other three-level Λ configurations with ratios different from $\omega_{13}/\omega_{12}=3$.

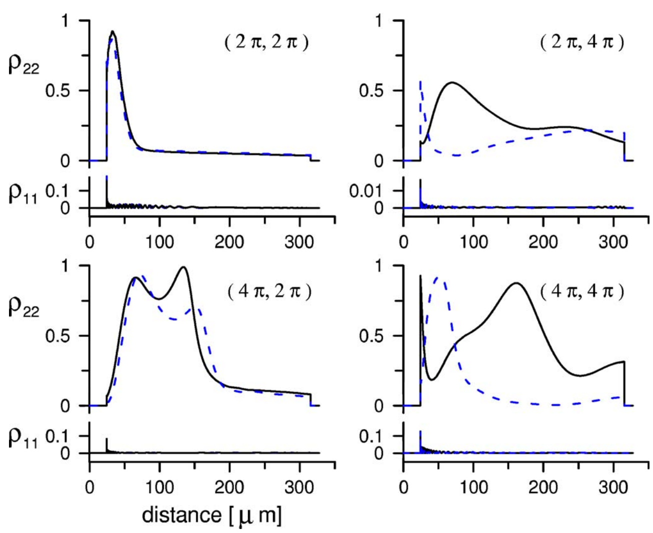

FIG. 8. (Color online) Phase sensitivity of the spatial distribution of the population of level 2 after propagation through the three-level Λ system with $\omega_{13}=3\omega_{12}$ of two-color $\omega_1=3\omega_2$ resonant $(m2\pi, n2\pi)$ pulses with initial phase difference $\phi=\pi$ (blue dashed lines) and $\phi=0$ (black solid lines). The zero and corresponding abrupt changes in ρ_{22} on the left and right sides define the free space before and after the medium on the spatial grid used in the calculations.

V. FAR-FROM-RESONANCE TWO-COLOR PULSE PROPAGATION: PHASE SENSITIVITY OF FOUR-WAVE MIXING

In this section, two-color ω-3ω initially synchronized pulses in the infrared region with spectral components such as $\lambda_{3\omega}=3.0\ \mu$m and $\lambda_{\omega}=9.0\ \mu$m are considered. Since the optical transition frequencies belong to the infrared region, the initial populations of levels have been defined by the Boltzmann distribution at room temperature. Initial coherences between all states are equal to zero. We assume here that both spectral components of the incident two-color pulses are far from any resonance of the three-level Λ system. In this case, the ratio of the optical transition frequencies, $\xi=\omega_{12}/\omega_{13}$, is taken as one of the control parameters, and the pulse temporal width is assumed to be $\tau_p=300$ fs. We will investigate phase-sensitive four-wave mixing phenomena and the existence of a point at which this sensitivity is suppressed by extending previous results reported by some of us [13,14] to the case of three-level Λ systems.

Four-wave mixing of a ω-3ω pulse produces emission at 5ω frequency. In a two-level system, the efficiency of four-wave mixing for different values of the relative phase (ϕ) between the ω and 3ω components of the incident pulse is related to the following parameter [14]:

$$\eta_{13} = (\omega_{13}/\omega)^2. \tag{8}$$

In our notation, the parameter η_{13} indicates how far from the ω_{13} resonance the frequency components of the incident pulse are. For short propagation distances [14], with $\eta_{13} < 5$ the amplitude of the 5ω component is larger for $\phi=\pi$, while with $\eta_{13}>5$ the amplitude of the 5ω component is larger for $\phi=0$. In the two-level approximation, ac Stark frequency shifts of the energy levels induced by the ω and 3ω spectral components compensate each other at $\eta_{13}=5$. The important result reported in Ref. [14] concludes that at this point—i.e., at $\eta_{13}=5$—the phase sensitivity of the 5ω four-wave mixing component disappears.

In a three-level Λ system a similar parameter can be introduced for the adjacent (1-2) transition as follows:

$$\eta_{12} = (\omega_{12}/\omega)^2 = \eta_{13}\xi^2. \tag{9}$$

Of course, simultaneous compensation of energy level shifts (induced by the ac Stark effect) on both transitions of the three-level Λ system is not feasible. However, when level 2 is not close to the upper level 1 [see Fig. 1(a)], one can

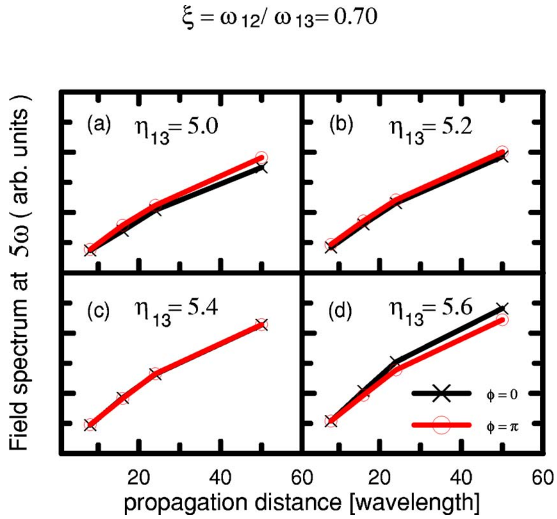

FIG. 9. (Color online) Spatial dependence of the maximum of the amplitude at 5ω in the propagation of two-color ω-3ω initially synchronized HS pulses. The initial area of the pulses is equal to 1.9π. The ratio of the optical transition frequencies of the three-level system is $\xi=\omega_{12}/\omega_{13}=0.7$. The results are shown for different values of η_{13} as indicated. Red lines with circles are for $\phi=\pi$, while black lines with crosses are for $\phi=0$. Cancellation of phase-sensitive four-wave mixing takes place in this case at $\eta_{13}=5.4$.

expect that there is a frequency ω at which the phase-sensitive character of the four-wave mixing process can also be suppressed in this case. We have indeed been able to find this cancellation effect in the three-level system for a value of η_{13} slightly higher than 5. This can be explained as follows: for $\eta_{13}>5$, the 1-3 transition gives preference for a larger amplitude of the four-wave mixing signal at 5ω for zero initial phase difference ($\phi=0$). Because in a three-level Λ system the ratio $\xi<1$ always holds, one can always have the relation $\eta_{12}<5$. In such a case, the 1-2 transition will give preference for a larger amplitude of the 5ω four-wave mixing component for $\phi=\pi$. Therefore, when $\eta_{13}>5$ and $\eta_{12}<5$, the two optical transitions of the three-level system will compete with each other in the four-wave mixing process and there will be a value of η_{12} (or equivalently a value of ξ) at which the changes in the amplitude of the wave-mixing signal at 5ω will cancel each other. This is shown in Figs. 9 and 10.

In Fig. 9, the spatial dependences of the spectral amplitude of the four-wave mixing signal at 5ω are presented for

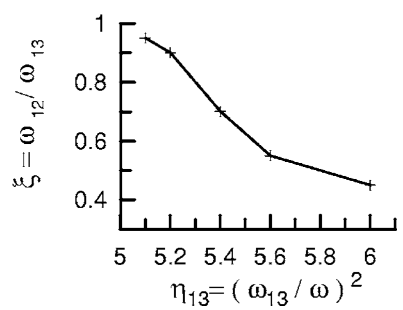

FIG. 10. Curve of phase-insensitive four-wave mixing in a three-level Λ system, plotted in the plane ξ-η_{13}.

the case of a three-level Λ system with the particular value of $\xi=0.7$ and for different values of the parameter η_{13} (i.e., for different values of the frequencies of the ω-3ω two-color pulse). From Fig. 9 one can see that the amplitude of the mixing signal at 5ω is controlled by the phase difference between the spectral components of the incident pulse. Moreover, it is evident that there is a value of η_{13} (or equivalently a frequency ω) at which the phase sensitivity of the four-wave mixing process disappears. For the particular case of $\xi=0.7$, the point at which the phase-sensitivity effect cancels is $\eta_{13}=5.4$ [see Fig. 9(c)]. In Fig. 9, only the curves for phase differences $\varphi=0$ and $\varphi=\pi$ are presented, although we have verified that the same phase-sensitivity cancellation remains for any value of the phase difference. We have calculated the phase-sensitivity cancellation point for other ξ values and illustrate the results in Fig. 10, where the curve of phase-insensitive four-wave mixing is represented in the plane (η_{13},ξ). It is worth noting that for small values of ξ the value of η_{13} has to be increased considerably in order to maintain $\eta_{12}<5$. In such a case, since the 3ω spectral component approaches the resonant frequency (ω_{13}) of the 1-3 transition, the phase-sensitive effect created by the 1-3 transition can no be longer compensated by the weaker effect created in the 1-2 transition. Also, in a more general case, the conditions for phase-insensitive four-wave mixing would depend on other parameters too, such as the relative strength (β) of the optical transitions or the relaxation rates (γ_{ij}) of the density matrix elements. The additional effects of these parameters are presently under investigation.

VI. SUMMARY AND CONCLUSIONS

In this paper we have introduced a FDTD and Runge-Kutta method for the solution of the Maxwell and density matrix equations of a three-level system beyond the RWA and SVEA in one spatial dimension. We have investigated coherent effects in the propagation of ultrashort pulses in two-level and three-level Λ systems. We have considered single pulses and also initially synchronized two-frequency pulses. It has been shown that the presented model is helpful for investigations of a wide range of nonlinear phenomena in two- and three-level systems. Since the RWA is not invoked in the theory, investigations of resonant and nonresonant phenomena can be performed.

It has been found that in a two-level medium, the propagation of two-color ω-3ω pulses depends on their initial phase difference. This phase-sensitivity effect is linked to the phase-sensitive phenomena observed in the four-wave mixing of ultrashort ω-3ω pulses in the region where they overlap, as was reported before in [13,14]. Such phase-sensitive phenomena might be used for optimal control of ultrashort pulse propagation (for instance, for precise tuning of the 3ω pulse retardation time) in a resonant two-level medium. Moreover, the phase-sensitive pulse-separation phenomena reported here might be also applied as a control mechanism for the generation of single-cycle pulses at 3ω frequency, by exploring the scheme of $m2\pi$ pulse separation into a chain of m pulses with 2π areas, as was proposed in Ref. [10].

Coherent propagation of femtosecond single- and two-color pulses has been investigated in the case where the pulse frequencies are in resonance with the optical transitions of a three-level Λ-type system. It has been found that in a three-level Λ system with arbitrary ratio of optical transition frequencies, the propagation of single-color $(2\pi,0)$ pulses does not exhibit preservation of the wave form. Such pulses undergo frequency down-conversion [conversion from a $(2\pi,0)$ pulse into a $(0,2\pi)$ one] at some propagation distance. In the small space domain where the conversion takes place, the population is efficiently transferred from the ground level 3 into the level 2 during the frequency down-conversion process. In this context, we have also studied the particular case where $\omega_{13}/\omega_{12}=3$. Although it is difficult to find a quantum system with a particular ratio of optical transition frequencies such as $\omega_{13}/\omega_{12}=3$, the phenomena that we investigate could still be accomplished in some measure by applying appropriate external fields to the system. We find that when $\omega_{13}/\omega_{12}=3$ is met, frequency down-conversion of a $(2\pi,\varepsilon)$ pulse is sensitive to the initial relative phase when an initially synchronized pulse with weak amplitude in resonance with the adjacent 1-2 transition is present. On the other hand, we have studied the propagation of single-color $(0,2\pi)$ pulses, the frequency of which is in resonance with a transition that is uncoupled from the ground state, in the electric-dipole approximation sense, and have obtained form-stable propagation in the general case. In the particular case where $\omega_{13}/\omega_{12}=3$ is met, propagation of $(0,2\pi)$ pulses is perturbed by multiphoton resonant processes, which populate excited states and induce coherences in the three-level Λ system.

We have observed that frequency down-conversion is enhanced in the case of two-color $(m2\pi,n2\pi)$ resonant pulse propagation in the three-level Λ-type system, due to the presence of a comparably strong pulse at the frequency of the adjacent 1-2 transition. Moreover, frequency down-conversion in the three-level Λ system with the particular ratio of optical transition frequencies, $\omega_{13}/\omega_{12}=3$, is governed by the relative phase of the ω-3ω $(m2\pi,n2\pi)$ resonantly propagating pulses. Namely, the position of the regions where there is effective population transfer to level 2 (i.e., effective frequency down-conversion) depends on the initial phase difference between the pulses. Phase-sensitive manipulation of population transfer could be useful, for instance, in information processing [3,4].

Finally, the propagation of femtosecond two-color ω-3ω initially synchronized hyperbolic-secant pulses in a three-level Λ system with an arbitrary ratio between the optical transition frequencies has been investigated in the case where the pulse frequencies are far from any one-photon resonance of the system. It has been shown that in a three-level Λ medium, the process of four-wave mixing of phase- and amplitude-matched ω-3ω ultrashort pulses is governed by the initial phase difference of these pulses, which extends the previous results found in two-level systems [13,14]. Moreover, it has been shown that coherent Stark nonlinear spectroscopy [14] is also feasible in three-level systems, for which we have shown the conditions for the suppression of four-wave mixing phase-sensitive phenomena.

In conclusion, we have investigated different coherent propagation effects in two- and three-level systems, including retardation, coherent population transfer, and frequency down-conversion phenomena, and have shown that the particular relation $\omega_{13}/\omega_{12}=3$ between the transitions involved in the three-level configuration together with the consideration of resonant two-color ω-3ω pulses might bring about many interesting phase-sensitive phenomena. Our investigation can have potential applications in fields as diverse as optoelectronics and materials research, in coherent control schemes, and in biological applications such as spectroscopy and imaging, among others.

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