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Complicated pulse structure and chaotic fronts in multispecies self-induced transparency

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Self-induced-transparency phenomena in systems with multiple atomic or molecular species are investigated theoretically. It is shown that under certain conditions relating relative concentrations and transition probabilities that (in the absence of loss processes) complex highly structured solitons can propagate through the system. For some parameters the inversion of some of the species and the field envelope can have very many maxima. Under other (more typical) conditions it is shown that a chaotic infinite train of irregular maxima and minima can propagate.

I. INTRODUCTION

The Hahn-McCall self-induced-transparency phenomenon involves absorption and stimulated emission processes which, for a pulse of sufficient amplitude, lead to unattenuated propagation in an absorbing medium when dissipative processes are negligible.[1-3] The structure of these pulses is simple; the electric field pulse envelope is symmetric and has a single maximum. Several authors have considered the effect of degeneracy on self-induced transparency.[4-9] In these cases the degeneracy was in the levels of a single species. If the system has more than one species with the same level spacing but with different concentrations and transition probability it is reasonable to expect that complex and interesting pulse structures and other phenomena could result. The purpose of this communication is to demonstrate that this is, in fact, the case.

In Sec. II we set forth the conservation laws for multispecies systems in the weakly varying envelope approximation. The system is specialized to the case of δ-function line shape, and analytical solutions for pulses of complex structure are obtained in Sec. III. Chaotic infinite pulse trains are discussed in Sec. IV and concluding remarks are made in Sec. V.

II. BASIC EQUATIONS OF MULTISPECIES SELF-INDUCED TRANSPARENCY

The one-species theory may be generalized in a rather straightforward way.[4] Since the purpose of the present study is to point out that very interesting phenomena can occur, we make the simplest possible assumptions that retain the essence of the phenomena. In Sec. V we shall briefly discuss relaxing some of the simplifications used.

A. Assumptions

The following assumptions are made (and are those usually taken for a simple exposition of the one-species case; see Refs. 1–9):

(1) The light frequency ω is identical with the frequency of the common energy spacing for all species.

(2) The line-shape functions for all species are taken to be delta functions.

(3) The density matrix for each atom is independent except as they are coupled through the light field.

(4) Collisional and other dissipative processes are neglected.

(5) The slowly varying envelope approximation is assumed.

(6) No phase shifts in the electric field occur.

Our goal is to find solitons under these assumptions.

B. Soliton EOM's

The derivation of the equations of motion (EOM) for constant velocity propagation is essentially the same as for the single-species case and are sketched in the Appendix. In that appendix we have introduced characteristic length, time, inversion, etc., so that the wave equations take the form

$$\frac{dv_j}{dt} = \mu_j \mathcal{E} w_j , \tag{2.1}$$

$$\frac{dw_j}{dt} = -\mu_j \mathcal{E} v_j , \tag{2.2}$$

$$\frac{d\mathcal{E}}{dt} = \sum_{i=1}^{s} \gamma_i \mu_i v_i , \tag{2.3}$$

where t is the time, $\mathcal{E}, w_i, v_i$ are quantities observed as the wave passes a given point in space, $\mathcal{E}$ is the electric field envelope function, and w_j, iv_j, γ_j, and μ_j are the inversion, off-diagonal single-atom density matrix, mole fraction, and dipole transition moment of species j of the s species system. All quantities are made dimensionless as outlined in the Appendix. For a pulse solution $w_i \to +1$, $v_i \to 0$, $\mathcal{E} \to 0$ as $t \to \pm\infty$. For the one-species case, $s = 1$, these equations reduce to the usual results.[1-3]

C. Conservation laws

The single-species conservation laws are easily generalized. Multiplying (2.1) by v_j and (2.2) by w_j and adding we get, upon invoking the boundary conditions in advance of the wave's arrival ($t \to -\infty$),

$$v_j^2 + w_j^2 = 1\,. \tag{2.4}$$

Next, multiplying (2.3) by $\mathcal{E}$ and using (2.2) we get

$$\tfrac{1}{2}\mathcal{E}^2 = \sum_{j=1}^{s} \gamma_j (1 - w_j)\,. \tag{2.5}$$

With these laws one can eliminate the $\{v_j\}$ and $\mathcal{E}$ from the problem and obtain closed equations in the scaled inversions $\{w_j\}$.

It is convenient to introduce a polar representation[1-3] such that

$$w_i = \cos\theta_i, \quad v_i = \sin\theta_i\,. \tag{2.6}$$

With this we obtain

$$\frac{d\theta_i}{dt} = \mu_i \mathcal{E}. \tag{2.7}$$

From this it is clear that the angular variables are related to the pulse area, i.e., since $\theta_i(-\infty) = 0$,

$$\theta_i(t) = \mu_i \int_{-\infty}^{t} dt\, \mathcal{E}(t')\,. \tag{2.8}$$

The field-inversion law (2.5) becomes

$$\mathcal{E} = \left(2 \sum_{i=1}^{s} \gamma_i (1 - \cos\theta_i) \right)^{1/2}, \tag{2.9}$$

and hence (2.7) can be reexpressed as a closed set of equations relating the angular variables, namely,

$$\frac{d\theta_j}{dt} = \mu_j \left(2 \sum_{i=1}^{s} \gamma_i (1 - \cos\theta_i) \right)^{1/2}. \tag{2.10}$$

A final, very useful, relation can be obtained among the θ_i by dividing both sides of (2.7) by a similar equation for θ_j; we obtain [using $\theta_k(-\infty) = 0$]

$$\theta_i = \frac{\mu_i}{\mu_j}\, \theta_j\,, \tag{2.11a}$$

$$\theta_{i \neq 1} = \mu_i \theta_1, \quad \mu_1 \equiv 1\,. \tag{2.11b}$$

D. Constant phase pulse criterion

The boundary conditions for pulse propagation $w_i(\pm\infty) = 1$ imply

$$\begin{aligned} &\theta_i(-\infty) = 0\,, \\ &\theta_i(+\infty) = 2\pi l_i\,, \end{aligned} \tag{2.12}$$

where l_i is some integer. From this we get $\theta_i(+\infty)/\theta_j(+\infty) = l_i/l_j$ and combining this with (2.11) we find

$$\frac{\mu_i}{\mu_j} = \frac{l_i}{l_j}\,. \tag{2.13}$$

Hence for propagation of a pulse through a multispecies medium, all the transition dipole moment ratios μ_i/μ_j must be rational fractions. This does not prove that pulse propagation is impossible for irrational ratios— we simply assert that they cannot propagate at constant phase. (See the Appendix for a definition of the phase.) We shall treat the more general case of varying phase phenomena elsewhere. Indeed it has been necessary to include the phase in the coupling of self-induced transparency to excitons.[10] Let us now concentrate on the most experimentally accessible case of two species to understand the nature of the rational fraction pulses.

III. TWO-SPECIES PULSES

For the two-species case a number of interesting pulse structures emerge. Let $\mu = \mu_2/\mu_1$, $\gamma = \gamma_1$. The equation of motion for the θ_1 variable then takes the form

$$\frac{d\theta_1}{dt} = \{2[\gamma(1-\cos\theta_1) + (1-\gamma)(1-\cos\mu\theta_1)]\}^{1/2}. \tag{3.1}$$

Note that $\gamma = N_1/N_T$ = the mole fraction of species 1 and hence is a readily controllable experimental parameter. Let us now discuss several special cases.

A. The case $\mu = 2$

For this case trigonometric identities may be used to reduce (3.1) to a form given in the tables.[11] Letting $\tau = 2(4-3\gamma)^{1/2} t$ and choosing the origin of coordinates such that $w_1(0) = -1$ we get

$$w_1(\tau) = \cos\theta_1(\tau) = 1 - \frac{4(4-3\gamma)}{8 - 7\gamma + \gamma\cosh\tau}\,, \tag{3.2}$$

$$w_2(\tau) = \cos 2\theta_1(\tau) = 2w_1^2 - 1\,, \tag{3.3}$$

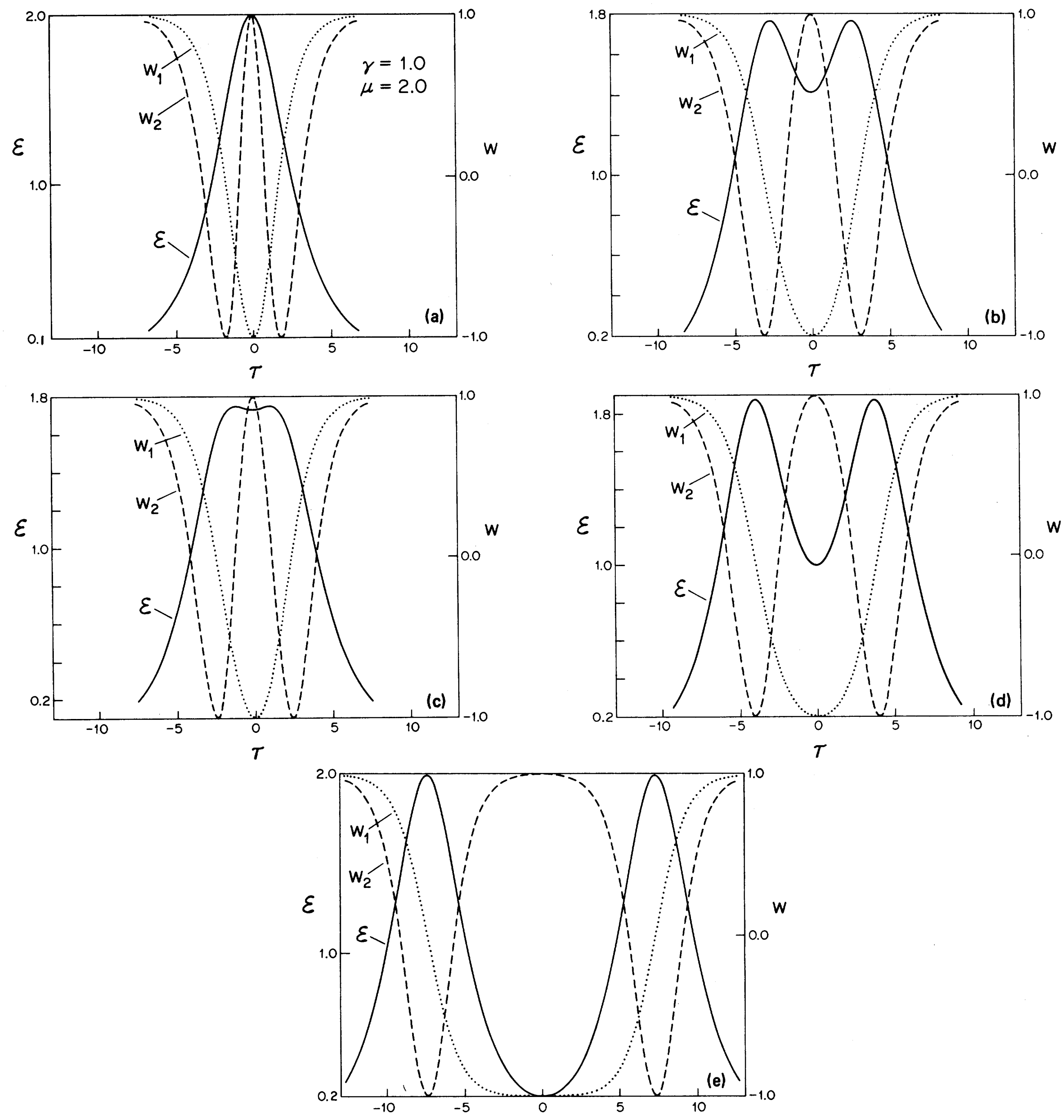

FIG. 1. Two-species self-induced transparency for the case $\mu_2/\mu_1 \equiv \mu = 2$. The parameter γ is the mole fraction of species 1. Notice that as γ decreases the character of the pulse changes from essentially the one-species singly peaked pulse ($\gamma = 1$) to a doublet of strongly interacting pulses. As $\gamma \rightarrow 0$ the pulse breaks up into two essentially single-(species 2) species pulses. (a) $\gamma = 1.0$, (b) $\gamma = 0.5$, (c) $\gamma = 0.75$, (d) $\gamma = 0.25$, and (e) $\gamma = 0.1$.

$$\mathcal{E}^2 = \frac{8\gamma(4-3\gamma)^2(1+\cosh\tau)}{[\gamma(1+\cosh\tau)+8(1-\gamma)]^2}\,. \tag{3.4}$$

It is clear from (3.2) that $w_1(\tau)$ has a single minimum. Hence w_2 must, as seen by (3.3), have two minima. Most interesting is the electric field envelope $\mathcal{E}$ which can be expressed more generally as

$$\mathcal{E}(\tau) = \{2[\gamma(1-\cos\theta_1) + (1-\gamma)(1-\cos\mu\theta_1)]\}^{1/2}\,. \tag{3.5}$$

From this it is clear that since $\cos\theta_1$ goes through one minimum, $\cos 2\theta_1$ has two minima and hence

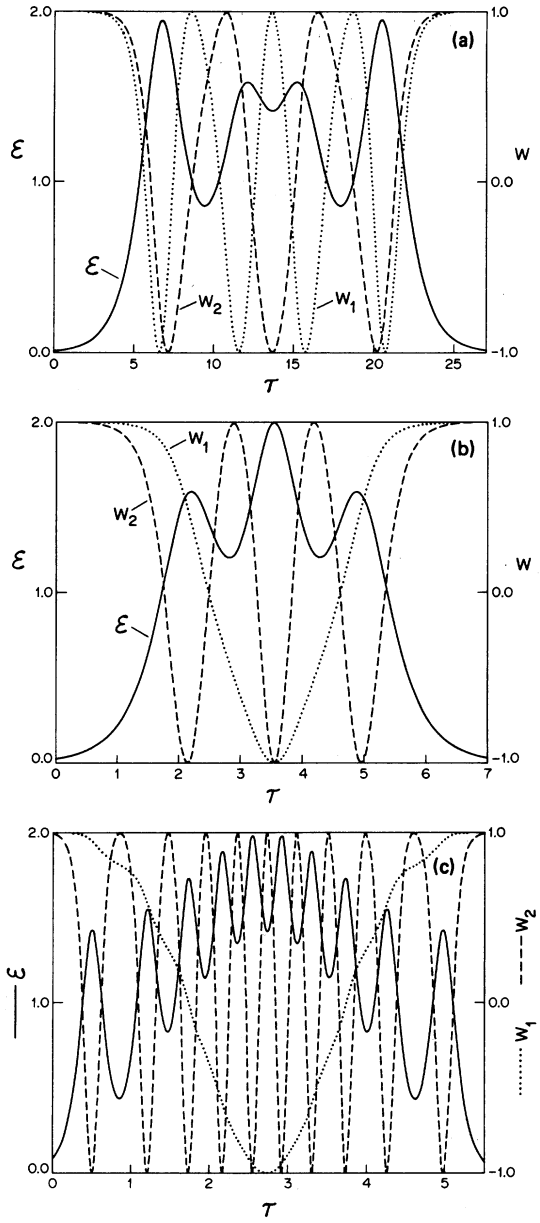

FIG. 2. Examples of two-species self-induced-transparency solitons for differing dipole moment ratios. The complex structures propagate with constant velocity and can be transformed significantly by varying the dipole moment ratio (μ) at $\gamma = 0.5$. The multiple maxima in the field envelope cannot be thought of as a train of independent pulses (except as one of the species are of negligible concentration); $\mu = 0.75$, 3, and 10 in (a), (b), and (c), respectively.

if γ is small, then $\mathcal{E}$ will be double peaked. In Fig. 1 we see several examples of mole fractions γ for the $\mu = 2$ case. Notice that by changing γ we can change the $\mathcal{E}$ profile from a single- to a double-peaked pulse. For $\gamma \neq 0$ the pulse structure is not that of two one-species pulses but a strong interaction between the field, inversions, and off-diagonal density matrix.

B. More complex rational pulses

As pointed out above one may find constant phase pulse solutions when μ is a rational fraction. To illustrate the possibilities we have run several numerical solutions of (3.1) for various choices of parameters. The results are summarized in Fig. 2. Notice that for a given transition dipole moment ratio the pulse can be changed dramatically by varying the mole fraction γ_1.

IV. CHAOTIC FRONTS

For irrational μ and nonzero γ we have shown that there are no constant phase pulse solutions. However, we have found other very meaningful types of solutions that can propagate with constant velocity and phase. These phenomena are infinite trains of highly coupled pulses that occur at irregular intervals and intensities. A typical case is shown in Fig. 3 for a $\mu = \sqrt{2}$ chaotic front.[12] Notice that the field envelope has no apparent simple predictable pattern. A situation closely related to this case has been studied experimentally by using Zeeman induced degeneracy of Na vapor; it was not possible to observe complete chaos since only an isolated input pulse was used although initial breakup was observed.[9] Also for this case, unlike in the present study, the mole fraction of the species is not at the control of the experimentalist.

The irrational case corresponds to evolution of the $\mathcal{E}$ field envelope as a nonlinear combination of functions of two noncommensurate periods. It has been shown[13] that such multiple noncommensurate periodic motion is transformed to true chaotic motion by a slight change in the dynamical equations. Thus we have used the word chaos to describe the phenomenon. The space-filling character in $\mathcal{E}$, w_1, w_2 space is also seen in Figs. 3(b)–3(d). The attractor is the surface defined by (3.5) for $-1 \leq w_1$, $w_2 \leq +1$ as seen in the projections of the attractor (or more precisely the trajectories corresponding to the wave profiles) on the three indicated planes.

Figure 4 shows how a general case approaches the rational case as μ converges on a given simple rational case. Notice that the wave tends to bunch up into $\mu = 2$ -like double pulses as $\mu \to 2$, although all doublets are seen to still interact strongly through the inversions. Between any two rational fractions there are infinitely many rational fractions. However, most of them are ra-

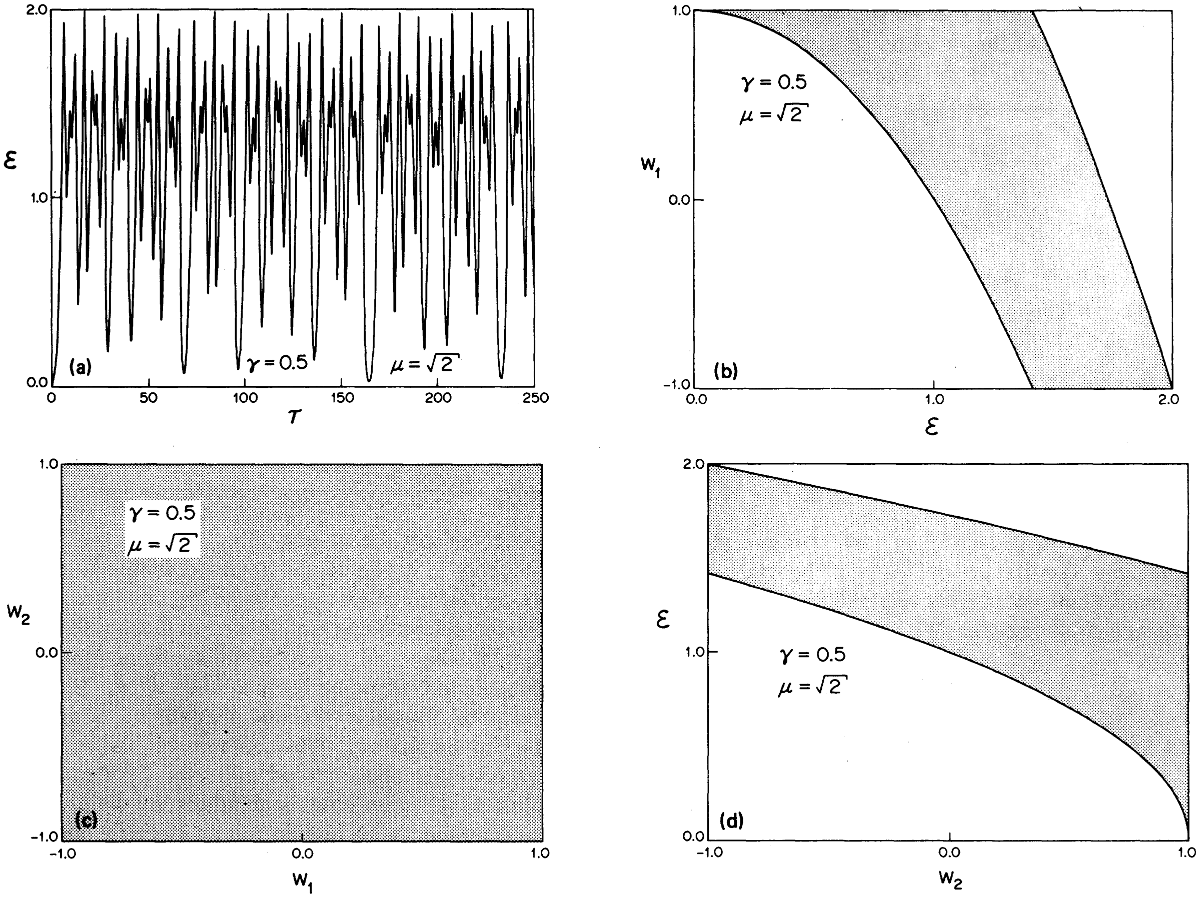

FIG. 3. A front is defined as being a propagating disturbance that leaves a system in a different state in its wake than in advance of its arrival (Ref. 12). In part (a) we see such a phenomenon corresponding to a semi-infinite sequence of irregular electric field intensity pulses. The inversion fluctuates, at irregular intervals, between ±1. Parts (b)–(d) show the space-filling character of phase plane trajectories for irrational cases.

tios of such large numbers as to lead to essentially infinitely long chains of pulses and will, because of fluctuations, be impossible to discriminate from the irrational case. Ratios of smaller numbers can, however, lead to readily observable structures, as seen in Figs. 1 and 2.

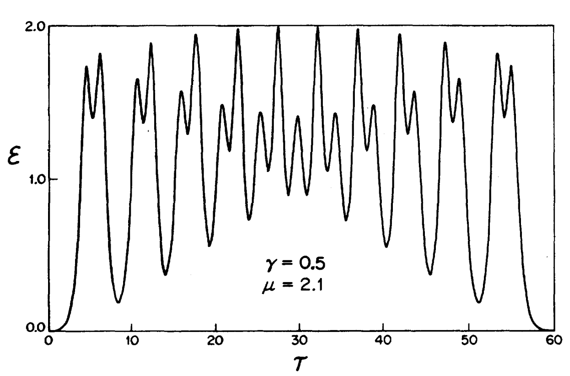

FIG. 4. Note the tendency to break up into ($\mu = 2$)-like doublets similar to those in Fig. 1. However, in this case there is a strong interdoublet coupling.

V. CONCLUDING REMARKS

Multiple-species self-induced transparency allows for an interesting set of phenomena. Our results show that for the rational μ case pulses of complex structures may propagate unattemated in the absence of relaxation processes. Furthermore, chaotic fronts in the form of a semi-infinite irregular pulse train may propagate in the irrational μ case. In both cases the wave form depends strongly on the mole fractions of the components.

An experimental verification of the present phenomena is as follows. Let us assume that μ for the two-species case is a ratio of integers, at least one of which is sufficiently small. Let us impose a pulse of light at one end and measure the velocity λ_μ through the medium and the maximum aplitude $\mathcal{E}_{\max}$ of the excited soliton. In the one-species case the velocity is simply related to the pulse width. Here, however, the complex structure of the pulse makes $\mathcal{E}_{\max}$ a more convenient parametrization. For example, in the case $\mu = 2$ we get

$$\begin{aligned} \lambda_2(\gamma_1, \mathcal{E}_{\max}) &= \frac{c}{n}\left[\mathcal{E}_{\max}^2/(\mathcal{E}_{\max}^2 + \gamma_1 A)\right] \quad (\gamma_1 > \tfrac{4}{5}), \\ &= \frac{c}{n}\left[16(1-\gamma_1)\,\mathcal{E}_{\max}^2/\ 16(1-\gamma_1)\,\mathcal{E}_{\max}^2 + (4-3\gamma_1)^2 A\right] (\gamma_1 < \tfrac{4}{5}), \end{aligned} \tag{5.1}$$

where $A = 2\omega c^2 \mu_0 N_T \hbar / n^2$ and γ_1 is the mole fraction of species 1. This dependence is shown in Fig. 5(a), wherein $\lambda_2 n/c$ is plotted as a function of γ_1 at fixed N_T and $\mathcal{E}_{\max}/A$. The concentration dependence of the doublet spacing Δ is also a characteristic quantity

$$\begin{aligned} \Delta(\gamma_1, \mathcal{E}_{\max}) &= 0 \quad (\gamma_1 > \tfrac{4}{5}) \\ &= \left[8\hbar n(c - n\lambda_2)/c^2 \mu_0 \mu_1 \omega N_T\right]^{1/2} \cosh^{-1}\left(\frac{8-9\gamma_1}{\gamma_1}\right) \quad (\gamma_1 < \tfrac{4}{5}), \end{aligned} \tag{5.2}$$

and is shown in Fig. 5(b). These types of measurements are direct tests of the theory.

A number of variations and extensions of this work are in progress. We are investigating solutions transient situations—i.e., what is the fate of a pulse of given shape imposed from the outside on the multispecies absorbing medium. Other critical issues being addressed are the effects of loss processes, of spectral densities of finite linewidth, and of allowing the phase to vary. Experimental verification of these phenomena is also under investigation. The phenomena shown here have possible applications in producing finite trains of extremely narrow pulses whose amplitude may easily be controlled by varying concentration ratios.

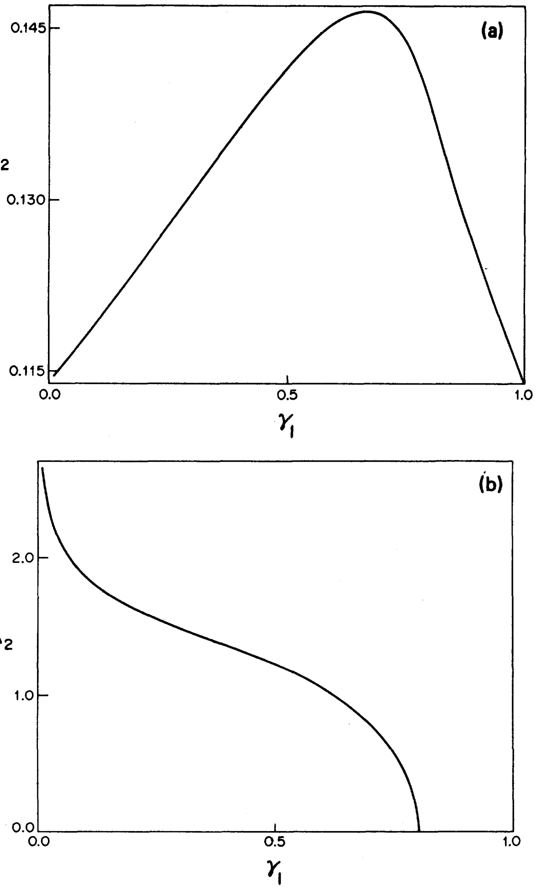

FIG. 5. Dependence of (a) velocity λ_2 and (b) peak separation Δ_2 for $\mu = 2$ doublet pulse on the number density of species 2, N_2, at constant N_1 and maximum pulse amplitude $\mathcal{E}_{\max}$.

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APPENDIX

Assumptions (1)–(5) of Sec. II imply for propagation in the z direction

$$\left(\frac{\partial}{\partial z} + \frac{n}{c}\frac{\partial}{\partial t}\right)\mathcal{E} = \frac{\omega c \mu_0}{2n} V, \tag{A1}$$

$$\mathcal{E}\left(\frac{\partial}{\partial z} + \frac{n}{c}\frac{\partial}{\partial t}\right)\phi = \frac{\omega c \mu_0}{2n} U, \tag{A2}$$

for the complex electric field envelope $\mathcal{E}e^{i\phi}$ and polarization envelope $U + iV$. Here ω, μ_0, c, and n are the light frequency, magnetic permeability, speed of light in free space, and index of refraction of the background medium. The polarization envelope functions U and V are related to the individual species polarization functions u_i, v_i ($i = 1, 2, \ldots, s$) via

$$\begin{Bmatrix} U \\ V \end{Bmatrix} = \sum_{i=1}^{s} \begin{Bmatrix} u_i \\ v_i \end{Bmatrix} , \tag{A3}$$

where u_i and v_i are related to the density matrix $\rho^{(i)}$ for species i; the latter satisfies

$$\begin{aligned} &\frac{\partial}{\partial t}[\rho_{11}^{(i)} - \rho_{22}^{(i)}] = 2i\mu^{(i)}[\rho_{21}^{(i)} - \rho_{12}^{(i)}]/\hbar , \\ &\frac{\partial \rho_{21}^{(i)}}{\partial t} = -i\omega\rho_{21}^{(i)} + i\mu^{(i)}E[\rho_{11}^{(i)} - \rho_{22}^{(i)}], \\ &\rho_{11}^{(i)} + \rho_{22}^{(i)} = 1, \quad \rho_{12}^{(i)} = \rho_{21}^{(i)*} , \end{aligned} \tag{A4}$$

where $E = \mathrm{Re}(\mathcal{E} e^{i\phi} e^{i\omega - ikz})$ is the electric field strength and $\mu^{(i)} = \langle 1 | \hat{\mu} | 2 \rangle^{(i)}$ is the matrix element of the dipole moment operator for the two states of species i in resonance with the electric field. With this we define $u_j + iv_j$ via

$$\rho_{21}^{(j)} = \frac{1}{2N_j\mu_j}(u + iv_j)e^{i(kz + \omega t + \phi)} , \tag{A5}$$

$$N_j\mu_j[\rho_{11}^{(j)} - \rho_{22}^{(j)}] = w_j , \tag{A6}$$

where N_j is the number density of species j. From the equation of motion (A4) we get

$$\frac{\partial u_j}{\partial t} = v_j \frac{\partial \phi}{\partial t} , \tag{A7}$$

$$\frac{\partial v_j}{\partial t} = -\mu_j \frac{\partial \phi}{\partial t} + \frac{\mu_j}{\hbar}\mathcal{E}\omega_j , \tag{A8}$$

$$\frac{\partial w_j}{\partial t} = \frac{\mathcal{E}}{\hbar}\mu_j v_j . \tag{A9}$$

With the above definitions (A1) and (A2) become

$$\left(\frac{\partial}{\partial z} + \frac{n}{c}\frac{\partial}{\partial t}\right)\mathcal{E} = -\frac{\omega c \mu_0}{2n}\sum_{i=1}^{s} v_i , \tag{A10}$$

$$\mathcal{E}\left(\frac{\partial}{\partial z} + \frac{n}{c}\frac{\partial}{\partial t}\right)\phi = \frac{\omega c \mu_0}{2n}\sum_{i=1}^{s} u_i . \tag{A11}$$

In the present study we shall seek solutions of constant velocity and constant phase, $\phi = 0$.

We find for the case of constant phase

$$\begin{aligned} &\frac{\partial u}{\partial t} j = 0 , \\ &\frac{\partial v_j}{\partial t} = \mu_j \mathcal{E} w_j/\hbar , \\ &\frac{\partial w_j}{\partial t} = -\mu_j \mathcal{E} v_j/\hbar , \end{aligned} \tag{A13}$$

$$\left(\frac{n}{c}\frac{\partial}{\partial t} + \frac{\partial}{\partial z}\right)\mathcal{E} = -\frac{\omega c \mu_0}{2n}\sum_{i=1}^{s} v_j .$$

Thus since u_j is initially zero it is always zero for the constant phase waves and will be deleted henceforth.

Next we seek constant velocity waves. Defining a time coordinate $\rho = t - z/\lambda$, where λ is the speed, we have

$$\begin{aligned} &\frac{\partial v_j}{\partial t} = \mu_j \mathcal{E} w_j/\hbar , \\ &\frac{\partial w_j}{\partial t} = -\mu_j \mathcal{E} v_j/\hbar , \\ &\frac{-1}{\lambda}\frac{\partial \mathcal{E}}{\partial \rho} + \frac{n}{c}\frac{\partial \mathcal{E}}{\partial t} = -\frac{\omega c \mu_0}{2n}\sum_{j=1}^{s} v_j , \end{aligned} \tag{A14}$$

where $w_j(\pm\infty) = N_j\mu_j$, $\mathcal{E} = v_j(\pm\infty) = 0$ for a pulse.

Defining dimensionless variables via

$$\begin{aligned} &\lambda' = \lambda n/c , \\ &v_j' = v_j/N_j\mu_j , \\ &w_j' = w_j/N_j\mu_j , \\ &t' = t\left(c^2\mu_0\mu_1^2\omega\sum_{i=1}^{s} N_i \Big/ 2\hbar n^2(1/\lambda' - 1)\right)^{1/2} , \\ &\mathcal{E}' = \mathcal{E}\left(2n^2(1/\lambda' - 1)\Big/\mu_0\sum_{i=1}^{s} N_i\right)^{1/2} , \\ &\gamma_j = N_j \Big/ \sum_{i=1}^{s} N_i , \\ &\mu_j' = \mu_j/\mu_1 , \end{aligned}$$

and noting that $\partial/\partial\rho = \partial/\partial t$ for a constant profile solution we obtain

$$\begin{aligned} &\frac{dv_j'}{dt'} = \mu_j'\mathcal{E}'w_j' , \\ &\frac{dw_j'}{dt'} = -\mu_j'\mathcal{E}'v_j' , \\ &\frac{d\mathcal{E}'}{dt'} = \sum_{i=1}^{s} \gamma_j\mu_j v_j' , \end{aligned}$$

where $w_j'(\pm\infty) = 1$, $v_j'(\pm\infty) = \mathcal{E}(\pm\infty) = 0$ for pulses. Finally, in the main text we drop the primes.

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