Index of Refraction for an Optical Medium with Clamped Quantum Phase

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In an adiabatic approximation for the simultaneous propagation of two optical pulses in a dielectric medium with clamped quantum phase, we reduce the Maxwell wave equation to a matrix equation with a non-Hermitian propagation kernel. Numerical simulations confirm novel consequences of an analytic solution that we identify as a two-photon-resonant propagation law. [S0031-9007(96)00102-0]

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The complex refractive index is the key parameter in the propagation of optical pulses in dielectric media. The real part determines optical path length and governs phase matching, and the imaginary part governs gain and loss in amplifiers and absorbers. The index is the parameter that specifies the most important length scales on which pulse propagation effects are measured, even when a pulse effect is not well described by an index, as, for example, in soliton and solitary wave propagation or in situations where the light intensity exceeds a nonlinear threshold or the pulse duration is too short. However, the refractive index is certainly the appropriate parameter to describe long and weak light pulses, even pulses of different colors traveling together through a dielectric so long as the different pulses excite the dielectric weakly and independently. For weak pulses, independence can normally be taken for granted because of rapid and ubiquitous incoherent material relaxation effects.

In this Letter we report on new results regarding the refractive index for weak multipulse propagation in a dielectric medium characterized by a quantum phase that is held fixed for long times despite the action of optical relaxation processes with very short lifetimes. Such an unusual medium (so-called phaseonium [1]) is unheard of in traditional spectroscopy, but is widely called for in recent proposals to use laser light for quantum control of reactions, devices, and processes of various kinds [2].

The consequences for pulse propagation in this nontraditional medium can be severe, leading, for example, to strong violations of Beer's law of absorption for the two pulses, even if both pulses are weak enough to be described by Beer's law separately. We show here that a two-photon-resonant propagation formula can be derived for phaseonium from approximate equations of propagation, and we confirm the validity of this formula by numerical simulations. We show that the formula predicts wave mixing that naturally incorporates earlier developments including matched pulses [3] and trapped and dressed fields [4] in an appropriate multifield vector space, and generalizes them to include inhomogeneous broadening processes.

In order to discuss our results most simply, we focus on propagation in a model for phaseonium that is an idealized version of a wide class of realistic absorbing media whose principal optical properties are determined by two-photon-resonant optical transitions from a pair of nondegenerate ground states to an upper level. As shown in Fig. 1, the transitions are affected by a variety of decay channels and are two-photon resonant, but may be far from individually resonant. A distribution of detunings Δ can be incorporated, and we have done this.

We inject two different-color plane-wave pulses into the phaseonium at $z = 0$ and calculate the changes in pulse amplitude and shape after propagation to position z. This can be done numerically case by case using well-known algorithms [5], and we give results below. However, this is not sufficient. For our main purpose, an analytic approach is necessary which we describe now.

The electric field vector for the two optical pulses can be written

$$\vec{E} = \hat{x}\mathcal{E}_a(z,t)e^{i(k_a z-\omega_a t)} + \hat{x}\mathcal{E}_b(z,t)e^{i(k_b z-\omega_b t)} + \text{c.c.}, \tag{1}$$

where $k_a c = \omega_a$ and $k_b c = \omega_b$; and $\mathcal{E}_a$ and $\mathcal{E}_b$ are the amplitudes of the electric fields of the two pulses. It is assumed that the two fields interact separately with

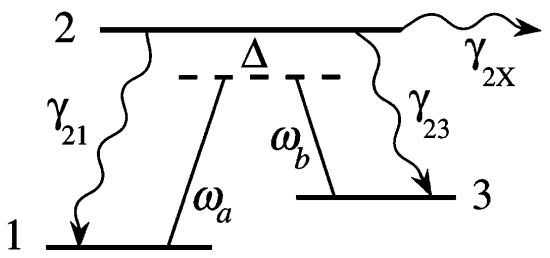

FIG. 1. Sketch of energy levels of typical three-level atom in absorbing medium.

the two transitions shown. Specifically, if we let $\delta\omega$ denote a generic bandwidth that roughly accounts for the incoherent phenomena that may be present, then we require $|\omega_a - \omega_b| \gg \delta\omega$, which is not difficult to satisfy when the fields are optical or near optical and in a Raman-type interaction as shown in Fig. 1.

The quantum phase of the two ground states is typically proposed to be clamped by external fields which do not interact with the phaseonium transitions of interest, and we do not concern ourselves with those fields here. If we account for background dispersion due to host and impurity atoms by interpreting c as the background propagation velocity, then in the narrow-band pulse limit the equations for the pulse amplitudes take the form

$$\frac{\partial\Omega_a}{\partial\zeta} = i\mu_a\langle\rho_{21}\rangle, \quad \frac{\partial\Omega_b}{\partial\zeta} = i\mu_b\langle\rho_{23}\rangle. \tag{2}$$

Here the notation is as follows: the Ω's are $\mathcal{E}$'s in frequency units, i.e., they are the corresponding complex Rabi frequencies: $\Omega_a \equiv 2d_a\mathcal{E}_a/\hbar$, and $\Omega_b \equiv 2d_b\mathcal{E}_b/\hbar$, while the ρ's are atomic density matrix elements appropriate to the two dipole transitions (1-2 and 3-2), and the μ parameters are standard propagation coefficients for the transitions individually: $\mu_a = 4\pi d_a^2 N\omega_a/\hbar c$, etc., where generally $\mu_a \neq \mu_b$. The angular brackets denote an average over the ensemble of source dipoles, i.e., an average over a distribution of Δ's arising from the Doppler effect or otherwise. Finally, we have defined local-time coordinates ζ and τ in the frame propagating with velocity c in the medium: $c\tau \equiv ct - z$ and $\zeta \equiv z$.

In addition to the two field equations there are, of course, equations for the nine atomic density matrix elements implied by the energy level diagram in Fig. 1, given by $i\hbar\partial\rho/\partial t = [H, \rho]$ + relaxation terms, as usual. These eleven equations are nonlinearly coupled and must be solved together, but for short medium response times, characteristic of the usual Beer's law (i.e., when $\gamma \gg \delta\omega$ and $\gamma\delta\omega \gg \Omega^2$, for all pulse bandwidths, Rabi frequencies, and damping rates), the atoms relax effectively instantly on the pulse time scale, and the relevant "adiabatic" solutions for $\langle\rho_{21}\rangle$ and $\langle\rho_{23}\rangle$ are

$$\langle\rho_{21}\rangle = \left\langle \frac{(\rho_{11} - \rho_{22})\Omega_a + \rho_{31}\Omega_b}{2(\Delta - i\gamma_a)} \right\rangle \tag{3}$$

and

$$\langle\rho_{23}\rangle = \left\langle \frac{\rho_{13}\Omega_a + (\rho_{33} - \rho_{22})\Omega_b}{2(\Delta - i\gamma_b)} \right\rangle. \tag{4}$$

Here γ is the total dipole damping rate of the individual transitions, i.e., $\gamma_a = \gamma_{21} + \gamma_{2X}$, etc., where γ_{2j} is the dephasing rate of the dipole transition between levels 2 and j.

Phaseonium assigns fixed relative quantum phase between states 1 and 3, and zero occupation probability to level 2, so we can take $\rho_{22} = 0$ and write the other density matrix elements in terms of fixed probability amplitudes, e.g., $\rho_{31} \equiv C_1^* C_3$, etc. In a conventional spectroscopic medium these off-diagonal coherences are zero or negligible, and one correctly expects that the differences in the present case must be significant.

Under present conditions, the right hand sides of both Eqs. (2) are proportional to a linear combination of Ω_a and Ω_b. Thus, if we designate by $|\Omega\rangle$ the two-field [6] column vector $[\Omega_a, \Omega_b]^T$, we can write the equations for the pulses in terms of a 2×2 propagation operator $\mathcal{K}$:

$$\mathcal{K} = \begin{bmatrix} K_a|C_1|^2 & K_a C_1^* C_3 \\ K_b C_1 C_3^* & K_b|C_3|^2 \end{bmatrix} = K\begin{bmatrix} rp & rq \\ sp & sq \end{bmatrix}.$$

In the second matrix we have introduced a simplified dimensionless notation for $\mathcal{K}$'s matrix elements: $p \equiv C_1$, $q \equiv C_3$, $r \equiv (K_a/K)C_1^*$, and $s \equiv (K_b/K)C_3^*$, with $K \equiv K_a|C_1|^2 + K_b|C_3|^2$, where $K_a \equiv \langle\mu_a/2(\Delta - i\gamma_a)\rangle$, etc. Note that we do not take $\mu_a = \mu_b$.

The propagation (scattering) equation equivalent to (2) is $\partial|\Omega\rangle/\partial\zeta = i\mathcal{K}|\Omega\rangle$, with the obvious solution

$$|\Omega(\zeta, \tau)\rangle = e^{i\mathcal{K}\zeta}|\Omega(0, \tau)\rangle. \tag{5}$$

This is a two-photon-resonant propagation law for phaseonium [7], and $\mathcal{K}$ is clearly a two-field (matrix) wave vector, but further analysis is required for interpretation. Note that the wave vector operator $\mathcal{K}$ is complex, nonsymmetric, and non-Hermitian [8]. It has complex eigenvalues κ_i, with $\kappa_0 = 0$ and $\kappa_1 = K$. The right and left eigenvectors of $\mathcal{K}$ are different and both will eventually be needed. For κ_0 we have $|u_0\rangle = [q, -p]^T$, and $\langle v_0| = [s, -r]$; and for κ_1 we have $|u_1\rangle = [r, s]^T$, and $\langle v_1| = [p, q]$. Note that $\langle v_0|u_1\rangle = \langle v_1|u_0\rangle = 0$ and $\langle v_0|u_0\rangle = \langle v_1|u_1\rangle = 1$, and the sum $\sum_i |u_i\rangle\langle v_i|$ is the unit operator.

Now we discuss the implications of the matrix propagation law (5) for phaseonium. Unusual features are found not previously associated with weak pulse propagation which illustrate the dramatic effects of perfect off-diagonal long-range order (ODLRO, or $\rho_{11}\rho_{33} \equiv \rho_{13}\rho_{31}$) in a new context.

Standard one-pulse results, such as Beer's law of absorption, cannot be obtained from (5) in the usual way by making the pulses arbitrarily weak or even by setting one of them initially to zero. However, if the off-diagonal coherences vanish ($\rho_{13} = \rho_{31} = 0$), then $\mathcal{K}$ is diagonal, the two components of $|\Omega\rangle$ evolve independently, and the two pulses recover their usual independence and Beer's law behavior, as shown in Fig. 2. For phaseonium, however, since one of the eigenvalues of $\mathcal{K}$ is identically zero (for arbitrary choices of quantum phase and γ), spontaneous wave mixing always gives rise to a nonattenuating part of the solution for both field components, independent of their initial values. The nonattenuating part of $|\Omega\rangle$ corresponds to the "trapped field" combination of amplitudes introduced earlier [4].

Given arbitrary inputs for the two fields Ω_a and Ω_b, the full solution for the a component is given by $\Omega_a(\zeta, \tau) = \langle a\,|\,\Omega(\zeta, \tau)\rangle$, where we will designate by $|a\rangle$ and $|b\rangle$

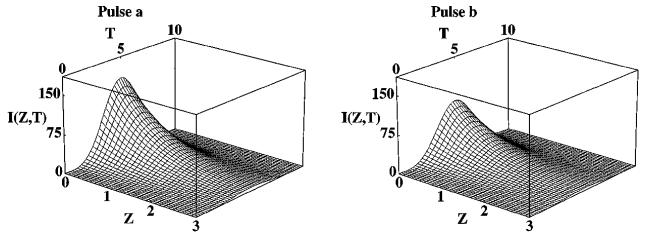

FIG. 2. Intensities $I(Z,T) \equiv (\Omega\tau_p)^2$ of pulses a (left) and b (right) as a function of space and time for a zero ODLRO medium. The parameters used are $\rho_{11} = 0.6$, $\rho_{33} = 0.4$, and $\rho_{13} = 0$, with $K_b = 1.5K_a$, $Z \equiv \mathrm{Im}(K)\zeta$, and $T \equiv \tau/\tau_p$, where τ_p is the width of pulse a.

the bare single-field states represented by $[1,0]^T$ and $[0,1]^T$, respectively. By exploiting both right and left eigenvectors of the propagation operator $\mathcal{K}$, we find

$$\begin{aligned}\Omega_a(\zeta,\tau) &= \sum_{i=0,1} \langle a|u_i\rangle e^{i\kappa_i\zeta}\langle v_i|\Omega(0,\tau)\rangle \\ &= (qs + rpe^{iK\zeta})\Omega_a(0,\tau) \\ &\quad - qr(1 - e^{iK\zeta})\Omega_b(0,\tau)\,. \qquad (6)\end{aligned}$$

A similar expression for Ω_b can be obtained. Note that (6) shows that if the τ dependences of the two pulses are the same at $\zeta = 0$ then they will be the same for all ζ. This is the "matched-pulse" effect noted by Harris [3] for non-phase-clamped media. In Figs. 3 and 4 we plot several predictions of formula (6). One sees the evolution toward matched shapes for $Z \gg 1$.

It is evident from formula (6) that the perfect offdiagonal long-range order plays a remarkable role in pulse propagation. In the top part of Fig. 3 we plot the evolution of the same input pulses of Fig. 2 in a perfect ODLRO medium. We observe that the input pulses oscillate in space (because of the nonzero imaginary part of the eigenvalue K) and decay to (because of the real part of the eigenvalue K) a spatially steady state. We should also mention that the phase of the off-diagonal order can affect the propagation significantly. This is illustrated in the bottom part of Fig. 3 where the degree of long-range order, the atomic level populations, and the input pulses are the same as for the top part, but the phaseonium phase is different by $\pi/4$. We note that because of the phase shift, the evolution is now completely different.

Formula (6) also implies that *deliberately unmatched pulses can be used for elaborate pulse shaping* since (5) specifies the degree to which the pulses will partially exchange some of their shape characteristics during propagation. This is shown in Fig. 4. In this case, pulse b was a pair of completely disjoint pulses injected before and after single pulse a. It is obvious that pulse shaping can be strongly dynamic both in time and in space.

In summary, we have obtained a new approximate propagation law for very weak light pulses in a quantum-phase-clamped medium (phaseonium). We have shown that the intrinsic two-photon-resonant nature of the propagation invalidates well-known aspects of weak pulse propagation. The key element in our analysis is an adiabatic approximation leading to a non-Hermitian matrix wave vector for the coupled fields. Despite heavily overdamped evolution of the atomic density matrix elements that couple to the fields, complex pulse shaping can be obtained in both space and time, and the reshaping is strongly quantum phase dependent. An unstated assumption has been the absence of decay from level 3 to level 1. In practice, this will add a small positive imaginary part to the zero eigenvalue of $\mathcal{K}$, leading to additional decay. However, both 1-2 and 2-3 are assumed to be dipole transitions, so $|1\rangle$ and $|3\rangle$ have equal parity and any 3-1 decay effects will normally be small. Finally, since the propagation law (5) was derived approximately, we have checked its validity by solving the coupled two-pulse Maxwell and Schrödinger equations fully numerically [5] in several cases, for comparison with the predictions of analytic formula (6). To illustrate the results, in Fig. 5 we show a comparison of the peak intensity of pulse a as computed by our analytic approximate

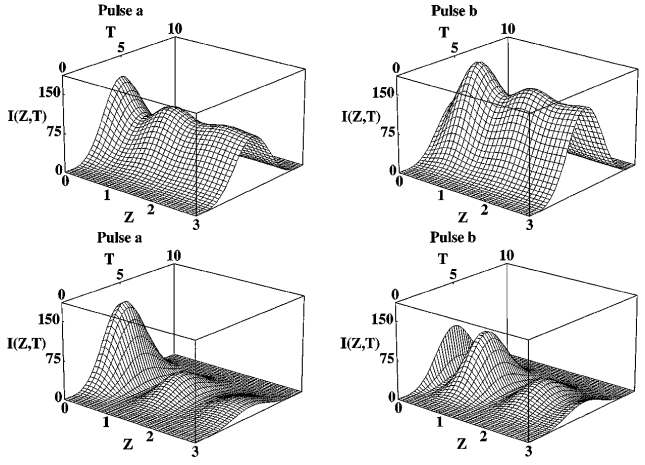

FIG. 3. Intensities $I(Z,T) \equiv (\Omega\tau_p)^2$ of pulses a (left) and b (right) as a function of space and time for a perfect ODLRO medium. The parameters are as in Fig. 2, except $\rho_{13} = -0.24$ in the top figures and $\rho_{13} = -\sqrt{0.24}\exp(-i\pi/4)$ in the bottom figures.

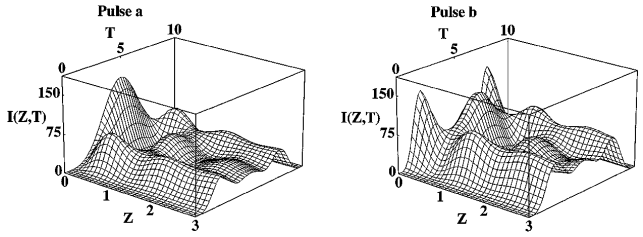

FIG. 4. Intensities $I(Z,T) \equiv (\Omega\tau_p)^2$ of pulses a (left) and b (right) as a function of space and time for a perfect ODLRO medium. The population parameters are as in Fig. 3 (top). Different input pulses are chosen, as shown.

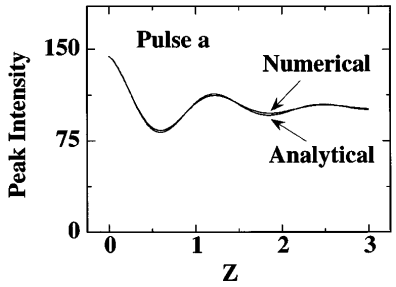

FIG. 5. Showing the agreement between the predictions of analytic propagation formula (6) and the exact numerical solution of the coupled Maxwell-Schrödinger equations.

theory and exactly numerically, as a function of propagation distance. The agreement is quite good.

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[1] Exploitation of the unusual properties of quantum phase-clamped atoms was initiated by M. O. Scully in Phys. Rev. Lett. **55**, 2802 (1985), and extended in Phys. Rev. Lett. **67**, 1855 (1991).

[2] Effects of recent interest include two-color interactions to control electron currents, molecular reactions, dielectric properties, various types of lasing, etc. Reviews of many of these effects, with wide reference lists, are given by M. O. Scully in Phys. Rep. **219**, 191 (1992), which treats phaseonium explicitly, and Quantum Opt. **6**, 201 (1994).

[3] S. E. Harris, Phys. Rev. Lett. **70**, 552 (1993); **72**, 52 (1994).

[4] J. H. Eberly, M. L. Pons, and H. R. Haq, Phys. Rev. Lett. **72**, 56 (1994).

[5] A good discussion of the numerical method for light propagation problem can be found in A. Icsevgi and W. E. Lamb, Jr., Phys. Rev. **185**, 517 (1969), and in F. P. Mattar, Appl. Phys. **17**, 53 (1978). We have used the general outline presented in these papers and actually used the fourth-order Runge-Kutta method for the time integration and the Euler method for the space integration as described in W. H. Press *et al.*, *Numerical Recipes in Fortran* (Cambridge University Press, Cambridge, England, 1992), p. 704.

[6] Clearly the two-field case can be generalized.

[7] In contrast to some earlier treatments [2], we take account of the mutual interaction of the two propagating fields, which in the presence of trapping states must be treated as a coherent joint excitation even when their bandwidths have no overlap at all. This is the physical motivation for a matrix formulation of the propagation equations. When the incident field is treated as a single pulse (see [1]), these considerations are not evident.

[8] Previous discussion of non-Hermitian propagation kernels has been motivated by consideration of excess spontaneous emission in laser amplifiers and oscillators. An early example is A. E. Siegman, Phys. Rev. A **39**, 1253 (1989); **39**, 1264 (1989).