Recall and Creation of Spatial Excitation Distributions in Dielectric Media

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We present an analytical theory for the optical properties of a novel three-level medium for which the degree of its initial excitation is a function of the position. The spatial distribution of the excitation can be imaged on the temporal profile of an electromagnetic signal that is created inside the medium when traversed by a cw-laser field. We propose an experimental scheme to generate arbitrary spatial excitations. [S0031-9007(97)04439-6]

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In recent years there has been a growing interest in exploring the unusual optical propagation properties of dielectric media that are coupled coherently to one or several laser fields. The dielectric material is typically initially in its lowest energetically possible state. However, recently, the optical transmission properties of an unusual medium—so-called phaseonium—have been investigated [1–4]. Phaseonium is a dielectric medium that has been prepared in a coherent superposition of two energy eigenstates. This nontraditional medium is the key element in recent proposals to use laser light for coherent quantum control of molecular reactions and various other processes [5]. The mechanism for electromagnetically induced transparency, pioneered by Harris and co-workers [6], is also related to coherent excitations that are maintained by external fields. Various types of solitary wave forms have been found for phaseonium [7], and it has been demonstrated [8] that this medium can allow for the formation of shape-invariant pulse pairs called adiabatons from arbitrary input fields.

In this Letter we report the first results for an optical medium in which the quantum mechanical state of the medium depends on the position. The degree of initial excitation of the medium becomes a new variable depending on space. The information contained in the spatial modulation of the excitation can be either an analog signal or a digital sequence of binaries. In the following, we will present a fully analytical theory for the optical properties of such a medium. If the space dependent excitation (SDE) is partially coherent, a laser field can be injected into the medium to "recall" this excitation. This recall field triggers the formation of a second laser pulse (called the signal field) with a different carrier frequency whose *temporal* envelope is an inverted mirror symmetric replica of the medium's original *spatial* excitation distribution. Below we first discuss the physical mechanism for the information recall from an SDE medium and then we show how the information contained in an amplitude-modulated optical beam can be converted into the space-dependent excitation of a medium that is initially in the ground state.

The two lowest lying states $|1\rangle$ and $|3\rangle$ of the medium are assumed to be dipole coupled to the same upper level $|2\rangle$. The SDE medium is in its ground state $|1\rangle$ with the exception of a spatial regime in which some of the population is also in the metastable state $|3\rangle$, $\rho_{11}(z, t = 0) = 1 - |G(z)|^2$ and $\rho_{33}(z, t = 0) = |G(z)|^2$. The excitation distribution $G(z)$ is an arbitrary function of the position z with $|G(z)|^2 \leq 1$. The initial off-diagonal coherence $\rho_{31}(z, t = 0)$ is related here to the diagonal density matrix elements via $|\rho_{13}| = \lambda\sqrt{[\rho_{11}\rho_{33}]}$ where $0 < |\lambda| < 1$. The complex parameter λ is identical to zero for thermally populated levels and $|\lambda| = 1$ for perfect atomic coherence.

The electromagnetic wave can be expressed in terms of its complex amplitude, where we assume that the recall field E_R as well as the signal field E_S are practically monochromatic:

$$E(z,t) = \hat{x}E_R(z,t)\exp[i\omega_R(t - z/c)] + \hat{y}E_S(z,t)\exp[i\omega_S(t - z/c)] + \text{c.c.}, \quad (1)$$

with the two resonant optical frequencies ω_R (transition $2 \leftrightarrow 3$) and ω_S (transition $1 \leftrightarrow 2$). We use the slowly varying envelope approximation such that the temporal and spatial evolution of each amplitude is governed by the well-known reduced wave equation. For $E_S(z, t)$ we have

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)E_S(z,t) = i\frac{2\pi\omega_S}{c}P_S(z,t), \quad (2)$$

and a similar equation for the recall field $E_R(z, t)$. The polarization depends on the medium's microscopic dipole moment $P(z, t) = \mathcal{N}\,\text{tr}[\rho(z, t)\hat{d}]$, where $\mathcal{N}$ denotes the density of atoms and $\hat{d}$ the slowly varying part of the corresponding atomic dipole operator. The notation is simplified if we replace the electric field amplitudes E_S and E_R by Rabi frequencies via $S \equiv 2d_S E_S/\hbar$ and $R \equiv 2d_R E_R/\hbar$ where d is the dipole moment between the relevant levels. The propagation constants are related to the density of atoms via $\mu_S \equiv 4\pi\mathcal{N}d_S^2\omega_S/\hbar c$ and similarly for μ_R. To obtain fully analytical solutions, we assume equal oscillator strength $\mu_S = \mu_R$ for both transitions.

To present our theory first for the simplest possible case, we begin with a SDE medium with a fully established atomic coherence ($\lambda = 1$) such that each atom can be described by state amplitudes $C_j(z, t)$ where $j = 1, 2, 3$. We assume that the response of the medium to

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the field is temporally adiabatic leading to the trapped-state condition $S(z,t) = -C_3(z,t)\sqrt{[R(z,t)^2 + S(z,t)^2]}$. If we assume that the input field R is slowly varying, this approximation is quite reasonable if the excitation function $G(z)$ does not change very rapidly:

$$\frac{\partial}{\partial z} G(z)\sqrt{[1 - |G(z)|^2]} \ll \frac{2\mu}{R} + \frac{R}{c}. \qquad (3)$$

Even though this inequality contains a *space* derivative, it follows directly from the *temporally* adiabatic atomic dynamics which guarantee that the evolution follows closely the instantaneous eigenvectors of the system.

The trapped-state condition applied to Eq. (2) together with a correction due to nonadiabaticity leads to a nonlinear wave equation [8] for the amplitude of state $|3\rangle$:

$$\left[\frac{\partial}{\partial z} + \left(\frac{1}{c} + \frac{2\mu}{R(z,t)^2 + S(z,t)^2}\right)\frac{\partial}{\partial t}\right] C_3(z,t) = 0, \qquad (4)$$

which needs to be solved under the initial condition of the medium $C_3(z, t = 0) \equiv G(z)$ and the boundary condition given by the two fields: $R(z = 0, t) \equiv R(t)$ and $S(z = 0, t) \equiv 0$. The denominator in Eq. (4) is proportional to the sum of the intensities and satisfies a simple linear wave equation such that its evolution is simply given by $R(z,t)^2 + S(z,t)^2 = R(t - z/c)^2$ [8,9]. The general solution for the signal field $S(z,t)$ can be found analytically and requires the inversion of an integral [10]. If we assume that the recall field is zero for $t < 0$ the solution is

$$S(z,t) = -R(t - z/c)G\left[z - \frac{1}{2\mu}\int_0^{t-z/c} dt'\, R(t')^2\right]. \qquad (5)$$

This expression shows that an electromagnetic signal $S(z,t)$ is created inside the medium as soon as the front edge of the recall field reaches that spatial domain in which $G(z)$ is nonzero. The maximum signal amplitude can approach that of the recall field if there are regions in the medium in which the population is entirely in the metastable state ($|G| = 1$). We discuss here the case which is of most practical importance. It corresponds to a recall field that has a constant amplitude after its turn-on. If the total length of the medium is larger than the domain in which $G(z)$ is nonzero, the created signal does not get attenuated or distorted and travels with invariant shape. The propagation velocity of the signal is $V_S = c(1 + 2\mu c R^{-2})^{-1}$ and can be much smaller than c, which indicates that the signal and the recall field interact strongly with the medium. As soon as the SDE information has been recalled, the medium passed by the signal is left behind in the ground state. As an interesting redundancy one should note that the recall field at output obtains some of the SDE information, but in inverted form. The spatial length of the emitted signal is identical to that of $G(z)$, which we denote by L. The temporal width T_S decreases with increasing intensity of the recall field, $T_S = L(1 + 2\mu c R^{-2})/c$.

To demonstrate the recall process graphically, we show in Fig. 1 temporal snapshots of the signal taken at various distances. In this example, the excitation function G was chosen to be a doubly peaked Gaussian (Fig. 2). The scaled propagation distance $Z \equiv z/L$ is in units of L such that $G \neq 0$ if $Z \in [0, 1]$. It follows that $Z = 1$ is the minimum propagation distance required to read off the excitation function $G(Z)$. When the wave front of the recall field reaches the regime in which $G(z)$ is nonzero, the signal is created. For distances $Z > 1$, the recall is completed and the (two-peak) signal pulse propagates shape invariant through the medium with reduced speed and without significant attenuation.

In Fig. 2 we show the "energy" of the signal $e_S(Z) \equiv \int dt' \, |S(Z,t')|^2$ as a function of the propagation distance. The energy growth rate depends on G, shown by the broken line. For larger distances $Z > 1$ the energy remains practically constant, indicating the shape-invariant propagation of the signal. In the figure we also show the energy of the recall field $e_R(Z) \equiv \int dt' |R(Z,t')|^2$. The decrease at $Z \in [0.2, 0.8]$ shows that some of the energy of the recall field has been converted to create the signal. The total electromagnetic energy $e_S + e_R$ is conserved, which demonstrates that the stored energy of the medium due to population in level $|3\rangle$ is not used for the signal. This physical mechanism is obviously different from the operation process of a standard light amplification in which the gain is paid for by atomic excitation energy.

We now turn to an interesting superposition law for nonlinear propagation. Even though the interaction between the medium and the fields is highly nonperturbative as the excitation of the medium evolves in space and time, the trapped-state relation between $C_3(z,t)$ and $S(z,t)$ indicates that the "operator" that relates the input data $[R(t), G(z)]$ to the output field $S(z,t)$ is linear with respect to $G(z)$. To illustrate this "linear" relation between the spatial distribution $G(z)$ and the temporal profile of the generated signal field, we have prepared the medium's degree of excitation $G(z)$ as a three-peaked function of space as shown in

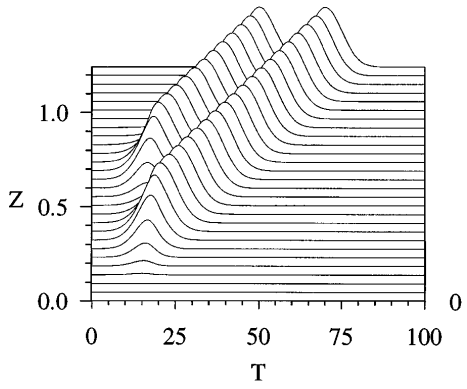

FIG. 1. The generation and propagation of the signal field $S(Z,T)$ inside the medium. The retarded time $T \equiv (t - z/c)R/10$ is measured in units of R^{-1}, where R denotes the Rabi frequency of the recall field. The created doubly peaked signal travels shape invariant with reduced speed for $Z > 1$. The initial excitation function $G(Z)$ is shown in Fig. 2. [The parameters are $R = 10$, $\mu = 1$, $L = 3000$, and $\lambda = -1$.]

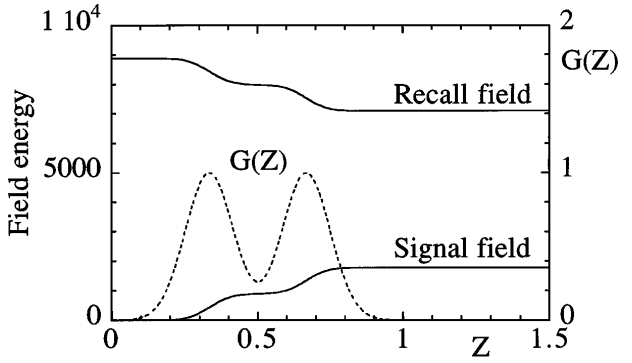

FIG. 2. The conversion of the electromagnetic energy of the recall field $e_R(Z) \equiv \int dt' |R(Z,t')|^2$ into that of the signal $e_S(Z)$. The broken line shows the excitation function $G(Z)$. Note that $e_R + e_S$ is conserved. [Same parameters as in Fig. 1.]

Fig. 3(a). In Fig. 3(b) we show the temporal profile of the resulting signal. As expected, this profile is inverted and mirror symmetric to the spatial distribution of $G(z)$ and every detail contained in the spatial excitation information has been transferred to the signal field. We present the real part of the amplitude in the figure to show that even the phase of $G(z)$ is mapped on the signal.

To check our approximate analytical formula we have also solved the Maxwell and Liouville equations numerically using a modified Runge-Kutta-Euler algorithm to solve the corresponding eleven coupled partial differential equations in space and time. We have included spontaneous emission from level 2, collisional broadening between each of the lower states and the upper state, as well as an irreversible decay from the upper level. The exact numerical solution is also displayed in Fig. 3(b) and the two curves are practically indistinguishable. This excellent agreement is typical for all of our calculations as long as the relaxation rates for dissipative processes mentioned above are smaller than the Rabi frequency R of the recall field. Even if the relaxation rates are comparable to R, the amplitude of the signal field was compressed by only 50%.

Finally, we want to show that the information transfer efficiency is reduced if the off-diagonal coherence between levels $|1\rangle$ and $|3\rangle$ is imperfect [$|\lambda| \neq 1$]. We have repeated our exact numerical simulation of the coupled Maxwell-Liouville equations for the same excitation function $G(z)$ shown in Fig. 3(a) but for various degrees of coherence λ. The growth pattern of the signal energy is illustrated in Fig. 4. As expected the transfer is reduced. On the other hand, the curve for $\lambda = 0.25$ shows that a medium with only 25% coherence is still suitable to generate a significant signal, merely with a smaller amplitude. The practically zero slope of the energy for sufficiently large distances indicates that perfect coherence is not necessary for the loss-free propagation. The signal pulse width (not shown) decreases with decreasing λ, but the basic temporal (three-peak) structure is qualitatively maintained even for λ as low as 0.25, which suggests that the recall effect is relatively robust.

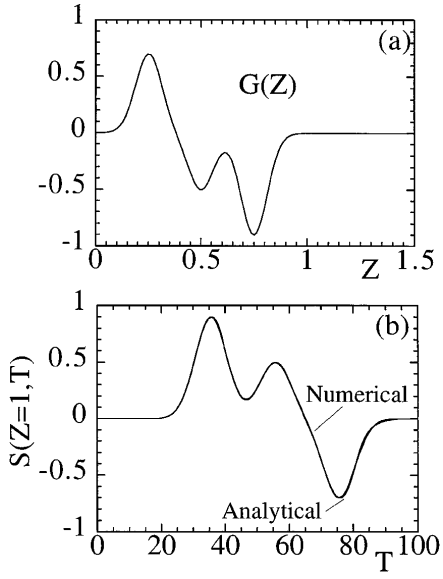

FIG. 3. Superposition law for nonlinear pulse propagation. Relation between the spatial profile of excitation $G(Z)$ and the temporal profile of the generated signal field $S(Z,T)$. (a) The medium's initial excitation distribution $G(Z)$ with perfect coherence ($\lambda = 1$). (b) The temporal profile of the signal field (in units of R) after it has been created by the medium. The second curve is the prediction according to the exact numerical solution of the coupled Maxwell-Liouville equations. The atomic relaxation times due to the dissipative processes mentioned in the text were chosen to be equal to the pulse duration of the generated signal pulse. The two curves are practically indistinguishable. [$R = 10$, $\mu = 1$, $L = 4000$.]

The curve labeled μ corresponds to the case of unequal propagation coefficients μ. Even though the energy growth pattern of the signal is similar to that for the case of equal couplings, no form-invariant propagation is found for $Z > 1$. The shape of the signal amplitude (not shown) displays fast oscillations that are a typical signature of nonadiabaticity in pulse propagation.

Let us now demonstrate the generation of a position dependent excitation inside a medium that is initially entirely in the ground state. We will show below that

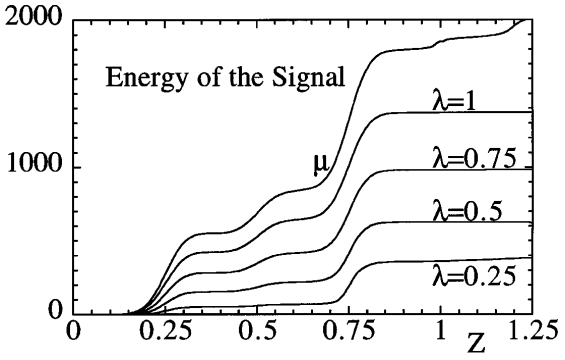

FIG. 4. The energy of the signal field for the same excitation function $G(Z)$ as shown in Fig. 3 but for various degrees of atomic coherence λ. All the parameters are as in Fig. 3. The graph denoted with μ is for perfect coherence ($\lambda = 1$) but unequal propagation constants ($\mu_S = 1.3$ and $\mu_R = 1$).

this can be accomplished using two amplitude-modulated optical pulses of different carrier frequency. Consider that the ground-state medium is subjected to two resonant laser pulses, Ω_1 pumping the 1-2 transition and Ω_3 coupling the levels 3 and 2. If the two fields are turned on in the counterintuitive sequence (Ω_3 enters the medium before Ω_1), then the trapped-state condition indicates that the final state of the medium depends only on the details of how the pulses are turned off [11]:

$$C_3(z,\infty) = -\lim_{\tau\to\infty} \frac{\Omega_1(z,\tau)}{\sqrt{\Omega_1^2(z,\tau) + \Omega_3^2(z,\tau)}}. \qquad (6)$$

The final excitation at the entry surface of the medium ($z = 0$) is determined by the input pulse shapes of the fields. Inside the medium ($z > 0$), however, the dynamics are more complicated as both pulses can change their shapes and the trailing edge of the fields can propagate with different velocities.

Using the theory of adiabatons [8,9], it is possible to find analytical expressions for the final excitation distribution inside the medium. This theory provides solutions for the space- and time-dependent amplitudes of two resonant laser pulses that are injected in the counterintuitive time sequence into a three-level medium initially in its ground state. Applied to our situation it predicts the final medium state as a function of the input fields:

$$C_3(z,\infty) = -\frac{\Omega_1(z=0, t=\chi(z))}{\sqrt{\Omega_1(z=0, t=\chi(z))^2 + \Omega_3(z=0, t=\chi(z))^2}} \qquad (7)$$

where the function $\chi(z) \equiv F^{-1}[F((\infty) - z)]$ and $F(x) \equiv (2\mu)^{-1}\int_0^x dt[\Omega_1(z=0,t)^2 + \Omega_3(z=0,t)^2]$ and $F^{-1}[y]$ is its inverse function. If the medium is subjected to two input pulses with specified pulse shapes and temporal delay between the pulses, any arbitrary excitation function $G(z) = C_3(z,\infty)$ can be generated inside the medium. In general, the SDE depends on the pulse shapes of both fields. It is helpful to point out a special case which has some similarity with the recall process when viewed in reversed time. In this situation the input pulse forms are complementary such that the sum of both intensities is constant ($\equiv\Omega$) at $z = 0$ for $0 < t < T_m$ and 0 otherwise. The required integral inversion can be done analytically and we obtain the simple solution $C_3(z,\infty) = -\Omega_1(z=0, t = T_m - 2\mu z/\Omega)/\Omega$. In this case the final medium excitation is precisely a mirror symmetric replica of the temporal pulse shape of only one of the input fields.

The applicability of the theory is not restricted to any specific laser pulse shapes or specific functional forms for $G(z)$. The recall mechanism in a material with a controlled SDE could be exploited to tailor pulses with any desired shape. The index of refraction depends directly on $|G(z)|^2$. As an example, a sequence of several pulses Ω_1 at input would generate a spatially modulated periodic index.

Our theoretical predictions can be tested experimentally in the optical regime. Harris' group [12] has provided the first experimental evidence for adiabatons [8]. A similar optical scheme could be used to first generate a SDE medium with two pulses and then to recall the excitation distribution with a single field. A sequence of experiments with variable timing of the recall field delay would show the lifetime of the SDE coherence. Hakuta *et al.* [13] have recently demonstrated experimentally that solid hydrogen can provide an ideal material to study coherences.

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