Coherent Control of the Effective Susceptibility through Wave Mixing in a Duplicated Two-Level System

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(Received 15 May 2008; published 20 November 2008)

We theoretically demonstrate the coherent control of the effective susceptibility of a duplicated two-level system. The control is obtained for a linearly polarized weak field in the presence of a much stronger orthogonally polarized field. For small optical depths, the effective susceptibility $\chi_{\rm eff}$ behaves as $\chi_{\rm lin}e^{2i\phi}$ ($\chi_{\rm lin}$ is the linear susceptibility, ϕ the phase shift) allowing coherent control of the optical response. For large optical depths, $\chi_{\rm eff} \approx \chi_{\rm lin}^*$, turning an absorber into an amplifier without affecting the dispersion.

DOI: 10.1103/PhysRevLett.101.213601 PACS numbers: 42.50.Gy, 42.65.−k

The problem of tailoring the optical response of a medium is one of the most active fields in optics because of its enormous potentiality of applications. Negative refractive index physics [1], slow, stored, and fast light [2], are inexhaustive examples of research fields where the modification of the linear response leads to a drastic modification of the electromagnetic fields. Of particular interest is the use of quantum interference to control the optical properties of a material. Electromagnetically induced transparency (EIT) [3], refractive index enhancement via quantum coherence [4], and lasing without inversion [5] are some spectacular examples of this technique. It has also been used by McCullough *et al.* who proposed bichromatic excitation of an atomic system prepared initially in a coherent superposition of states to control the optical response, but the control still depended on the field characteristics [6]. Schmidt *et al.* proposed a nonlinear scheme in a four-level system that cancels absorption (through EIT) and yields a giant Kerr nonlinearity [7]. It was later experimentally observed [8] and a lot of work that takes advantage of this effect was carried out in various contexts [9].

In this Letter, we present the coherent control of the nonlinear response of an atomic medium independent of the control field characteristics. The medium is an assembly of duplicated two-level atoms excited by a pair of two identical fields having orthogonal polarizations (Fig. 1) with one field (pump) much stronger than the other (probe). We identify two regimes where the medium exhibits an effective susceptibility ($\chi_{\rm eff}$) for the probe that is independent of the pump intensity and is as large as the linear susceptibility (giant response). For small optical depths, we obtain $\chi_{\rm eff} \approx \chi_{\rm lin}e^{2i\phi}$ where $\chi_{\rm lin}$ represents the linear susceptibility and ϕ is the phase shift between the two fields. Coherent control of both the refraction index and absorption coefficient is thus possible through the gain-dispersion coupling that appears. This behavior is explained by the interference between different excitation paths that are available to the probe. Moreover, the absorption by the population (linear response) is compensated by a cross-Kerr-type effect, and a different quantum path (associated with conjugate phase) accounts for the phase dependent medium response. In the method described here, no trapping (dark) state exists in the medium, in contrast with the giant Kerr effect [8,9] based on EIT and for which no phase control can be obtained. When the optical depth increases, this result is no longer valid, and we obtain $\chi_{\rm eff} \approx \chi_{\rm lin}^*$. Anomalous dispersion is now associated with an absorption hole in contrast with the usual linear response.

The duplicated two-level system excited by a pair of orthogonally polarized fields has been investigated before in both the femtosecond regime [10], where the coherent control of the medium gain for the probe was demonstrated, and in the long pulse regime where transparency leading to slow light was shown to occur thanks to coherent Zeeman oscillations [11].

We consider the $F = 1/2 \rightarrow F = 1/2$ transition (for instance, $^2S_{1/2}F = 1/2 \rightarrow {}^2P_{1/2}F = 1/2$ transition of ^{6}Li at 671 nm) excited by two copropagating, linearly polarized fields having the *same frequency* ω and mutually orthogo-

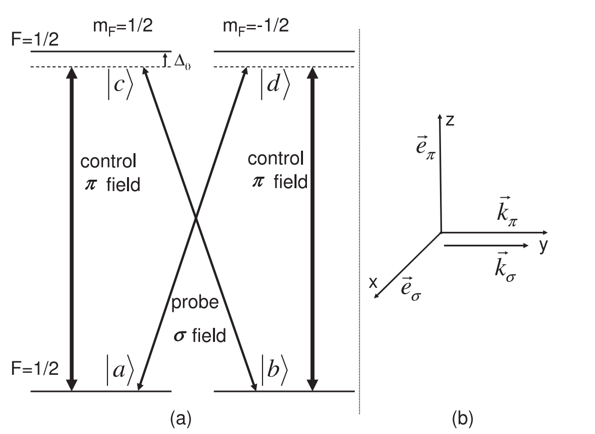

FIG. 1. (a) The duplicated two-level system and (b) fields configurations.

nal polarizations (Fig. 1). The electric fields are $\vec{E}_\pi(y,t) = \vec{e}_z\epsilon_\pi(y)e^{-i(\omega t-ky)} + \text{c.c.}$ and $\vec{E}_\sigma(y,t) = \vec{e}_x\epsilon_\sigma(y) \times e^{-i(\omega t-ky)}e^{-i\phi} + \text{c.c.}$ with corresponding Rabi frequencies $\Omega_\pi = D\epsilon_\pi/\hbar$ and $\Omega_\sigma = D\epsilon_\sigma/\hbar$. D is the dipole matrix element given by $D = \langle a|\hat{\vec{D}} \cdot \vec{e}_z|c\rangle$, $\Delta_0 = \omega_0 - \omega$ is the detuning, ϕ is the phase shift and $\epsilon_\pi(y=0)$ and $\epsilon_\sigma(y=0)$ are real. For such excitation, the system is equivalent to a double two-level system with the π polarized control field $\vec{E}_\pi$ coupling the transitions with identical m_F and the σ polarized probe field $\vec{E}_\sigma$ coupling the levels with different m_F. We determine in the following the susceptibility of the medium for the weak probe in the presence of much stronger control field. The time evolution of the system is given by the following density matrix equations:

$$
\begin{aligned}
i\dot{\rho}_{aa} &= [(\Omega_\pi\rho_{ac} + \Omega_\sigma\rho_{ad}e^{-i\phi}) - cc] + i\Gamma(\rho_{cc} + 2\rho_{dd})/3\\
i\dot{\rho}_{bb} &= [(-\Omega_\pi\rho_{bd} + \Omega_\sigma\rho_{bc}e^{-i\phi}) - cc] + i\Gamma(\rho_{dd} + 2\rho_{cc})/3\\
i\dot{\rho}_{cc} &= [(-\Omega_\pi\rho_{ac} - \Omega_\sigma\rho_{bc}e^{-i\phi}) - cc] - i\Gamma\rho_{cc}\\
i\dot{\rho}_{dd} &= [(\Omega_\pi\rho_{bd} - \Omega_\sigma\rho_{ad}e^{-i\phi}) - cc] - i\Gamma\rho_{dd}\\
i\dot{\rho}_{ca} &= \Omega_\pi(\rho_{cc} - \rho_{aa}) + \Omega_\sigma e^{-i\phi}(\rho_{cd} - \rho_{ba})\\
&\quad + (\Delta_0 - i\Gamma_d)\rho_{ca}\\
i\dot{\rho}_{db} &= -\Omega_\pi(\rho_{dd} - \rho_{bb}) + \Omega_\sigma e^{-i\phi}(\rho_{dc} - \rho_{ab})\\
&\quad + (\Delta_0 - i\Gamma_d)\rho_{db}\\
i\dot{\rho}_{ba} &= \Omega_\pi^*\rho_{da} + \Omega_\pi\rho_{bc} - \Omega_\sigma^* e^{i\phi}\rho_{ca} + \Omega_\sigma e^{-i\phi}\rho_{bd}\\
i\dot{\rho}_{dc} &= \Omega_\pi^*\rho_{da} + \Omega_\pi\rho_{bc} + \Omega_\sigma^* e^{i\phi}\rho_{db} - \Omega_\sigma e^{-i\phi}\rho_{ac}\\
&\quad - i\Gamma_{ze}\rho_{dc}\\
i\dot{\rho}_{da} &= \Omega_\pi(\rho_{ba} + \rho_{dc}) + \Omega_\sigma e^{-i\phi}(\rho_{dd} - \rho_{aa})\\
&\quad + (\Delta_0 - i\Gamma_d)\rho_{da}\\
i\dot{\rho}_{cb} &= -\Omega_\pi(\rho_{ab} + \rho_{cd}) + \Omega_\sigma e^{-i\phi}(\rho_{cc} - \rho_{bb})\\
&\quad + (\Delta_0 - i\Gamma_d)\rho_{cb}.
\end{aligned} \tag{1}
$$

cc denotes complex conjugate, and Γ is population damping rate. In the absence of nonradiative homogeneous dephasing processes, relaxation rates (Γ_{ze}, Γ_d) reduce to (Γ, $\Gamma/2$). The modification of the probe field is determined by the behavior of the coherence $\rho_\sigma = \rho_{cb} + \rho_{da}$ that radiates the σ polarized light ($\rho_\pi = \rho_{ca} - \rho_{db}$ is responsible for π polarized radiated field). The evolution of ρ_σ is given by the equation

$$
i\dot{\rho}_\sigma = \Omega_\pi(\rho_Z^* - \rho_Z) + \Omega_\sigma e^{-i\phi}(n_e - n_g) + (\Delta_0 - i\Gamma_d)\rho_\sigma. \tag{2}
$$

Here, $\rho_Z = \rho_{ab} + \rho_{cd}$; $n_g = \rho_{aa} + \rho_{bb}$; $n_e = 1 - n_g$. The coherence ρ_σ builds up through two competing phenomena: The diffraction of the pump by the Zeeman coherences (first term) and the absorption of the probe by the population (second term). It can be shown that due to the symmetry of the system $\rho_{dc} = 0$ in the stationary regime. Only the ground Zeeman coherence ρ_{ab} contributes to ρ_σ. We can work out the following relations in the stationary regime $\rho_\sigma = 2\rho_{da} = 2\rho_{cb} = (\Omega_\sigma e^{-i\phi}/\Omega_\pi)^*\rho_\pi$ with

$$
\rho_\sigma = \frac{(\Omega_\pi^2 + \Omega_\sigma^2 e^{-2i\phi})(i\Gamma_d + \Delta_0)\Omega_\sigma^* e^{i\phi}}{4\Gamma_d\Gamma^{-1}|\Omega_\pi^2 + \Omega_\sigma^2 e^{-2i\phi}|^2 + (|\Omega_\pi|^2 + |\Omega_\sigma|^2)(\Gamma_d^2 + \Delta_0^2)}. \tag{3}
$$

For the matching condition $\Omega_\pi^2 + \Omega_\sigma^2 e^{-2i\phi} = 0$, we have $\rho_\sigma = \rho_\pi = 0$. The system is transparent for both π and σ polarized fields. This is the well-known case where a dark state is realized in the system and all population is trapped in that dark state [12]. Indeed, the total polarization $\vec{e}_{\text{tot}} = \vec{e}_z \pm i\vec{e}_x$ is circular, and all the population is, respectively, trapped in the eigenstates of F_y with $m_y = \pm 1/2$. Another limiting case is $|\Omega_\pi| \ll |\Omega_\sigma|$. Here, ρ_σ can be approximated to $\frac{(i\Gamma_d + \Delta_0)\Omega_\sigma e^{-i\phi}}{\Gamma_d^2 + \Delta_0^2 + 4\Gamma_d\Gamma^{-1}|\Omega_\sigma|^2}$. The influence of the control is negligible, and only the probe can saturate the medium. The situation that interests us is that of strong control and weak probe $|\Omega_\pi| \gg |\Omega_\sigma|$. ρ_σ in this case simplifies to

$$
\rho_\sigma \approx \left(\frac{\Omega_\pi^2}{|\Omega_\pi|^2}\right)\frac{(i\Gamma_d + \Delta_0)\Omega_\sigma^* e^{i\phi}}{4\Gamma_d\Gamma^{-1}|\Omega_\pi|^2 + \Gamma_d^2 + \Delta_0^2} \tag{4}
$$

Both coherences ρ_σ and ρ_π vanish as long as $|\Omega_\pi| \gg \sqrt{\Gamma_d\Gamma}$: the saturation of the medium by the pump renders the medium transparent for both σ and π polarized fields. If $|\Omega_\pi| \ll \sqrt{\Gamma_d\Gamma}$ and if we neglect the phase grow during propagation (this condition will be discussed latter), Ω_π and Ω_σ are still real, and we get $\rho_\sigma \approx \Omega_\sigma e^{i\phi}/(\Delta_0 - i\Gamma_d)$. The effective susceptibility $\chi_{\text{eff}} = (2\alpha_0\Gamma_d/k)\rho_\sigma e^{i\phi}/\Omega_\sigma$ becomes

$$
\chi_{\text{eff}} = \chi_{\text{lin}} e^{2i\phi}. \tag{5}
$$

$\chi_{\text{lin}} = (2\alpha_0\Gamma_d/k)/(-i\Gamma_d + \Delta_0)$ is the linear susceptibility of the system and $\alpha_0 = ND^2\omega_0/2c\hbar\epsilon_0\Gamma_d$ is the field absorption coefficient at resonance (N is the atomic density). This is the central result of this Letter. The medium behaves as a linear medium with a modified susceptibility χ_{eff} that is connected through a simple expression to the true linear susceptibility. An important feature is that the response of the medium no longer depends on the pump intensity. The phase dependence of the effective susceptibility leads to the coupling between dispersion and gain as displayed in Fig. 2, allowing a real phase control of the linear response of the medium. As is shown in the Figure, the medium turns from an absorber at $\phi = 0$ to an amplifier at $\phi = \pi/2$ and can have normal or anomalous "dispersion like" absorption profile for $\phi = 3\pi/4$ and $\phi = \pi/4$, respectively.

To highlight the physical processes described here, it is instructive to go through a description in terms of quantum paths involved in the creation of the coherence $\rho_\sigma = \rho_{cb} + \rho_{da}$. Figure 3 shows the quantum paths involved for the coherence ρ_{da} and ρ_{cb}. For simplicity, we discuss

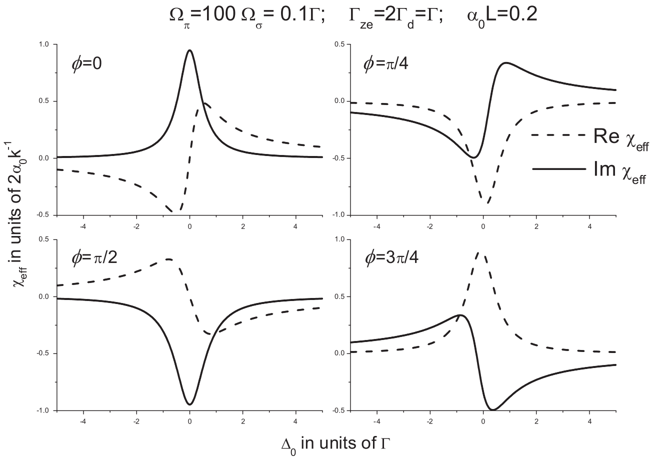

FIG. 2. Coherent control of the susceptibility for the probe beam. We have ($\Delta_0 = \omega_0 - \omega$).

only the case for ρ_{da}. Equivalent features hold for ρ_{cb}. Note that in the stationary regime, $\rho_{dc} = 0$ and does not contribute to the signal. From Eq. (1), we see that at the lowest order (with respect to probe amplitude), ρ_{da} results from the absorption of the probe by the population difference on transition $|a\rangle \leftrightarrow |d\rangle$ (case a) and the diffraction of the control by the ground state Zeeman coherence (case b and c). The ground Zeeman coherence in turn involves excitation by the probe of the transition $|a\rangle \leftrightarrow |d\rangle$ with the phase $e^{-i\phi}$ (case b) and along $|c\rangle \leftrightarrow |b\rangle$ with $e^{i\phi}$ (case c). The two paths correspond, respectively, to crossed-Kerr and phase conjugation-type effects. For $|\Omega_\pi| \gg |\Omega_\sigma|$ and in the stationary state, absorption (linear response) is completely compensated by the "cross-Kerr" term, and the "phase conjugate" term determines the phase dependent response of the medium [from (4)]. Moreover, if an angle is introduced between the control and the probe, this "phase

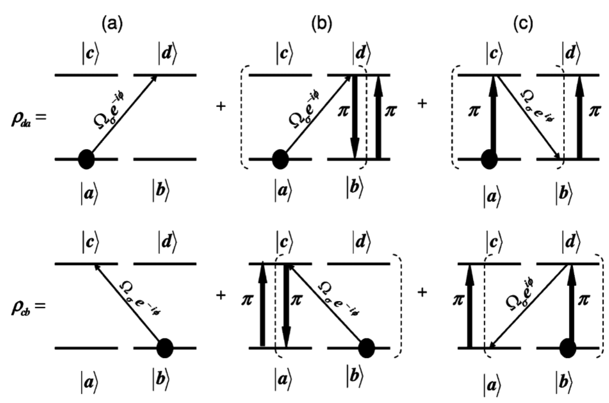

FIG. 3. Quantum paths giving rise to the coherences that modify the σ field behavior. (a) Absorption path. (b) Cross-Kerr type path. Zeeman coherence between the ground states is induced by simultaneous action of the pump and the probe (brackets). The pump is then diffracted. This path cancels with that of absorption. (c) Phase conjugation-type path. The diffraction of the pump leads to a wave conjugate with the probe and determines the optical response.

conjugate" term can be spatially separated and transparency can be induced for the probe [11]. The phase control of the effective susceptibility can thus be related to a wave mixing process where the only radiated field is the conjugate wave and that when added to the incident wave, gives rise to interference inducing a gain-dispersion coupling.

The above description is valid only in the regime of low optical depths. For large optical depths, the phase accumulated during propagation by both the probe and the control cannot be neglected. Again, considering $|\Omega_\pi| \ll \sqrt{\Gamma_d \Gamma}$ and writing $\Omega_j = |\Omega_j| e^{-i\phi_j}$ with $j = \sigma$, π, the effective susceptibility can be written (4) as $\chi_{\text{eff}} = \chi_{\text{lin}} e^{2i\Delta\phi}$ where $\Delta\phi = \phi + \phi_\sigma - \phi_\pi$. The propagation equations for the two fields are $\partial_y \Omega_\pi = i\alpha_0 \Gamma_d \rho_\pi$ and $\partial_y \Omega_\sigma e^{-i\phi} = i\alpha_0 \Gamma_d \rho_\sigma$ [13]. These can be used to write the propagation equation for $\Delta\phi$, and that latter can be analytically solved. By writing $\chi_{\text{lin}} = |\chi_{\text{lin}}| e^{i\phi_L}$ [with $\tan\phi_L(\Delta_0) = \Gamma_d/\Delta_0$], $\Delta\phi$ after propagation distance y is given by

$$\tan\Delta\phi(y) = \frac{\tan\phi_L}{[\frac{\tan\phi_L}{\tan\phi} + 1]e^{-2\alpha_0 y \sin^2\phi_L} - 1}. \tag{6}$$

For $\phi = 0$, $\Delta\phi = 0$, and $\chi_{\text{eff}} = \chi_{\text{lin}}$. Indeed, the total field is linearly polarized and because both the pump and the probe are weak ($|\Omega_j| \ll \sqrt{\Gamma_d \Gamma}$), the response of the medium is linear. The pump can modify the effective susceptibility for the probe only if the total field is elliptic. When $\phi \neq 0$ and for small optical depths such as $\alpha_0 L \sin^2\phi_L \ll 1$, $\Delta\phi \approx \phi$ (L is the length of the medium). No additional phase is introduced during propagation, and the phase control described above is realized. Figure 2 with $\alpha_0 L = 0.2$ corresponds to this case. For optical depths such as $\alpha_0 L \sin^2\phi_L \gg 1$, the phase saturates after a propagation distance $y \approx 1/(\alpha_0 \sin^2\phi_L)$ to the value $\Delta\phi = -\phi_L$ and thus $\chi_{\text{eff}} \approx \chi^*_{\text{lin}}$. This effect can be understood from the equation of evolution of $\Delta\phi$ that is given by $\partial_y(\Delta\phi) = -(k/2)[\chi'_{\text{lin}}(\cos 2\Delta\phi - 1) - \chi''_{\text{lin}} \sin 2\Delta\phi]$ with $\chi_{\text{lin}} = \chi'_{\text{lin}} + i\chi''_{\text{lin}}$. The phase evolves under the action of linear dispersion (χ'_{lin} term) and gain coupling (χ''_{lin} term). The two effects compensate when $\Delta\phi = -\phi_L$ and $\Delta\phi$ no longer evolves. The medium behaves as a linear medium with a response that depends neither on the pump beam intensity nor on the phase shift between the two fields (provided $|\Omega_\sigma/\Omega_\pi| \ll 1$ is fulfilled during the whole propagation). Moreover, the effective susceptibility is the conjugate of the true linear susceptibility. The dispersive properties of the medium are not changed, but the medium is now an amplifier. The Kramers-Kronigs relations are thus violated and it seems as an apparent contradiction. The contradiction is removed by noting that the expression of the effective susceptibility in this situation is valid only for spectral components such as $\alpha_0 L \sin^2\phi_L(\Delta_0) \gg 1$ and cannot be satisfied for arbitrary large frequencies. Equivalently, the distance (and thus the time) needed for the linear response to be established for a pulse with an

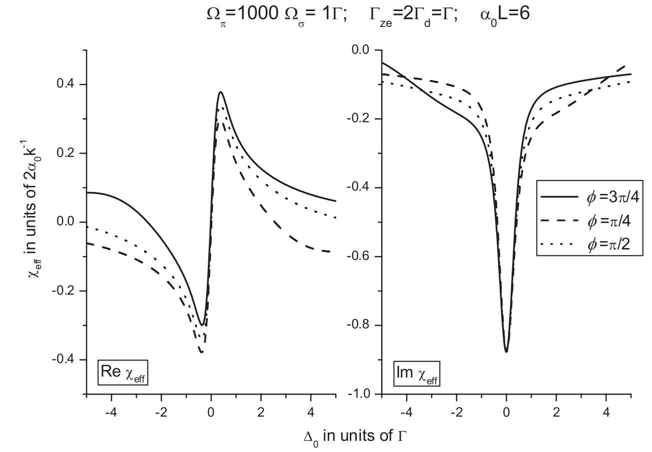

FIG. 4. Behavior of χ_{eff} after propagation in a thick optical medium ($\alpha_0 L \gg 1$). Here, the effective susceptibility saturates with the phase shift for detuning such as $|\Delta_0| \leq \Gamma$.

arbitrary spectrum bandwidth diverges. Causality no longer implies the well known Kramers-Kronig relations. These effects have already been noticed to occur in the (perturbative) four wave mixing processes [14].

Figure 4 exhibits the phase saturation and the behavior of the effective susceptibility after propagation for $\alpha_0 L = 6$. The medium turns into an amplifier with anomalous dispersion in contrast with usual linear medium where a gain peak is accompanied by a normal dispersion. Here, the phase saturation occurs only for the frequencies close to the atomic resonance. Finally, we note that the optical depth cannot be arbitrarily increased as the probe amplification, and the pump absorption violates the condition $|\Omega_\sigma/\Omega_\pi| \ll 1$ that is necessary to observe these effects. Interestingly, when propagation leads to $|\Omega_\sigma/\Omega_\pi| = 1$ and $\Delta\phi = \pm\pi/2$, the matching conditions required to realize a dark state in this system [12] are realized, and the system becomes transparent for both the control and the probe.

In conclusion, we have studied the response of a duplicated two-level system driven by a strong linearly polarized pump field for a weak probe field that is orthogonally polarized to the pump. Two interesting regimes have been identified where the pump and the probe strengths are such as $|\Omega_\sigma| \ll |\Omega_\pi| \ll \sqrt{\Gamma_d \Gamma}$. For small optical density such as $\alpha_0 L \sin^2\phi_L \ll 1$, we have $\chi_{\mathrm{eff}} \approx \chi_{\mathrm{lin}} e^{2i\phi}$. Coherent control of the medium susceptibility is obtained, and the effective susceptibility no longer depends on the strong field characteristics. For large values of the optical density such as $\alpha_0 L \sin^2\phi_L \gg 1$, the effective susceptibility is $\chi_{\mathrm{eff}} \approx \chi^*_{\mathrm{lin}}$ for $y > (\alpha_0 \sin^2\phi_L)^{-1}$ and is independent from both the strong field parameters and the dephasing. Note that the main limitation comes from the condition $|\Omega_\sigma| \ll |\Omega_\pi|$. Indeed, for $|\Omega_\pi| \geq \sqrt{\Gamma_d \Gamma}$, the effective susceptibility depends on the pump intensity, but the phase control and the phase saturation presented here are still possible. The ability to modify and to control the linear response opens the possibilities for many applications. For instance, the medium can switch from an absorber to an amplifier by adjusting the relative phase. The medium also behaves as a fast light medium for $\phi = 0$ and slow light medium for $\phi = \pi/2$ rendering coherent control of the group velocity. Another application of these effects with strong potential for applications emerges from the fact that dipole force applied on the atom depends in such configurations on the relative phase between the fields. Coherent control of the mechanical action of light on atoms can then be considered.

We wish to acknowledge J. Weiner for fruitful discussions.

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