Resonant Nonlinear Dielectric Response in a Photonic Band Gap Material

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We study the dielectric response of impurity two-level atoms in a photonic band gap (PBG) to an applied laser field. In this system, the atoms may exchange energy coherently by resonance dipole-dipole interaction (RDDI) which is assumed to be strong compared to the spontaneous emission rate. When the applied Rabi frequency exceeds the RDDI energy scale, nonlinear saturation of the absorptive part of the susceptibility occurs while the real part of the nonlinear susceptibility remains large. This suggests that doped PBG materials may act as nearly lossless, but highly nonlinear dielectrics.

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Localization of light is an effect which has been predicted [1] to occur in certain strongly scattering dielectric microstructures. It leads to novel strong coupling effects between photons and atoms. The reduction in propagative pathways for photons in such dielectrics enhances the likelihood of photon-photon interactions and nonlinear effects. This is most graphically illustrated in periodic dielectric microstructures exhibiting a complete photonic band gap (PBG) [2–4]. The PBG is a range of frequencies for which no propagating electromagnetic modes are allowed for any direction in three-dimensional space. It was suggested that this would be accompanied by the inhibition of spontaneous emission [5], strong localization of light [6,7], and photon-atom bound states [8]. This bound state is the optical analog of an electronic-impurity bound state in the gap of a semiconductor. Numerous applications of PBG materials have been discussed in Refs. [2–10]. When an atomic transition lies deep inside the gap, the spontaneous emission is strongly suppressed, whereas coherent dipole-dipole interaction between identical atoms persists on a length scale given by the localization length of the dielectric microstructure [8,11]. On length scales short compared to the optical wavelength, the resonance dipole-dipole interaction (RDDI) is mediated by the exchange of high energy virtual photons between the atoms. This leads to new coherent processes such as photon hopping conduction [10] and the formation of a quantum-optical spin glass state [12].

In this Letter we describe the response of two-level atoms placed within an imperfect PBG from an applied laser field. This laser field couples to localized states within the gap. When the average interatomic spacing is less than an optical wavelength the dominant contribution to RDDI comes from the traceless matrix interaction between the dipole vectors on each atom. The absence of ordinary, vacuum, spontaneous emission is manifest in the cutoff of the tranverse (propagating) part of the RDDI on the scale of the localization length. We consider a simplified scalar model using two-level atoms in which the random atomic positions are modeled by means of a Gaussian distribution of RDDI's.

Localized states appear within the PBG by means of dielectric defects within the otherwise periodic microstructure. Alternatively, a small density of propagating states may occur within an incomplete PBG [13]. In either case, spontaneous emission occurs selectively into such a PBG cavity mode. Strong coupling occurs when the energy scale of RDDI, J is large compared to both the rate of atomic population decay $1/T_1$ and the rate of dipole dephasing $1/T_2$. In addition, J is larger than the extent of inhomogeneous broadening of the atomic line due to the random crystal field in the PBG microstructure. In this case, novel nonlinear effects occur: When the Rabi interaction energy scale $\hbar\Omega$ of the applied laser field exceeds the RDDI energy scale J, the absorptive part of the dielectric susceptibility saturates, whereas the real part of the nonlinear response remains large. This suggests that the PBG system may act as nearly lossless, nonlinear material. As we show below, this is a direct consequence of a novel spin-glass-type polarization of the atomic dipoles by the external classical field. In addition, the spectrum of the glass-order parameter exhibits a nonlinear resonant dip, analogous to the Lamb dip in inhomogeneously broadened media. This resonant dip may be valuable for high resolution nonlinear spectroscopy of PBG materials.

We study the response to an applied field of N two-level atoms placed with a PBG material with a pseudogap of localized states. This pseudogap consists of surface-localized modes, localized defect modes in the interior of the photonic crystal, and, in certain cases [13], isolated propagating modes along specific symmetry directions of the periodic microstructure. The coherent part of the model Hamiltonian can be written, in the interaction picture, in the form (choosing units in which $\hbar = 1$)

$$H = -\frac{\delta}{2}\sum_{i}^{N}\sigma_i^z - \Omega\sum_{i}^{N}(\sigma_i^\dagger + \sigma_i) + \sum_{i\neq j}^{N} J_{ij}\sigma_i^\dagger\sigma_j^- . \tag{1}$$

Here, $\sigma_i^\dagger$ and σ_i^- describe atomic excitation and deexcitation of the ith atom, respectively; σ^z describes the atomic inversion; $\Omega = \mu E$ is the resonant Rabi frequency where μ is the atomic dipole moment and E is the amplitude of the applied field; $\delta = \omega - \omega_a$ is the detuning of laser

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frequency ω from the atomic resonant fequency ω_a; and $J_{ij} = J_{ji}$ denotes the RDDI between atoms i and j. The detailed, microscopic evaluation of J_{ij} as a function of atomic distance r_{ij} and atomic configurations may be found in Ref. [14] for free space and Refs. [8,11] for a PBG. In a realistic description, the RDDI term in (1) would be replaced by a traceless tensor interaction for atomic excitation transfer between two sets of triply degenerate atomic orbitals. We simulate this interaction in the simpler two-level atom system by the Gaussian random scalar variable J_{ij} with zero mean value and variance J. In the case of nonzero mean value J_0, the susceptibility has been shown to display "blue" or "red" spectral shifts [15,16]. In this Letter, we focus only on the nonlinear effects caused by fluctuations of RDDI's in a PBG for which $J \neq 0$ and $J_0 \cong 0$.

In the semiclassical (mean field) approximation, the optical Bloch equations for the operator expectation values are [17,18]

$$\frac{d}{dt}\langle\sigma_j^-\rangle = (-1/T_2 + i\delta)\langle\sigma_j^-\rangle - i(\Omega - F_j)\langle\sigma_j^z\rangle, \tag{2a}$$

$$\frac{d}{dt}\langle\sigma_j^z\rangle = -(\langle\sigma_j^z\rangle - \langle\sigma^z\rangle_0)/T_1 + 2i\Omega(\langle\sigma_j^\dagger\rangle - \langle\sigma_j^-\rangle) - 2i(\langle\sigma_j^\dagger\rangle F_j - \langle\sigma_j^-\rangle F_j^*). \tag{2b}$$

Here, $F_j \equiv \sum_{i(\neq j)} J_{ji}\langle\sigma_i^-\rangle$; T_1 and T_2 are the relaxation times of $\langle\sigma^z\rangle$ and $\langle\sigma^-\rangle$, respectively; and $\langle\sigma^z\rangle_0$ is the atomic population inversion in the thermal equilibrium. We use the fact that in the optical domain $\langle\sigma^z\rangle_0 \cong -1$ even at the room temperature. In general, T_1 and T_2 describe spontaneous emission into extraneous modes as well as nonradiative broadening. The collective effects of the incoherent interaction are assumed to be small compared to RDDI's and are simply ignored. Steady-state solutions of (2) satisfy the conditions

$$w_j^2 + w_j + 4(T_1/T_2)\,|p_j|^2 = 0, \tag{3a}$$

$$-\Omega w_j + (\delta + i/T_2)p_j + w_j \sum_{i(\neq j)} J_{ji}p_i = 0. \tag{3b}$$

Here, $p_j \equiv \langle\sigma_j^-\rangle_s$, $w_j \equiv \langle\sigma_j^z\rangle_s$ are steady-state expectation values. It is clear from Eq. (3a) that the population difference w_j is negative for all atoms and is not directly driven by random fluctuations of J_{ij}. On the other hand, random fluctuations in RDDI drive *phase* changes in the atomic dipoles p_i. These phase fluctuations dominate the system behavior in the long time limit. We have performed detailed numerical simulations of Eq. (3) for a small number of atoms which confirm this picture. Below, we present a simple mean-field solution of the steady-state equation (3), compatible with the conclusions of our more general numerical studies.

Configuration averaging over the random atomic positions corresponds to performing a statistical average over the possible values of J_{ij} using the Gaussian distribution. It is denoted by the square brackets $[\cdots]_c$. Using the mean-field approximation $[\langle\sigma_j^z\rangle^2]_c \cong [\langle\sigma_j^z\rangle]_c^2$ (verified numerically), Eq. (3a) becomes $w^2 + w + 4(T_1/T_2)q = 0$. Here, $q \equiv (1/N)\sum_j^N [|p_j|^2]_c$ is the Edwards-Anderson order parameter [19,20] and $w \equiv (1/N)\sum_j^N [w_j]_c$ is the average atomic population difference. Mean-field theory may also be applied to Eq. (3b) by replacing w_j by its average value w. It is convenient to decompose the symmetric matrix J_{ij} using the spectral representation [21] $J_{ij} = \sum_\lambda^N J_\lambda \langle i \mid \lambda\rangle\langle\lambda \mid j\rangle$. Here, J_λ and $\langle\lambda \mid i\rangle$ are the eigenvalues and orthonormal eigenvectors of J_{ij}, respectively. The polarization eigenmode $p_\lambda \equiv \sum_j^N \langle\lambda \mid j\rangle p_j$ is then given by $p_\lambda = \Omega w(\delta + i/T_2 + wJ_\lambda)^{-1}\sum_j\langle\lambda \mid j\rangle$. In the limit of large N, the eigenvalue density $\rho(J_\lambda)$ obeys a semicircular law [22]: $\rho(J_\lambda) = (2\pi\tilde{J}^2)^{-1}(4\tilde{J}^2 - J_\lambda^2)^{1/2}$, where $\tilde{J} = J\sqrt{N}$. Using the semicircular distribution of the eigenvalue J_λ we can easily find average atomic polarization $p = (1/N)\sum_j^N [p_j]_c = \chi\Omega$. Here,

$$\chi \equiv w \int dJ_\lambda \rho(J_\lambda)(\delta + i/T_2 + wJ_\lambda)^{-1} \tag{4}$$

is the susceptibility per atom [17]. The scaled spin-glass order parameter $q_\Omega \equiv q/\Omega^2$ is then given by $q_\Omega = -wT_2\chi''$, where $\chi'' \equiv \mathrm{Im}\chi$. The atomic population difference w is found than as $w = I\chi'' - 1$, where $I \equiv 4T_1\Omega^2$ is a scaled intensity parameter. Using the semicircular eigenvalue density, the susceptibility (4) can be expressed implicitly in the form

$$\chi = (1/2\tilde{J}^2 w)\{\delta + i/T_2 - [(\delta + i/T_2)^2 - 4w^2\tilde{J}^2]^{1/2}\}. \tag{5}$$

In the weak coupling case of $\tilde{J} \ll 1/T_2$, χ and q_Ω reduce to

$$\chi = -(\delta - i/T_2)/[\delta^2 + 1/T_2^2 + 4T_1\Omega^2/T_2] \tag{6}$$

and $q_\Omega = |\chi|^2$. Equation (6) is the conventional nonlinear susceptibility for ordinary materials [17] and exhibits conventional saturation in both the real and imaginary parts. A second limiting case may be recaptured for weak applied field $I \ll 1$. Here, $w \cong -1$ and χ becomes independent of Ω (linear susceptibility χ_L):

$$\chi_L = -(1/2\tilde{J}^2)\{(\delta + i/T_2) - [(\delta + i/T_2)^2 - 4\tilde{J}^2]^{1/2}\}. \tag{7}$$

In this linear regime, it is instructive to consider the relative importance of glassy configurations which are measured by the ratio of the ferroelectric to ferroglass order parameters. At exact resonance $\delta = 0$, $|\chi|^2/q_\Omega = [(1 + 4\tilde{J}^2T_2^2)^{1/2} - 1]/2\tilde{J}^2T_2^2$. Clearly $|\chi|^2/q_\Omega \ll 1$ for $\tilde{J} \gg 1/T_2$. That is, the glassy behavior of the atomic dipoles becomes dominant relative to any macroscopic ferroelectric response. As we will see shortly, the glass-order parameter and atomic polarization change dramatically with nonzero detuning and increased intensity of the

applied field. This suggests that the quantum spin-glass state may be created and controllably altered by means of an external laser field. This also leads to nearly lossless, resonant nonlinearity at high fields.

In Fig. 1 we plot the real (χ') (solid curve) and imaginary (χ'') (dashed curve) parts of the *linear* susceptibility and associated the glass-order parameter q_Ω (dotted curve) as a function of $\delta = \omega - \omega_a$ for the case of $\tilde{J} \gg 1/T_2$. As a result of photon hopping conduction, the excited levels of the impurity atoms broaden into a photonic impurity band [10]. The imaginary part of the linear susceptibility describes the density of states of this impurity band. Outside the impurity band $|\delta| > 2\tilde{J}$, the index of refraction $(\chi')^{1/2}$ remains large while the absorption coefficient (χ'') becomes negligible. Important applications of a linear, transparent, high-index optical material have been suggested in Ref. [23]. In contrast to Ref. [23], the absorptionless index of refraction is present over a large frequency interval. A second feature of the linear susceptibility is that, near resonance, the glass-order parameter is much larger than atomic polarization while far from resonance they are of the same order. We emphasize that in the PBG the linear absorption in the frequency interval $|\delta| \leq 2\tilde{J}$ (dashed curve in Fig. 1) is the result of collectively enhanced photon hopping conduction between impurity atoms rather than single atom relaxation. This kind of absorption within the photonic impurity band, as we now show, can be greatly suppressed by a strong applied field.

For $\tilde{J} \gg 1/T_2$, and in the frequency interval $1/T_2 \ll |\delta| \leq 2\tilde{J}$, $1/T_2$ in Eq. (5) can be ignored. The atomic population difference is then found for the case of strong applied field as $w \cong -|\delta|/2\tilde{J}$. It yields $\chi' \cong -\mathrm{sgn}(\delta)/\tilde{J}$ and $\chi'' \cong (1 - |\delta|/2\tilde{J})/I$. Clearly, in the frequency interval $|\delta| \leq 2\tilde{J}$, χ'' is strongly suppressed ($\sim 1/I$) as intensity of the applied field increases, whereas χ' remains large.

In Fig. 2 we plot the nonlinear susceptibility and the glass-order parameter numerically evaluated from Eq. (5) for the case of $\tilde{J} \gg 1/T_2$. The strong nonlinear suppression of absorption accompanied by large real refractive is particularly evident in Fig. 2(b) where $T_2 \gg I/\tilde{J} \gg 1$. Such a photonic material may have important applications in high-refractive nonlinear devices. It may lead to nonlinear transparency in the form of gap soliton propagation. On the other hand, when dipole dephasing and nonradiative relaxation rates exceed the rate of RDDI hopping conduction both the real and imaginary parts of nonlinear susceptibility exhibit conventional power broadening with increasing intensity of the applied field. In ordinary vacuum, where rapid spontaneous emission also occurs, T_1 and T_2 are many orders of magnitude smaller than in a PBG. This leads to the requirement of high atomic density and very strong applied field Ω for realization of the condition $I = 4T_1\Omega^2 \gg \tilde{J} \gg 1/T_2$ to see the nonlinear effects described in Fig. 2.

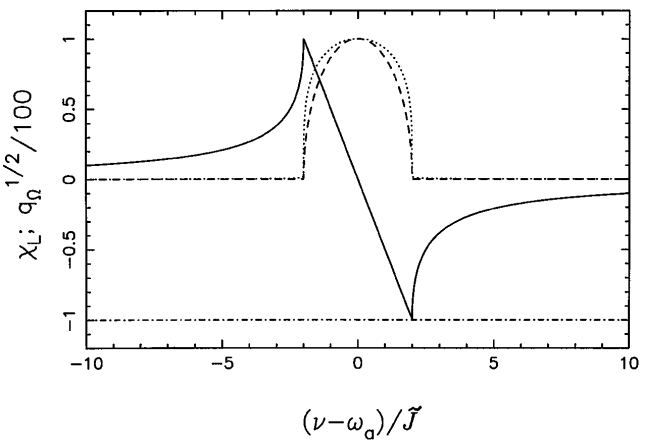

FIG. 1. Real (χ', solid curve) and imaginary (χ'', dashed curve) parts of the linear susceptibility χ_L, associated glass-order parameter $q_\Omega^{1/2}$ (dotted curve) (in units of $\tilde{J} = 1$) and atomic population difference w (dash-dotted curve) as a function of detuning $(\omega - \omega_a)/\tilde{J}$ and for $1/\tilde{J}T_2 = 10^{-4}$.

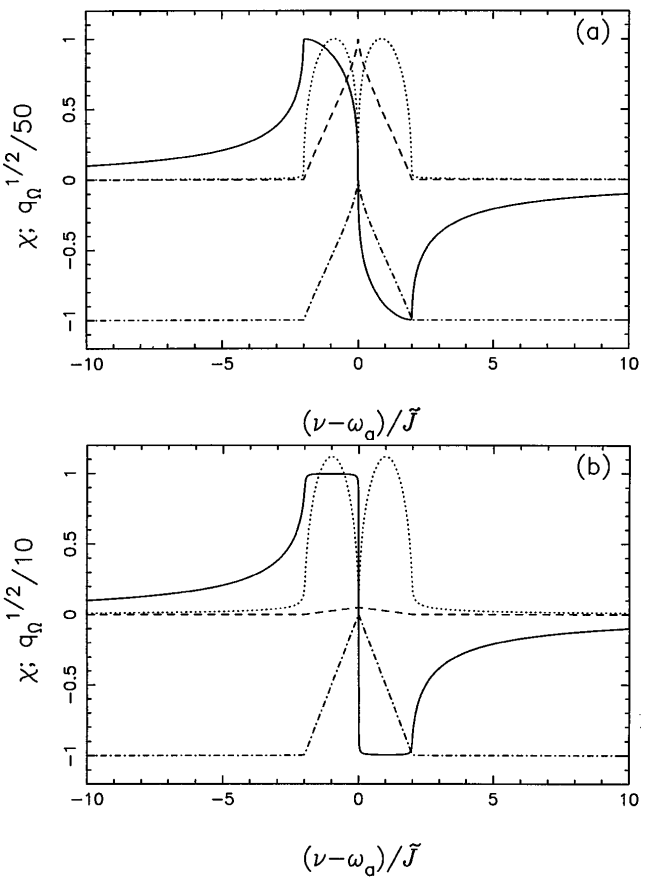

FIG. 2. Real (χ', solid curves) and imaginary (χ'', dashed curves) parts of the nonlinear susceptibility χ, glass-order parameter $q_\Omega^{1/2}$ (dotted curves) (in units of $\tilde{J} = 1$) and atomic population difference w (dash-dotted curves) as a function of detuning $(\omega - \omega_a)/\tilde{J}$ for $1/\tilde{J}T_2 = 10^{-4}$ and for (a) $I/\tilde{J} = 1$ and (b) $I/\tilde{J} = 20$.

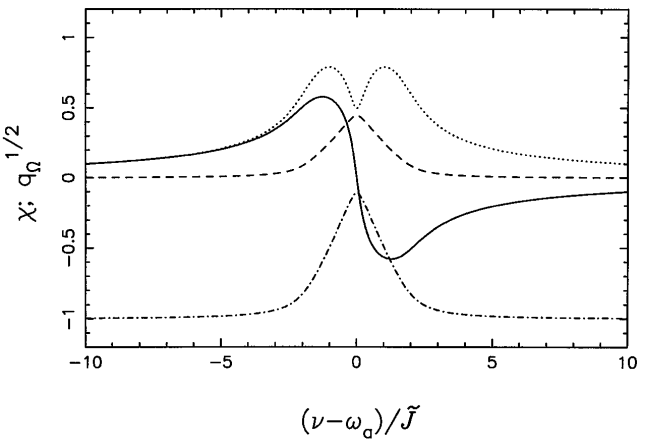

FIG. 3. The same as in Fig. 2 but for $1/\tilde{J}T_2 = 0.2$ and $I/\tilde{J} = 2$.

Unlike the atomic susceptibility, the glass-order parameter displays a resonant dip (dotted curves in Fig. 2). This resonant dip is evident even when $\tilde{J}$ is not much larger than $1/T_2$ (Fig. 3) suggesting that this hole-burning effect can be easily seen in a broad range of PBG materials. The resonant dip appears only in the nonlinear regime and it is associated with saturated absorption to the dressed states with largest probability in the distribution $\rho(J_\lambda)$. Unlike Stark or Doppler broadened media [24], the resonance dip does not appear in the spectrum of atomic population difference (dash-dotted curve). The glass-order parameter can be related to the long time dynamics of the first-order autocorrelation function of the local atomic dipole moment [19]. This in turn can be measured via the electric field autocorrelation function of resonance fluorescence. Spectral hole burning and the Lamb dip have played an important role in nonlinear laser spectroscopy of inhomogeneous media [24]. Hole burning in the glass order parameter field may play an analogous role in PBG materials.

In conclusion, our study of the nonlinear suceptibility of impurity two-level atoms in a PBG has shown that this system exhibits high linear and nonlinear index of refraction accompanied by negligible absorption. For a broad range of parameters, the glassy behavior of the atomic system is dominant. This suggests a practical way of creating and controlling quantum optical spin-glass behavior by a classical applied field. Resonant, lossless, nonlinearity associated with the glass state may lead to low-threshold nonlinear wave propagation and self-induced transparency in a PBG material. A detailed study of the full Maxwell-Bloch equations describing the coupling of the atomic impurity band to propagating, band-edge electromagnetic waves will be presented elsewhere.

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