Coherent Control of Spontaneous Emission near a Photonic Band Edge: A Single-Atom Optical Memory Device

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We demonstrate coherent control of spontaneous emission from a three-level atom with one resonant frequency near the edge of a photonic band gap. As a result of quantum interference and photon localization, spontaneous emission can be totally suppressed or strongly enhanced depending on the relative phase between the control and pump laser fields. The fractionalized steady state inversion of the atom depends sensitively on the initial conditions, suggesting the possibility of a phase-sensitive, optical memory device on the atomic scale. [S0031-9007(97)04879-5]

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Photon localization is a fundamental effect predicted [1] to occur in certain strongly scattering dielectric microstructures. The realization of this effect has been facilitated by the prediction [2,3] and development [4–7] of photonic band gap (PBG) materials. These are lossless materials which exhibit a range of frequencies for which electromagnetic wave propagation is classically forbidden. In addition to strong localization of light [3] at the classical level, these systems lead to the suppression of spontaneous emission [2,8] and the formation of photon-atom bound states [9]. Near a photonic band edge, spontaneous emission dynamics is anomalous [10] and leads to a fractionalized steady-state inversion [8,10,11] for a single atom. Nonexponential spontaneous emission decays into frequency-dependent reservoirs, and other non-Markovian effects in cavities and waveguides have been discussed in the early works of Lewenstein *et al.* [12]. These studies have been extended to the case of a two-level atom driven by an external laser field [13]. Unlike these earlier studies, in a PBG system the emitted photon remains partially localized in the vicinity of the emitting atom. This, in turn, leads to long term memory effects and non-Markovian behavior in the collective spontaneous emission from many atoms [14]. These remarkable changes in the radiative properties of atoms and molecules arise purely from the dielectric environment of the PBG host, without the presence of an external field. On the other hand, coherent interaction of atoms with external laser fields can have a profound effect on radiative dynamics [13], even in ordinary vacuum [15–19].

In this paper we investigate the combined effects of coherent control and photon localization on spontaneous emission from a three-level atom with one resonant frequency at the edge of a PBG. In our system, a pump laser pulse is used to create an excited state of the atom with an atomic Bloch vector specified by the "area" of the incident pulse. A control, cw, laser field with a specific phase relation to the pump laser pulse stimulates radiative transitions between the upper two excited states. It is shown that spontaneous emission can be totally suppressed or strongly enhanced by changing the optical paths and, hence, the relative phase, between the control laser field and the initial atomic Bloch vector determined by the pump laser pulse. Unlike the free space case, the steady-state inversion of the atomic system is strongly dependent on the externally prescribed initial conditions. As a result, such a system may be relevant for a single-atom, phase-sensitive, optical memory device.

The model we consider consists of a three-level atom with two upper levels $|3\rangle$ and $|2\rangle$ and a ground state $|1\rangle$ (Fig. 1). The level $|3\rangle$ is coupled by radiation modes to the ground level $|1\rangle$, and the resonant transition frequency ω_{31} is assumed to be equal to the band edge frequency ω_c of a photonic band gap [9–11,14]. The transition between the two upper levels $|3\rangle$ and $|2\rangle$ is driven by a resonant ($\omega_L = \omega_{32}$) control laser field. The Hamiltonian of the system in the interaction picture has the form,

$$H = i\hbar\Omega(e^{i\phi_c}\sigma_{23} - e^{-i\phi_c}\sigma_{32}) + \sum_{\lambda} \hbar\Delta_\lambda a_\lambda^\dagger a_\lambda + i\hbar \sum_{\lambda} g_\lambda(a_\lambda^\dagger \sigma_{13} - \sigma_{31} a_\lambda). \tag{1}$$

Here $\sigma_{ij} = |i\rangle\langle j|$ $(i, j = 1, 2, 3)$ are the atomic operators, a_λ and $a_\lambda^\dagger$ are the radiation field annihilation and creation operators, Ω is the resonant Rabi frequency, ϕ_c is the phase of the control laser beam which depends on its optical path, and $\Delta_\lambda = \omega_\lambda - \omega_{31}$ is the detuning of the radiation mode frequency ω_λ from the atomic resonant frequency ω_{31}. In the final term of (1), $g_\lambda = (\omega_{31} d_{31}/\hbar)(\hbar/2\epsilon_0\omega_\lambda V_0)^{1/2}\mathbf{e}_\lambda \cdot \mathbf{u}_d$ is the atom-radiation field coupling, where d_{31} and $\mathbf{u}_d$ are the magnitude and unit vector of the atomic dipole moment of the transition $|3\rangle \rightarrow |1\rangle$, V_0 is the quantization volume, $\mathbf{e}_\lambda \equiv \mathbf{e}_{k,\sigma}$

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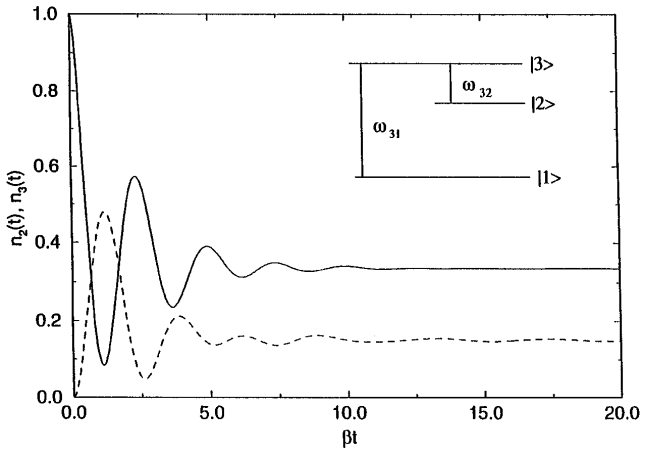

FIG. 1. Atomic population on the upper states $n_3(t) = |b_3(t)|^2$ (solid curve) and $n_2(t) = |b_2(t)|^2$ (dashed curve) as a function of the scaled time βt. Here, $\Omega/\beta = 1$ and the atom is initially on the upper state $|3\rangle$ ($\cos\theta = 1$, $\sin\theta = 0$). The scheme of the three-level atom is shown in the inset.

are the transverse (polarization) unit vectors, and ϵ_0 is the Coulomb constant. In this model we have ignored spontaneous emission $|3\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |1\rangle$. For a V system, level $|2\rangle$ and level $|3\rangle$ have the same symmetry, and the transition frequency ω_{21} is assumed to be far inside the gap so that the spontaneous emission from level $|2\rangle$ to the ground state $|1\rangle$ is ignored [10,11]. Accordingly, the control laser field drives a two-photon transition between state $|2\rangle$ and $|3\rangle$. For a Λ system, $|1\rangle$ and $|2\rangle$ have the same symmetry and the transition $|3\rangle \rightarrow |2\rangle$ is assumed to be far inside the gap. Accordingly, the control laser must be injected into a localized mode arising from a defect in the periodic microstructure.

The atom is assumed to be initially in a coherent superposition of the two upper states and the radiation field is in the vacuum state. The state vector of the system at an arbitrary time t can then be written as

$$|\Psi(t)\rangle = b_3(t)\,|3,\{0\}\rangle + b_2(t)\,|2,\{0\}\rangle + \sum_\lambda b_{1\lambda}(t)e^{-i\Delta_\lambda t}|1,\{\lambda\}\rangle, \quad (2)$$

where the state vectors $|3,\{0\}\rangle$ and $|2,\{0\}\rangle$ describe the atom in its upper states $|3\rangle$ or $|2\rangle$ and no photons are present, and the state vector $|1,\{\lambda\}\rangle$ describes the atom in the ground state $|1\rangle$ and a single photon in a mode $\{\lambda\}$. The time-dependent Schrödinger equation, projected on the one-photon sector of the Hilbert space, takes the form,

$$\frac{d}{dt}\,b_{1\lambda}(t) = g_\lambda b_3(t)e^{-i\Delta_\lambda t}, \quad (3a)$$

$$\frac{d}{dt}\,b_2(t) = \Omega e^{i\phi_c} b_3(t)\,, \quad (3b)$$

$$\frac{d}{dt}\,b_3(t) = -\Omega e^{-i\phi_c} b_2(t) - \sum_\lambda g_\lambda b_{1\lambda}(t)e^{-i\Delta_\lambda t}. \quad (3c)$$

The formal solution of Eq. (3a) is $b_{1\lambda}(t) = g_\lambda \int_0^t b_3(t')e^{i\Delta_\lambda t'}dt'$. Substituting it into Eq. (3c), we get

$$\frac{d}{dt}\,b_3(t) = -\Omega e^{-i\phi_c} b_2(t) - \int_0^t G(t - t')b_3(t')dt', \quad (4)$$

where $G(t - t') = \sum_\lambda g_\lambda^2 e^{-i\Delta_\lambda(t-t')}$ is the delay Green's function. In what follows, we consider both an isotropic PBG as well as a 3D anisotropic PBG. Both models exhibit qualitatively similar behavior. For an isotropic PBG described by the "effective mass" dispersion relation $\omega_k = \omega_\lambda \cong \omega_c + A(k - k_0)^2$ with $A \cong \omega_c/k_0^2$ [9–11] the Green's function takes the form [14], $G(t - t') = \beta^{3/2}e^{-i\pi/4}/\sqrt{\pi(t - t')}$, where $\beta^{3/2} \equiv \omega_{31}^{7/2}d_{31}^2/6\pi\epsilon_0\hbar c^3$. This power law decay with time $t - t'$ describes long term memory effects in spontaneous emission dynamics.

The Laplace transform $\tilde{b}_3(s) = \int_0^\infty e^{-st}b_3(t)dt$ is found from Eqs. (4) and (3b) to be $\tilde{b}_3(s) = (-\Omega e^{i\phi}\sin\theta + s\cos\theta)/D(s)$. Here, the initial amplitudes, $b_3(0) = \cos\theta$ and $b_2(0) = \sin\theta e^{i\phi_p}$, are defined by the "pulse area" and the phase of the pumping laser pulse used for preparing the atom in a coherent superposition of states [20], $\phi = \phi_p - \phi_c$ is the relative phase between the controlling and pumping lasers, and $D(s) = s^2 + s\tilde{G}(s) + \Omega^2$, where $\tilde{G}(s) = \beta^{3/2}e^{-i\pi/4}/\sqrt{s}$ is the Laplace transform of the delay Green's function $G(t)$. It follows that $D(s) = \prod_{j=1}^4(\sqrt{s} - \sqrt{i}\,\upsilon_j)$, where υ_j ($j = 1,\ldots,4$) are the roots of the quartic equation $x^4 + \beta^{3/2}x + \Omega^2 = 0$ and are given by [21] $\upsilon_{1,3} = \sqrt{u}/2 \pm (A - u/4)^{1/2}$ and $\upsilon_2 = \upsilon_4^* = -\sqrt{u}/2 - i(A + u/4)^{1/2}$, where $A = (u^2/4 + \Omega^2)^{1/2}$, $u = (\beta/2^{1/3})[(B + 1)^{1/3} - (B - 1)^{1/3}]$, and $B = [1 + (4/27)(2\Omega/\beta)^6]^{1/2}$.

The amplitude $b_3(t)$ is found from the inverse Laplace transform of $\tilde{b}_3(s)$ through the Mellin inversion formula $b_3(t) = (2\pi i)^{-1}\int_{\epsilon-i\infty}^{\epsilon+i\infty} e^{st}\tilde{b}_3(s)ds$. Here the real number ϵ is chosen so that $s = \epsilon$ lies to the right of all singularities (poles and branch points) of the function $\tilde{b}_3(s)$. The inverse Laplace transform of $\tilde{b}_3(s)$, which contains pole and branch cut contributions, yields

$$b_3(t) = a_1e^{i\upsilon_1^2 t} + a_2e^{i\upsilon_2^2 t} - (\beta^{3/2}e^{-i\pi/4}/\pi) \times \int_0^\infty \frac{e^{-xt}\sqrt{x}\,(\Omega e^{i\phi}\sin\theta + x\cos\theta)}{(x^2 + \Omega^2)^2 - i\beta^3 x}\,dx\,, \quad (5)$$

where $a_1 = 2\upsilon_1(i\Omega e^{i\phi}\sin\theta + \upsilon_1^2\cos\theta)/[(\upsilon_1 - \upsilon_2) \times (\upsilon_1 - \upsilon_3)(\upsilon_1 - \upsilon_4)]$ and $a_2 = 2\upsilon_2(i\Omega e^{i\phi}\sin\theta + \upsilon_2^2\cos\theta)/[(\upsilon_2 - \upsilon_1)(\upsilon_2 - \upsilon_3)(\upsilon_2 - \upsilon_4)]$. The amplitude $b_2(t)$ is easily found by similar methods . It is easy to verify that at $\Omega = 0$ we have $\upsilon_1 = \beta^{1/2}$ and $\upsilon_2 = -\beta^{1/2}(1 - i\sqrt{3})/2$, and Eq. (5) reduces to the two-level spontaneous emission case in [10]. Clearly, the first term on the right-hand side of Eq. (5) corresponds to the photon-atom bound dressed state [9,10] at the frequency $\omega_c - \upsilon_1^2$ inside the gap and with no decay. The

second term corresponds to the dressed state at the frequency $\omega_c + A$ outside the gap with the decay rate equal to $u^{1/2}(A + u/4)^{1/2}$. It is easy to see that this dressed state splitting is the combined effect of the vacuum Rabi splitting caused by the singularity of the density of modes at the band edge [9,10] and the Autler-Townes splitting by the external field. The last term describes the branch cut contribution and tends to zero as $t \to \infty$. As a result of interference between these terms, the spontaneous emission displays oscillatory behavior as depicted in Figs. 1 and 2, where we plot atomic population on the upper levels as a function of the scaled time βt. Clearly, spontaneous emission dynamics is strongly dependent on the control laser and on the initial state of the atom. In the long time limit, only the first term in Eq. (5) remains dominant. The atomic steady-state population on the exited state $|3\rangle$ can be then written as

$$n_{3s} = \lim_{t\to\infty} |b_3(t)|^2 = \frac{4v_1^2|i\Omega e^{i\phi}\sin\theta + v_1^2\cos\theta|^2}{|(v_1 - v_2)(v_1 - v_3)(v_1 - v_4)|^2}. \tag{6}$$

The steady-state atomic population on the upper state $|2\rangle$ can be found analogously as $n_{2s} = \lim_{t\to\infty}|b_2(t)|^2 = \Omega^2 n_{3s}/v_1^4$. Clearly, the steady-state atomic population is strongly dependent on the control laser intensity and the initial state of the atom. For a strong control laser field, n_{3s} and n_{2s} reduce to $n_{3s} = n_{2s} = (1 - \sin 2\theta \sin\phi)/4$. Clearly, when the atom is initially on the upper state $|3\rangle$ ($\theta = \pi$) or state $|2\rangle$ ($\theta = \pi/2$), the steady-state atomic populations, for the case of a strong control laser field, are given by $n_{3s} = n_{2s} = 1/4$. That is, n_{2s} and n_{3s} are independent of the initial relative phase ϕ. In this case, the steady-state atomic population keeps the memory of only the intensity and duration of the pumping laser pulse used for preparing the initial atomic state. If the atom is initially prepared in a coherent superposition of the two upper states $|3\rangle$ and $|2\rangle$, the steady-state atomic population will also keep the memory of the initial relative phase ϕ. For example, when $\theta = \pi/4$, $n_{3s} \cong n_{2s} \cong 0.5$ for $\phi = 3\pi/2$ and $n_{3s} \cong n_{2s} \cong 0$ for $\phi = \pi/2$. This means that the spontaneous emission can be totally suppressed $[(n_{3s} + n_{2s} \cong 1)]$ or strongly enhanced $[(n_{3s} + n_{2s} \cong 0)]$, depending on the initial relative phase ϕ (Fig. 3). As a result, we can control the steady-state atomic population by changing the optical paths of the pumping and controlling lasers. Because of the effects of the photon localization, the atom keeps the memory of the intensity and the phase of the pumping (input) laser. As a result, such a system may behave as an optical memory device on the atomic scale.

Although the isotropic photon dispersion model is formally simple, it overestimates the actual photon density of states at the band edge. For a real dielectric crystal in three dimensions with an allowed point-group symmetry, the band edge is associated with a point $\mathbf{k} = \mathbf{k}_0$ (or a finite collection of symmetry related points) rather than the entire sphere $|\mathbf{k}| = |\mathbf{k}_0|$. In the anisotropic model, we choose the effective mass dispersion relation to be of the form $\omega_{\mathbf{k}} \cong \omega_c + A(\mathbf{k} - \mathbf{k}_0)^2$. Using this anisotropic dispersion relation, the Laplace transform of the delay Green's becomes [10] $\tilde{G}_3(s) \cong \beta_3^{1/2} e^{i\pi/4}\sqrt{s}$, where β_3 is a constant. The Laplace transform $\tilde{b}_3(s)$ has the same form as in the isotropic model if we replace $\tilde{G}(s)$ in $D(s)$ by $\tilde{G}_3(s)$. The amplitude $b_3(t)$ in an anisotropic PBG can be found from the inverse Laplace transform of $\tilde{b}_3(s)$. In Fig. 4 we plot atomic population on the upper level $|3\rangle$ as a function of the scaled time $\beta_3 t$. As in the isotropic PBG case, the dynamics and steady state of the atomic system are strongly dependent on the atomic initial state, and the atom keeps the memory of the intensity and phase of the pumping laser. The most significant difference between the isotropic and anisotropic models occurs in the weak field ($\Omega \to 0$) limit. For the isotropic model, vacuum

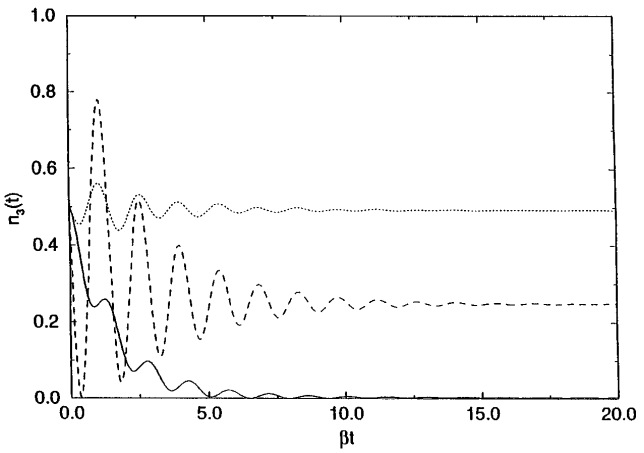

FIG. 2. Atomic population $n_3(t)$ as a function of the scaled time βt for $\Omega/\beta = 2$. The atom is initially in the coherent superposition of the upper states with $\theta = \pi/4$ and for $\phi = 0$ (dashed curve), $\phi = -\pi/2$ (dotted curve), and $\phi = \pi/2$ (solid curve).

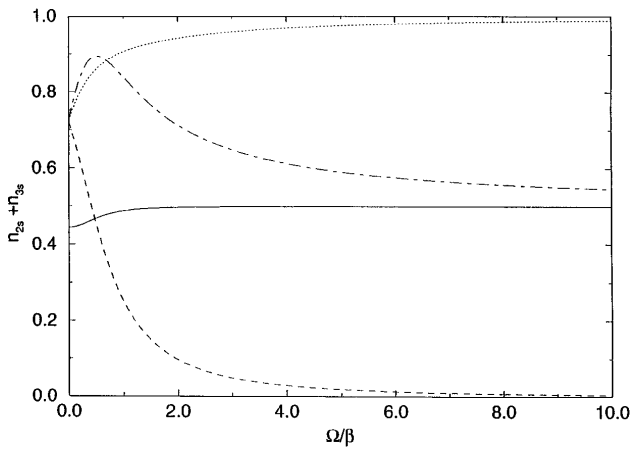

FIG. 3. Steady-state atomic population $n_{2s} + n_{3s}$ as a function of Ω/β. The solid curve describes the atom initially on the upper state $|3\rangle$. The remaining curves describe the atom initially in the coherent superposition of the upper states with $\theta = \pi/4$, and for $\phi = 0$ (dot-dashed curve), $\phi = -\pi/2$ (dotted curve), and $\phi = \pi/2$ (dashed curve).

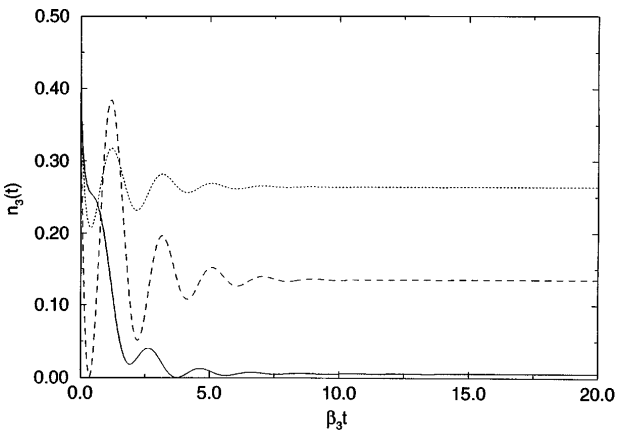

FIG. 4. Atomic population $n_3(t)$ as a function of the scaled time $\beta_3 t$ for the anisotropic PBG and for $\Omega/\beta_3 = 2$. The atom is initially in the coherent superposition of the upper states with $\theta = \pi/4$, and for $\phi = 0$ (dashed curve), $\phi = -\pi/2$ (dotted curve), and $\phi = \pi/2$ (solid curve).

Rabi splitting and fractionalized steady-state inversion occur for all $\omega_{31} \cong \omega_c$. For the anisotropic model, fractionalized inversion occurs only when ω_{31} is slightly below ω_c. However, when the external field Ω is strong, Autler-Townes splitting of the transition $|3\rangle \rightarrow |1\rangle$ is dominant and this distinction is no longer present.

In a real PBG material, dephasing effects on the atomic dipoles caused by interaction with lattice vibrations are a significant perturbation. In particular, the phonon interactions may cause the energy levels of the atom to experience small, random, time-varying Stark shifts. This phenomenon can be expressed mathematically by adding random shifts $\delta\omega_{31}(t)$ and $\delta\omega_{21}(t)$ to the energy differences ω_{31} and ω_{21}, respectively. In the Markovian approximation for atom-phonon interaction [22], $\delta\omega_{31}(t)$ and $\delta\omega_{21}(t)$ can be considered Gaussian random numbers with temporal correlations of the form $\langle\delta\omega_{31}(t)\delta\omega_{31}(t')\rangle = 2\gamma_{31}\delta(t - t')$ and $\langle\delta\omega_{21}(t)\delta\omega_{21}(t')\rangle = 2\gamma_{21}\delta(t - t')$, where γ_{31} and γ_{21} are the dephasing rates. We have carried out a preliminary study of these effects. Even when dephasing rates γ_{31} and γ_{21} are comparable to β, we find that the phase-sensitive memory effects which we obtained above can be recaptured, provided that external Rabi frequency Ω is large compared to the dephasing rates.

In conclusion, we have considered coherent control of spontaneous emission near the edge of a photonic band gap. Because of quantum interference, as well as coherent photon localization, spontaneous emission can be totally suppressed or strongly enhanced, depending on the relative phase of the initial atomic Bloch vector (determined by the pump laser pulse) and the control laser field. The steady-state atomic population on the upper levels keeps the memory of the atomic initial state as well as the phase of the external field.

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