Tailoring Optical Nonlinearities via the Purcell Effect

Peter Bermel, Alejandro Rodriguez, John D. Joannopoulos, and Marin Soljačić
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
(Received 25 April 2007; published 3 August 2007)

We predict that the effective nonlinear optical susceptibility can be tailored using the Purcell effect. While this is a general physical principle that applies to a wide variety of nonlinearities, we specifically investigate the Kerr nonlinearity. We show theoretically that using the Purcell effect for frequencies close to an atomic resonance can substantially influence the resultant Kerr nonlinearity for light of all (even highly detuned) frequencies. For example, in realistic physical systems, enhancement of the Kerr coefficient by one to two orders of magnitude could be achieved.

DOI: 10.1103/PhysRevLett.99.053601 PACS numbers: 42.50.Ct, 42.70.Qs

Optical nonlinearities have fascinated physicists for many decades because of the variety of intriguing phenomena that they display, such as frequency mixing, supercontinuum generation, and optical solitons [1,2]. Moreover, they enable numerous important applications such as higher-harmonic generation and optical signal processing [2–4]. On a different note, the Purcell effect has given rise to an entire field based on studying how complex dielectric environments can strongly enhance or suppress spontaneous emission (SE) from a dipole source [5–9]. In this Letter, we demonstrate that the Purcell effect can also be used to tailor the effective nonlinear optical susceptibility. While this is a general physical principle that applies to a wide variety of nonlinearities, we specifically investigate the Kerr nonlinearity, in which the refractive index is shifted by an amount proportional to intensity. This effect occurs in most materials, modeled here as originating from the presence of a collection of two-level systems. We show theoretically that using the Purcell effect for frequencies close to an atomic resonance can substantially influence the resultant Kerr nonlinearity for light of all (even highly detuned) frequencies.

In hindsight, the modification of nonlinearities through the Purcell effect could be expected intuitively: optical nonlinearities are caused by atomic resonances, hence varying their strengths should influence the strengths of nonlinearities as well. Nevertheless, to the best of our knowledge, this interesting phenomenon has not thus far been described in the literature. Moreover, as we show below, it displays some unexpected properties. For example, while increasing SE strengthens the resonance by enhancing the interaction with the optical field, it actually makes the optical nonlinearity weaker. Furthermore, phase damping (e.g., elastic scattering of phonons), which is detrimental to most optical processes, plays an essential role in this scheme, because in its absence, these effects disappear for large detunings (i.e., the regime where low-loss switching occurs).

A simple, generic model displaying Kerr nonlinearity is a two-level system. Its susceptibility has been calculated to all orders in both perturbative and steady state limits [2]. However, this derivation is based on a phenomenological model of decay observed in a homogeneous medium and does not necessarily apply to systems in which the density of states (DOS) is strongly modified, such as a cavity or a photonic crystal (PhC) band gap. Following an approach similar to Ref. [10], the validity of this expression can be established from a more fundamental point of view. Start by considering a collection of N two-level systems per unit volume in a PhC cavity, whose levels are labeled a and b. The corresponding Hamiltonian is given by the sum of the self-energy and interaction terms. Using the electric dipole approximation, one obtains:

$$H = \hbar[\omega_a \sigma_{aa} + \omega_b \sigma_{bb} + \Omega(t)\sigma_{ab} + \Omega^*(t)\sigma_{ba}], \quad (1)$$

where $\sigma_{ij} = c_i^\dagger c_j$ is the operator that transforms the fermionic state j to the fermionic state i, $\Omega(t) = -\vec{\mu}\cdot\vec{E}(t)/\hbar$ is the Rabi amplitude of the applied field as a function of time, and the scalar dipole moment μ is defined in terms of its projection along the applied field $\vec{E}(t)$. In general, if this system is weakly coupled to the environmental degrees of freedom, then the time scale for the observable dynamics of the system is less than the time scale of the "memory" of the environment. In this case, information sent into the environment is irretrievably lost—this is known as the Markovian approximation [11]. The dynamics of this system can then be modeled by the Lindbladian $\mathcal{L}$, which is a superoperator defined by $\dot{\rho} \equiv \mathcal{L}[\rho]$. In general, one obtains the following master equation from the Lindbladian:

$$\dot{\rho} = -(i/\hbar)[H,\rho] + \sum_\mu [L_\mu \rho L_\mu^\dagger - \tfrac{1}{2} L_\mu^\dagger L_\mu \rho - \tfrac{1}{2}\rho L_\mu^\dagger L_\mu]. \quad (2)$$

Using the only two quantum jump operators that are allowed in this system on physical grounds—$L_1 \equiv \sigma_{ab}/\sqrt{T_1}$ and $L_2 \equiv \sigma_{bb}\sqrt{\gamma_{\text{phase}}}$ [11]—one can obtain the following dynamical equations:

$$\frac{d\rho_{ba}}{dt} = -(i\omega_{ba} + T_2^{-1})\rho_{ba} + i\Omega(t)(\rho_{bb} - \rho_{aa}) \quad (3)$$

$$\frac{d(\rho_{bb} - \rho_{aa})}{dt} = -\frac{(\rho_{bb} - \rho_{aa}) + 1}{T_1} - 2i[\Omega(t)\rho_{ab} - \Omega^*(t)\rho_{ba}], \quad (4)$$

where $\omega_{ba} \equiv \omega_b - \omega_a$, T_1^{-1} is the rate of population loss of the upper level, and $T_2^{-1} = (1/2)T_1^{-1} + \gamma_{\rm phase}$ is the rate of polarization loss for the off-diagonal matrix elements. The prediction of exponential decay via SE is known as the Wigner-Weisskopf approximation [12]. Although it has been shown that the atomic population can display unusual oscillatory behavior in the immediate vicinity of the photonic band edge [13,14], theoretical [10] and experimental considerations [15,16] show that this approximation is fine for resonant frequencies well inside the photonic band gap. In the rest of this Letter, this is assumed to be the case. Near resonance, one makes the rotating wave approximation for Eqs. (3) and (4), and then solves for the steady state. If the polarization is defined by $P = N\mu(\rho_{ba} + \rho_{ab}) = \chi E$, where χ is the total susceptibility to all orders, one obtains the following well-known expression for the susceptibility [2,10]:

$$\chi = -\frac{N\mu^2(\omega - \omega_{ba} - i/T_2)T_2^2/\hbar}{1 + (\omega - \omega_{ba})^2 T_2^2 + (4/\hbar^2)\mu^2|E|^2 T_1 T_2}. \quad (5)$$

In general, Eq. (5) may be expanded in powers of the electric field squared. Of particular interest is the Kerr susceptibility, also in Ref. [2]:

$$\chi^{(3)} = \frac{4}{3}N\mu^4 \frac{T_1 T_2^2(\Delta T_2 - i)}{\hbar^3(1 + \Delta^2 T_2^2)^2}, \quad (6)$$

where $\Delta \equiv \omega - \omega_{ba}$ is the detuning of the incoming wave from the electronic resonance frequency. For large detunings $\Delta T_2 \gg 1$, one obtains the approximation that:

$$\mathrm{Re}\,\chi^{(3)} \approx \frac{4}{3}N\mu^4\left(\frac{1}{\hbar\Delta}\right)^3 \frac{T_1}{T_2}. \quad (7)$$

Of course, there are many types of materials to which a simple model of noninteracting two-level systems does not apply. However, it has been shown that some semiconductors such as InSb (a III-V direct band gap material) can be treated as a collection of independent two-level systems with energies given by the conduction and valence bands, and they yield reasonable agreement with experiment [17]. If the parameter Δ is defined in terms of the band gap energy such that $\Delta \equiv \omega_G - \omega$, then one can look at the regime $\Delta T_2 \gg 1$ studied above and obtain the following equation:

$$\mathrm{Re}\,\chi^{(3)} \approx -\frac{1}{30\pi\hbar^3}\left(\frac{eP}{\hbar\omega}\right)^4 \left|\frac{2m_r}{\hbar\Delta}\right|^{3/2} \frac{T_1}{T_2}, \quad (8)$$

where P is a matrix element discussed in Ref. [17], ω_G is the direct band gap energy of the system, and m_r is the reduced effective mass of the exciton. This equation displays the same scaling with lifetimes as Eq. (7), so the considerations that follow should also apply for such semiconductors.

Now, consider the effects of changing the SE properties for systems modeled by Eqs. (7) and (8). When SE is suppressed, as in the photonic band gap of a PhC, T_1 will become large while T_2 remains finite, thus enhancing $\chi^{(3)}$ by up to one or more orders of magnitude (for materials with the correct properties). For large detunings (where $\Delta T_{2,\rm vac} \gg 1$), we expect that $\chi^{(3)}$ will scale as T_1/T_2. The enhancement of the real part of $\chi^{(3)}$ is defined to be $\eta \equiv \mathrm{Re}\chi^{(3)}_{\rm Purcell}/\mathrm{Re}\chi^{(3)}_{\rm hom}$, where $\chi^{(3)}_{\rm Purcell}$ is the nonlinear susceptibility in the presence of the Purcell effect, while $\chi^{(3)}_{\rm hom}$ is the nonlinear susceptibility in a homogeneous medium. Since $T_1^{-1} = \Gamma_{\rm rad} + \Gamma_{\rm nr}$, the maximum enhancement is predicted to be roughly:

$$\eta \approx \frac{T_{1,\rm Purcell}T_{2,\rm vac}}{T_{1,\rm hom}T_{2,\rm Purcell}} = \frac{\frac{1}{2}\Gamma_{\rm nr} + \gamma_{\rm phase}}{\frac{1}{2}(\Gamma_{\rm nr} + \Gamma_{\rm rad}) + \gamma_{\rm phase}} \frac{\Gamma_{\rm rad} + \Gamma_{\rm nr}}{\Gamma_{\rm nr}}, \quad (9)$$

where $\Gamma_{\rm rad}$ is the radiative decay rate in vacuum. Since the Purcell effect increases the amplitude of $\chi^{(1)}$, one might also expect it to increase the amplitude of $\chi^{(3)}$; however, according to Eq. (9), the opposite is true. This can be understood by noting that Purcell enhancement decreases the allowed virtual lifetime, and thus, the likelihood of nonlinear processes to occur [18]. Moreover, since the Purcell factor [5] is calculated by only considering the photon modes [7], one would not necessarily expect phase damping effects to play a role, in marked contrast with Eq. (9). This result comes about because $\chi^{(3)}$ comes directly from the polarization of the medium, which exhibits a significant T_2 dependence. The presence of large phase damping effects makes T_2 effectively constant, which means that suppression of SE (caused by the absence of photonic states at appropriate energies [6]) can enhance Kerr nonlinearities by one or more orders of magnitude, while enhancement of SE can suppress these nonlinearities. For the case where Purcell enhancement takes place, T_1 decreases while T_2 does not change as rapidly, when $T_1 \gg \gamma_{\rm phase}^{-1}$. Otherwise, for sufficiently small T_1, T_2 will scale in the same way and $\chi^{(3)}$ will remain approximately constant for large detunings, where $\Delta T_2 \gg 1$. This opens up the possibility of suppressing nonlinearities in photonic crystals (to a certain degree). For processes such as four-wave mixing or cross phase modulation, $\chi^{(3)}$ will generally involve a detuning term and will differ from Eq. (6). It is also interesting to note that this enhancement scheme will generally not increase nonlinear losses, which are a very important consideration in all-optical signal processing. If the nonlinear switching figure of merit ξ is defined by $\xi = \mathrm{Re}\chi^{(3)}/(\lambda \mathrm{Im}\chi^{(3)})$ [19], then $\xi_{\rm Purcell}/\xi_{\rm vacuum} = T_{2,\rm Purcell}/T_{2,\rm vacuum} \geq 1$, for all cases of suppressed SE.

The general principle described thus far should apply for any medium where the local DOS is substantially modified. In what follows, we show how this effect would manifest itself in one such exemplary system: a PhC. This example serves as an illustration as to how strong nonlinearity suppression or enhancement effects could be achieved in realistic physical systems. It consists of a 2D triangular lattice of air holes in dielectric ($\epsilon = 13$), with a two-level system placed in the middle, as in Fig. 1.

Note that the vast majority of PhC literature is generally focused on modification of dispersion relations at the frequency of the light that is sent in as a probe. By contrast, in the current work, it is only essential to modify the dispersion relation for the frequencies close to the atomic resonances; the dispersion at the frequency of the light sent in as a probe can remain quite ordinary.

First, consider the magnitude of the enhancement or suppression of SE in this system. Clearly, since there are several periods of high contrast dielectric, two effects are to be expected. First, there will be a substantial but incomplete suppression of emission inside the band gap. Second, there will be an enhancement of SE outside the band gap (since the DOS is shifted to the frequencies surrounding the band gap). For an atom polarized in the direction out of the 2D plane, only the TM polarization need be considered.

We numerically obtain the enhancement of SE by performing two time-domain simulations in Meep [20], a finite difference time-domain code which solves Maxwell's equations exactly with no approximations, apart from discretization (which can be systematically reduced) [21]. First, we calculate the SE of a dipole placed in the middle of the PhC structure illustrated in Fig. 1, then divide by the SE rate observed in vacuum. The resulting values of T_1 and T_2 are calculated numerically, and Eq. (6), in conjunction with the definition of the enhancement factor η, is used to plot Fig. 3. The results are plotted in Fig. 2.

A GaAs-AlGaAs single quantum well can lie in the interesting regime discussed above, where the radiative loss rate $\Gamma_{\rm rad}$ dominates the nonradiative loss rate $\Gamma_{\rm nr}$ as well as the overall loss rate of the quantum well, for certain temperatures [22]. Equation (8) implies that substantial enhancement of the Kerr coefficient occurs in that regime.

At a temperature of about 200 K, $\Gamma_{\rm nr} \approx 0.1\Gamma_{\rm rad}$ [22]. Although experimental measurements for $\gamma_{\rm phase}$ are unavailable to the authors, the presence of a substantial phonon bath at that temperature leads one to expect a fairly large value, which may be conservatively estimated by $10\Gamma_{\rm rad}$. These results are displayed in Fig. 3(a). Note that enhancement is primarily observed inside the photonic band gap (cf. Fig. 2). We observe an enhancement in the real part of the Kerr coefficient up to a factor of 12, close to the predicted maximum enhancement factor of 10.48 in the regime of large detunings ($\Delta T_2 \gg 1$).

Also, at a temperature of about 225 K, $\Gamma_{\rm nr} \approx \Gamma_{\rm rad}$ [22], and again we take $\gamma_{\rm phase} = 10\Gamma_{\rm rad}$. These results are displayed in Fig. 3(b). In this case, we observe an enhancement up to a factor of 2.5, close to the predicted maximum enhancement factor of 1.91 in the regime of large detunings ($\Delta T_2 \gg 1$).

Finally, we note that close to room temperature (285 K), the system in Ref. [22] displays $\Gamma_{\rm nr} \approx 10\Gamma_{\rm rad}$, which is predicted to yield a maximum enhancement factor of 1.06. Since this number is fairly negligible, it illustrates that this approach has little impact when nonradiative losses dominate the decay of the electronic system.

On the other hand, some recent work has demonstrated that a single quantum dot can demonstrate predominantly radiative decay in vacuum even at room temperature, e.g., single CdSe/ZnS core-shell nanocrystals with a peak emission wavelength of 560 nm, with $\Gamma_{\rm rad} \approx 39\Gamma_{\rm nr}$ [23]. Even bulk samples of similar nanocrystals have been shown to yield a significant radiative decay component, corresponding to $\Gamma_{\rm rad} \approx \Gamma_{\rm nr}$ [24]. Thus, we predict that with strong suppression of radiative decay, nonlinear enhancement of a factor of 2 or more could be observed at room temperature.

We now discuss the implications of this effect on previous work describing nonlinearities in photonic crystals, such as Ref. [25], and the references therein. Most past

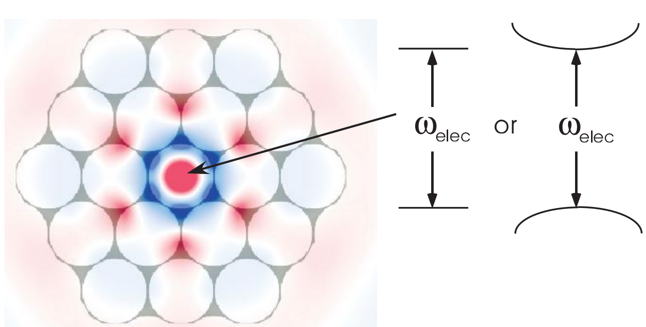

FIG. 1 (color). A 2D triangular lattice of air holes in dielectric ($\epsilon = 13$). On top of the dielectric structure in gray, the E_z field is plotted, with positive (negative) values in red (blue). A small region of nonlinear material is placed exactly in the center of the structure. This material may be, for example, either two-level atoms, quantum wells, or some semiconductors such as InSb.

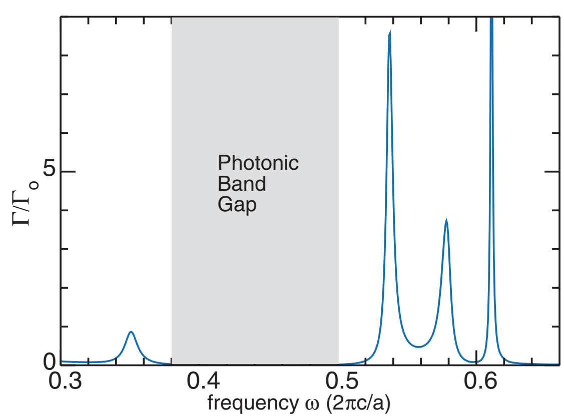

FIG. 2 (color). Relative enhancement of the TM local DOS for Fig. 1, as measured in the time-domain simulation rate of emission, Γ, normalized by the emission rate in vacuum, Γ_o

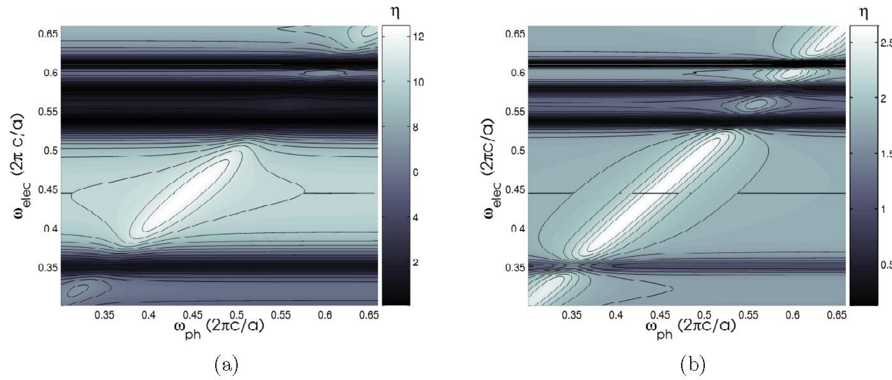

FIG. 3 (color). Contour plot of Kerr enhancement $\eta \equiv \mathrm{Re}\chi^{(3)}_{\mathrm{Purcell}}/\mathrm{Re}\chi^{(3)}_{\mathrm{hom}}$ as a function of probe (ω_{ph}) and electronic transition (ω_{elec}) frequencies, for a system like a single quantum well of GaAs-AlGaAs, assuming $\gamma_{\mathrm{phase}} = 0.025(2\pi c/a)$ (a) at $T = 200$ K, with $0.1\gamma_{\mathrm{phase}} = 10\Gamma_{\mathrm{nr}} = \Gamma_{\mathrm{rad}}$, and (b) at $T = 225$ K, with $0.1\gamma_{\mathrm{phase}} = \Gamma_{\mathrm{nr}} = \Gamma_{\mathrm{rad}}$.

experiments should not have observed this effect, because they were designed with photonic band gaps at optical frequencies significantly smaller than the frequencies of the electronic resonances generating the nonlinearities, in order to operate in a low-loss regime. Furthermore, in most materials, nonradiative decays will dominate radiative decays at room temperature. Finally, all the previous *analyses* are still valid as long as one considers the input parameters to be effective nonlinear susceptibilities, which come from natural nonlinear susceptibilities modified in the way described by this Letter.

In conclusion, we have shown that the Purcell effect can be used to tailor optical nonlinearities. This principle manifests itself in an exemplary two-level system embedded in a PhC; for realistic physical parameters, enhancement of Kerr nonlinearities by more than an order of magnitude is predicted. The described phenomenon is caused by modifications of the local DOS near the resonant frequency. Thus, this treatment can easily be applied to analyze the Kerr nonlinearities of two-level systems in almost any geometrical structure in which the Purcell effect is substantial (e.g., PhC fibers [26], optical cavities). It also presents a reliable model for a variety of materials, such as quantum dots, atoms, and certain semiconductors. Future investigations will involve extending the formalism in this manuscript to other material systems.

The authors would like to thank R. Boyd, E. Ippen, D. Kleppner, V. Vuletic, M. Segev, S. G. Johnson, and M. Rudner for valuable discussions. This work was supported in part by the Army Research Office through the Institute for Soldier Nanotechnologies under Contract No. DAAD-19-02-D0002.

[1] P. Drazin and R. Johnson, *Solitons: An Introduction* (Cambridge University Press, Cambridge, England, 1989).
[2] R. W. Boyd, *Nonlinear Optics* (Academic, New York, 1992).
[3] M. Nielsen and I. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, England, 2000).
[4] G. Agrawal, *Applications of Nonlinear Fiber Optics, Optics and Photonics* (Academic, New York, 2001).
[5] E. Purcell, Phys. Rev. **69**, 681 (1946).
[6] D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981).
[7] H. Y. Ryu and M. Notomi, Opt. Lett. **28**, 2390 (2003).
[8] P. Bermel, J. D. Joannopoulos, Y. Fink, P. A. Lane, and C. Tapalian, Phys. Rev. B **69**, 035316 (2004).
[9] D. Englund, D. Fattal, E. Waks, G. Solomon, B. Zhang, T. Nakaoka, Y. Arakawa, Y. Yamamoto, and J. Vuckovic, Phys. Rev. Lett. **95**, 013904 (2005).
[10] S. John and T. Quang, Phys. Rev. Lett. **76**, 2484 (1996).
[11] J. Preskill, Quantum Computation Lecture Notes, http://www.theory.caltech.edu/people/preskill/ph229/, 2004.
[12] C. Cohen-Tannoudji, B. Diu, and F. Laloë, *Quantum Mechanics* (Wiley, New York, 1977).
[13] S. John and T. Quang, Phys. Rev. A **50**, 1764 (1994).
[14] P. Lambropoulos, G. M. Nikolopoulos, T. R. Nielsen, and S. Bay, Rep. Prog. Phys. **63**, 455 (2000).
[15] M. Bayer, T. L. Reinecke, F. Weidner, A. Larionov, A. McDonald, and A. Forchel, Phys. Rev. Lett. **86**, 3168 (2001).
[16] P. Lodahl, A. F. van Driel, I. S. Nikolaev, A. Irman, K. Overgaag, D. Vanmaekelbergh, and W. L. Vos, Nature (London) **430**, 654 (2004).
[17] D. Miller, S. Smith, and B. Wherrett, Opt. Commun. **35**, 221 (1980).
[18] J. J. Sakurai, *Modern Quantum Mechanics* (Addison-Wesley, Reading, MA, 1994).
[19] G. Lenz, J. Zimmermann, T. Katsufuji, M. E. Lines, H. Y. Hwang, S. Spälter, R. E. Slusher, S.-W. Cheong, J. S. Sanghera, and I. D. Aggarwal, Opt. Lett. **25**, 254 (2000).
[20] A. Farjadpour, D. Roundy, A. Rodriguez, M. Ibanescu, P. Bermel, J. D. Joannopoulos, S. G. Johnson, and G. Burr, Opt. Lett. **31**, 2972 (2006).
[21] A. Taflove and S. C. Hagness, *Computational Electrodynamics* (Artech House, Norwood, MA, 2000), 2nd ed.
[22] S. Kraiem, F. Hassen, H. Maaref, X. Marie, and E. Vaneelle, Opt. Mater. (Amsterdam) **17**, 305 (2001).
[23] X. Brokmann, L. Coolen, M. Dahan, and J. P. Hermier, Phys. Rev. Lett. **93**, 107403 (2004).
[24] D. V. Talapin, A. L. Rogach, A. Kornowski, M. Haase, and H. Weller, Nano Lett. **1**, 207 (2001).
[25] M. Soljacic and J. D. Joannopoulos, Nat. Mater. **3**, 211 (2004).
[26] N. M. Litchinitser, A. K. Abeeluck, C. Headley, and B. J. Eggleton, Opt. Lett. **27**, 1592 (2002).