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Tuning and switching of the spontaneous emission in one-dimensional photonic crystals

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Abstract

If the modal density available to an excited atom is varied on the time scale of its lifetime, then we can expect the natural process of spontaneous emission (SE) to become dynamically manipulable. We consider various experimental possibilities and focus on an atom embedded in a photonic crystal designed to have a band edge in the vicinity of the frequency of the emitted light. Specifically, we calculate the rate of SE by erbium ions (radiating at the wavelength $1.54 \mu m$) implanted in a one-dimensional silicon/silica photonic crystal. The semiconductor layers are assumed to be strongly doped; by tuning the impurity density the free carrier concentration changes and the photonic bands shift. As a result, the SE rate exhibits significant dependency on the level of charge injection. © 2006 Published by Elsevier B.V.

Is it possible to vary spontaneous emission SE *actively* – in real time? The very idea seems to involve a contradiction of terms: "spontaneous" implies a process that is inherent to the emitting atom, independent of experimental conditions! Nevertheless, in a pioneering paper [1], Purcell pointed out that SE is altered if the radiated field is constrained to satisfy a set of boundary conditions. In other words, the lifetime of an excited atomic state changes if the atom is located in a material environment, rather then radiating in free space. The rate of SE can be inhibited (and even prohibited) or enhanced, depending on the modal density (or density of states). If now the external conditions are varied on the time-scale of the excited state's lifetime, then we can expect that the natural process of SE becomes manipulable in real time. Is this feasible?

Let us consider three types of emitters. (a) *Rare earth* ions. In Er^{3+} the transition $4I_{13/2} \rightarrow 4I_{15/2}$, involving f-shell electrons, gives rise to emission at the important communications wavelength $\lambda_0 = 1.54 \,\mu\text{m}$. The radiative lifetime (with the ions implanted in glass) is [2] $\tau = 22 \,\text{ms}$. (b) *Nano*-

particles (quantum dots). These are frequently II–VI or III– V semiconductor compounds such as CdSe; they have been implanted in opal-type photonic crystals (PCs) [3]. Also, silicon nanocrystals have been implanted in silica [4]. Such quantum dots emit in the visible or near infrared; the lifetimes vary over many orders of magnitudes and can be as long as ~10 ms. (c) *Recombination in semiconductors*. The radiative lifetime due to recombination of photo-excited electrons and holes varies widely among semiconductors and depends strongly on the impurity density. For instance, with a density of $10^{17}/\text{cm}^3$ of majority carriers, the minority carrier lifetime in Si, Ge, and GaP is on the order of a ms. For intrinsic Si and Ge it is, respectively, 4.6 h and 0.61 s [5]. Next, are the temporal scales of tuning compatible with these lifetimes?

To answer this question, we discuss four possibilites of tuning of the optical response. (A) *Manipulation of the free-carrier density in semiconductors*. The mechanisms of thermal excitation and doping in PCs have been explored in a series of papers [6,7]. In particular, we found that the transmission of light through a one-dimensional (1D) PC of alternating Si/SiO₂ layers can be sensitively tuned at the wavelength $1.54 \mu m$, [7] mentioned in (a). The intensity of high photoexcitation applied to semiconductors can be modulated as rapidly as the laser beam can be pulsed

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(repetition rates ≤ 1 ns). An even more rapid band-edge tuning, in a 2D silicon PC, was achieved by means of 0.3 ps pulses [8]. And the response time of forward biasing at heterojunctions is ~ 1 ns [9]. (B) Tuning of liquid crystal*infilled PCs* is a very active field, recently reviewed in Ref. [10]. Such tuning may be performed by changing the temperature or by applying a variable electric field, or by shining on pulsating high-intensity light; the response time is on the order of 1 ms. We quote a few notable papers [11]. (C) Employing the thermo-optic effect. Very recently, Vlasov et al. [12] have taken advantage of this effect to tune, in real time, the group velocity of waves propagating in a PC waveguide. The time-scale of the tuning was 0.1 µs. (D) Nonlinear modulation of the refractive index of an insulator. To quote Yablonovitch [13], "ideally, in nonlinear optics, we regard the refractive index of a medium to be a time-variable function totally under the control of the experimentalist". With solid state, (mode locked) pulsed lasers it is possible to achieve repetition rates as small as ~ 1 ns with fs pulses [14].

As we see from the list (A)–(D), the *slowest* tuning agent considered varies on the time scale of 1 ms. Then, comparing with the lifetimes for SE in the examples (a)–(c), we note that the *quickest* emission occurs also in about 1 ms. Hence, we come to the conclusion that external intervention in the process of SE should be feasible. We expect the rate of emission to depend in an important way on the details of the tuning agent. To quantify this problem we assume that the radiating atom is embedded in a dielectric medium whose permittivity ϵ is being dynamically tuned by some unspecified mechanism. If the temporal dependence $\epsilon = \epsilon(t)$ is given – what is the corresponding rate of SE?

We are not aware of any measurement of the SE by an atom in a dynamically varying host medium. If $\epsilon = \epsilon(t)$, the nature of the normal modes in the medium will strongly depend on the specific form of the time-dependence. As a result, the behavior of the SE should also change qualitatively. Even for slow temporal variation of the dielectric function, the adiabatic principle cannot be expected to be indicative of the real-time behavior of the SE rate. A theory of SE in a temporally varying medium is now being developed by the first author (PH). For the time being, we will not concern ourselves with SE in real time. Rather, we will focus on a specific example that demonstrates appreciable dependence of the SE rate on an external agent of tuning. PCs [15] are often characterized by sharp photonic band edges and are, therefore, singularly attractive choices for hosting the radiating atom. This is true because the PC can be designed so as to have one of its band edges approximately coincide with the frequency of emission. Then a modest variation of the tuning agent is expected to produce a sizeable change in the emission rate. Regarding the choices (a)-(c) and (A)-(D) discussed above, we select the (a)-(A) combination. Specifically, this Letter is concerned with the simulation of SE by Er^{3+} ions embedded in a 1D PC of alternating Si and SiO₂ layers [16]. These materials have been selected because of their ease of fabrication, technological importance, and large dielectric contrast. The Er ions are chosen because they emit at the near-infrared wavelength $\lambda_0 = 1.54 \,\mu\text{m}$, very important for communications by optical fibers. We assume that the Si layers are strongly doped with impurity donor atoms. The concentration of these impurities is varied externally – and this is the tuning agent commented on above. Presumably, the mechanism would be carrier injection - either photoexcitation or forward biasing; the specifics are of no consequence for the considerations that follow. For highly extrinsic Si, the density of free electrons in the conduction band is practically equal to the donor density. The tuning of these densities modulates the dielectric constant of the semiconductor and this, in turn, shifts the photonic band edges. We then conclude that the tuning of the charge injection modulates the density of states available to the radiating atom. This explains the idea behind tuning of the SE rate for the particular example considered.

The semiconductor layers of the PC are modeled as a plasma of free electrons in the conduction band and free holes in the valence bands. The density of these, as function of the donor density N (and no acceptors) is [17] $n_{e,h} = [N^2 + n_i^2(T)]^{1/2} \pm N/2$ where the intrinsic density $n_i(T)$ is very small in comparison to N. The corresponding electron and hole plasma frequencies are $\omega_{pe,h} = (4\pi n_{e,h}e^2/\epsilon_{\infty}m_{e,h})^{1/2}$. Here ϵ_{∞} is the high-frequency dielectric constant equal to 11.7 for Si. We use $m_e = 0.26m_0$, where m_0 is the true electron mass. Also, the conductivity effective mass for holes (m_h) can be obtained from the effective masses of the light (m_{h}) and heavy (m_{hh}) holes: $m_h = m_{hh}(1 + r^{3/2})/(r + r^{3/2})$, where $m_{hh} = 0.49m_0$ and $r = m_{hh}/m_{lh}$ [18]. The plasma model for the dielectric function takes into account absorption:

$$\epsilon(\omega) = \epsilon_{\infty} \{ 1 - \omega_{\rm pe}^2 / [\omega(\omega + i/\tau_{\rm e})] - \omega_{\rm ph}^2 [\omega(\omega + i/\tau_{\rm h})] \}.$$
(1)

The scattering times τ_e and τ_h are gotten from the carrier mobilities: $\tau_{e,h} = m_{e,h}\mu_{e,h/e}$ with $\mu_{e,h}$ derived from empirical models that take into account phonon and impurity scattering [19]. In Fig. 1 we plot the real and imaginary parts of Eq. (1) as function of the donor concentration N. The circular frequency has been fixed at $\omega = \omega_0 = 1.224 \times$ 10^{15} /s, corresponding to the emission wavelength of interest $\lambda_0 = 1.54 \,\mu\text{m}$. Re ϵ and Im ϵ change rapidly for $N \geq 1.54 \,\mu\text{m}$ 10^{19} /cm³; while Re ϵ decreases, Im ϵ increases with N. We are thus confronted with opposing demands. On the one hand, it is desirable to have N large enough for the plasma frequency to reach values $\omega_p(N) \leq \omega_0$ (see inset of Fig. 1) where the tunability is largest. On the other hand, we also wish to keep absorption in check. As a reasonable compromise, we limit the impurity density to the range $10^{19} \le N \le 10^{20}$ /cm³ where, for the upper limit, Im $\epsilon \simeq 0.0046$ Re ϵ . Henceforth, we will take into account only the real part of $\epsilon(N)$. We note that similar models were used for tuning the band structure and reflectance in 1D and 2D PCs [6,7]. For effective Bragg diffraction the period d should be of the same order as the wavelength $(d \sim$



Fig. 1. Real (left axis) and imaginary (right axis) part of dielectric function of Si as function of impurity concentration. The wavelength is fixed at $\lambda_0 = 1.54 \,\mu\text{m}$. The electron plasma frequency is shown in the inset. The arrow points at a hypothetical plasma frequency $2\pi c/\lambda_0$.

 $\lambda_0 \sim \lambda_p = 2\pi c/\omega_p$). We have chosen $d = 0.73 \ \mu\text{m}$ and layers of equal thickness. Using $\epsilon_1 = 2.09 \ [18]$ for the silica layers and ϵ_2 given by the *real part* of Eq. (1) for the silicon layers, we obtain the band structure shown in Fig. 2. At low frequencies ($\omega \leq \omega_p$), a forbidden gap just below the first band appears in the band structure. Because the lower edge of the first band approximately coincides with ω_p , this gap is associated with the fact that for $\omega \leq \omega_p$, Re $\epsilon < 0$ and light cannot propagate in the semiconductor. The first



Fig. 2. TE band structure of a one-dimensional photonic crystal with period $d = 0.73 \,\mu\text{m}$. Alternating Si and SiO₂ layers have equal widhts. Two values of the electron concentration are considered: $N = 10^{19}$ and $10^{20}/\text{cm}^3$. Gray areas correspond to allowed bands for the lower concentration. For the higher concentration, only the band edges are shown (dotted). In the inset, the lower band edges of the third band are amplified for both concentrations.

band is the most sensitive to change in N because $\omega \sim \omega_{\rm p}$. The PC considered was designed to give rise to an on-axis band edge around $\lambda = 1.54 \,\mu{\rm m}$, see the inset in Fig. 2. Thus, when $N = 10^{20}/{\rm cm}^3$ the lower edge of the third band is just above $\omega_0 = 1.224 \times 10^{15}/{\rm s}$ (corresponding to $\lambda_0 = 1.54 \,\mu{\rm m}$). For this frequency then, the *radiative* density of modes (with $k_{\parallel} < \omega/c$) vanishes and there can be no SE. On the other hand, if $N = 10^{19}/{\rm cm}^3$, the frequency $\omega = 1.224 \times 10^{15}$ is located within the third allowed band and, consequently, the radiative density of modes is finite and radiation *can* occur. In Ref. [20] it was found that the SE changes abruptly at the on-axis band edges if the radiating atom is located at an antinode of the modes.

Now we proceed to calculate the rate of SE of an atom hosted by the above described PC. Glauber and Lewenstein [21] have developed a modal quantum electrodynamical theory for the SE rate of an atom embedded in an inhomogeneous dielectric medium, characterized by an arbitrary, position-dependent dielectric function $\epsilon(\mathbf{r})$. A similar, classical theory, was derived by Dowling and Bowden [22], who replaced the two-level atom by a dipole (moment μ) oscillating with the frequency $\omega_0 = (E_2 - E_1)/\hbar$. It is important to observe that the two theories [21,22] are equivalent because the Wigner–Weisskopff approximation, namely weak coupling of the emitted light to the atom, was assumed in Ref. [21]. This, in fact, corresponds to the situation observed in most experiments of SE. The main result of Dowling and Bowden [22] is the radiated power

$$P_{\rm p} = \pi^2 \omega_0^2 \int d\mathbf{k} |\mathbf{a}_{\mathbf{k}p}(\mathbf{r_0}) \cdot \boldsymbol{\mu}|^2 \delta(\omega_{\mathbf{k}p} - \omega_0), \qquad (2)$$

where the index p stands for TE or TM polarization, $\omega_{\mathbf{k}p}$ are the frequency eigenvalues, $\mathbf{a}_{\mathbf{k}p}(\mathbf{r}_0)$ are the vector potential eigenvectors evaluated at the atom's position, the Dirac-delta function takes care of energy conservation, and the integration is over all normal modes for a given polarization. The SE rate is, simply, $\Gamma_p = P_p/\hbar\omega$.

Very recently, we have applied Eq. (2) to the calculation of the emission rate by an atom embedded in an (undoped) 1D PC [20,23]. Here we investigate the variation, with impurity concentration, of $\Gamma_{\rm p}$ for the Si/SiO₂ PC described above. In Fig. 3 we show the rate of emission as a function of the emitter's position for several carrier concentrations. The decay rate is normalized to the rate of emission in vacuum $(\Gamma/\Gamma_0 = P/P_0)$. It is seen that the tunability is considerably greater for an ion embedded in the SiO2 (white background), than it is for an ion in the Si (grey). For dipoles oriented parallel to the interfaces, the rate of emission into TE modes, Fig. 3(a), is very sensitive to N. When the donor concentration of Si is 10²⁰/cm³ there is no radiative mode because the lower edge of the third band is slightly above the frequency of the spontaneously emitted light. Thus, for this carrier concentration, the radiative contribution to the rate of emission vanishes for any position of the atom. Decreasing the carrier concentration, the rate of emission becomes finite when the lower edge of the third band shifts to frequencies that are below



Fig. 3. Radiative contribution to the rate of spontaneous emission by an Er ion (at $\lambda = 1.54 \,\mu\text{m}$) implanted in a Si/SiO₂ one-dimensional photonic crystal. The rate of emission is normalized with that in air and is plotted as function of the donor impurity concentration and of the ion's position in the SiO₂ (left) and Si (right) layers. (a) TE modes, (b) TM modes for dipole parallel to the interfaces, and (c) TM modes for dipole perpendicular to the interfaces.

 $1.224 \times 10^{15} \text{ s}^{-1}$ (see the behavior of the on-axis band edge near $\lambda = 1.54 \,\mu\text{m}$ in Fig. 2). This means that the emitted light can now couple to radiative modes. The emitter's position at which the rate of emission is most sensitive to changes in the carrier concentration is $s_0 = 0.25$ (at the middle of the SiO₂ layer). The antinodes of the electric-field at the lower band edges indeed coincide with the middle of the low-index layers, as found in Ref. [20]. If μ forms an angle ψ with the interface, then Γ plotted in Fig. 3(a) must be multiplied by $\cos^2 \psi$.

Consider an excited Er ion at the middle of a SiO₂ layer of a PC with $N = 10^{19}$ /cm³. It would radiate TE–polarized light at a rate $\Gamma \simeq 0.27\Gamma_0$. If, now, the carrier concentration is raised abruptly to 10^{20} /cm³, the radiative TE–polarized emission will be prohibited. It is notable that this *switching* occurs when the dielectric function of Si suffers a change of only 6% (see Fig. 1). On the other hand, a *gradual* variation of the free-electron concentration from 10^{19} to 9×10^{19} /cm³ would result in *tuning* of the SE. In contrast to the emission into radiative modes, the rate of emission into evanescent modes (not shown in Fig. 3) is much less sensitive to change in carrier concentration.

For TM polarization, two independent orientations (parallel and perpendicular to the interfaces) contribute to the SE. For dipoles parallel to the interfaces [$\psi = 0$, Fig. 3(b)], the radiative contribution to the SE presents considerable change when the carrier concentration varies from 10^{19} to $10^{20}/\text{cm}^3$. The radiative rate of emission,

however, never vanishes because the Brewster effect gives rise to closing of the band gaps for $0 < k_{\parallel} < \omega/c$. When the dipole is perpendicular to the interfaces $[\psi = \pi/2,$ Fig. 3(c)], the coupling of the light to the dipole is quite weak; therefore, the rate of emission is not very sensitive to changes in the carrier concentration. Also, the total rate of emission is discontinuous across the interfaces. This is a result of the discontinuity of the component of the electric field normal to the interfaces.

The explicit dependence of the emission rate on the impurity density is displayed in Fig. 4. In part (a) the emitter is positioned in the middle of a silica layer. If the dipole moment is parallel to the interfaces, we observe considerable tunability for both polarizations, and the TE component strictly vanishes for $N \cong 9.4 \times 10^{19}$ /cm³. The discontinuity in the slope $d\Gamma/dN$ at this concentration persists even for a uniform distribution of the donors in the SiO₂ layer, Fig. 4(b). Here we averaged over all the dipole positions and orientations and over the two polarizations.

In this paper we have calculated the rate of SE of an atom embedded in a tunable 1D PC. We designed it so that it has a band edge near the frequency of the spontaneously emitted light corresponding to the important wavelength $\lambda = 1.54 \,\mu\text{m}$. Therefore, we obtain considerable sensitivity of the radiative SE on the donor impurity density, especially when the emitter's position coincides with an antinode of the normal modes. The results presented are a demonstration, through an example, of the idea that it should be possible to intervene *actively* in the process of SE. As discussed in this Letter, in principle there are many



Fig. 4. Normalized rate of spontaneous emission by an Er ion as function of the impurity density in the layers. (a) The radiating ion is located at the center of SiO₂ the layer; note that for TE polarization $\Gamma = 0$ for $N \ge 9.4 \times 10^{19} / \text{cm}^3$. (b) Average rate of SE (per ion) for Er ions evenly distributed in the layer.

other possibilities for manipulating the relaxation of an atom via photon emission. We believe that dynamic alteration of SE is feasible experimentally. This could have important consequences for sources of light such as LEDs and lasers.

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