Spontaneous emission in the near field of two-dimensional photonic crystals

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We show theoretically that photonic crystal membranes cause large variations in the spontaneous emission rate of dipole emitters, not only inside but also in the near field above the membranes. Our three-dimensional finite-difference time-domain calculations reveal an inhibition of more than five times and an enhancement of more than ten times for the spontaneous emission rate of emitters with select dipole orientations and frequencies. Furthermore, we demonstrate theoretically the potential of a nanoscopic emitter attached to the end of a glass fiber tip as a local probe for mapping the large spatial variations of the photonic crystal local radiative density of states. This arrangement is promising for on-command modification of the coupling between an emitter and the photonic crystal in quantum optical experiments. © 2005 Optical Society of America

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It is well known that the rate of spontaneous emission can be controlled by the geometry of the medium surrounding the fluorescent species. In particular, many recent research efforts have been devoted to studying spontaneous emission in photonic crystals[1–3] (PCs). The quantitative interpretation of these experiments, however, remains frustrated by lack of detailed information about many parameters that strongly affect the emission dynamics. These include the exact position of the emitters on the subwavelength scale and the orientation of the emission dipole moments, as well as systematic effects such as surface-induced quenching[4] or other chemical or electronic surface phenomena. An ideal arrangement would require accurate placement of a single emitter at an arbitrary location in a PC. Very recently Badolato *et al.*[5] achieved this by precise fabrication of a PC structure around a given semiconductor emitter.[5] In this Letter we discuss the *in situ* control of the position and thereby modification of the spontaneous emission rate of a single emitter close to or in a two-dimensional PC slab.

Two-dimensional (2D) PCs fabricated in thin semiconductor membranes promise to achieve many of the long-standing goals of photonic bandgap materials. Indeed, recently it was demonstrated that it is possible to achieve very high-Q and low-mode-volume cavities in these structures.[6,7] Owing to their planar nature, PC membranes can be easily accessed by subwavelength probes such as optical fibers[8] or atomic force microscope tips.[9] Motivated by this opportunity, we investigate the prospects of coupling between a PC and nanoscopic optical emitters located at the end of sharp probes.[10–12] Although 2D crystals do not yield a zero density of states, we show that both inside and in the near field above a PC membrane the emission rate of properly oriented dipoles can be strongly modified. We show that the nanometer accuracy in scanning probe positioning allows the direct mapping of the dependence of the emission rate on the spatial coordinates of the subwavelength emitter.

We have used the 3D finite-difference time-domain method[13–15] to calculate the local radiative density of states (LRDOS), accounting for the position dependence of the photon states available for fluorescent decay of a quantum emitter.[16] This calculation relies on the fact that the LRDOS appearing in the formulation of Fermi's golden rule for the spontaneous emission rate also describes the total emitted power of a classical point-dipole antenna run at a fixed current.[13] We consider semiconductor membranes with dielectric constant $\epsilon = 11.76$ and thickness $d = 250$ nm, surrounded by up to 1 μm of air above and below. We take the membrane to contain a hexagonal array of holes with radius $r = 0.3a$ at a lattice spacing of $a = 420$ nm. Such a structure possesses a bandgap for a/λ in the range 0.25 to 0.33 for the transverse electric (TE) mode where the electric field is parallel to the plane of the membrane. The ratio a/λ is used as normalized frequency units throughout our work. We used discretization with 14 or 20 points per lattice constant and employed volume averaging of the

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dielectric constant to reduce staircasing errors.[9] We considered finite hexagonally shaped PC structures up to 25 holes across, terminated by the unperforated slab extending into Liao's absorbing boundary conditions. By broadband temporal excitation of the dipole, we simulated the emission power spectrum over a wide frequency range. After dividing the resulting spectrum by that of an identically excited dipole in vacuum, we obtained the LRDOS normalized to the vacuum LRDOS.[15]

For an emitter halfway deep in a PC membrane, the solid black spectrum in Fig. 1 shows a strong inhibition of fluorescence, by over a factor of 7, in the bandgap as compared with its vacuum rate. In this case the dipole was laterally centered in the structure, and its orientation was chosen to be in the x direction (see Fig. 1). The slab was taken to be $13a$ across, and we verified that no significant further reduction of the emission rate was obtained if we increased the size of the structure. However, the magnitude of the enhancement at the blue edge of the gap, as well as the Fabry–Perot oscillations at frequencies below the gap, depend on the finite size of the PC structure. For all tested structures wider than seven holes across, we find emission enhancements larger than a factor of 15, representing a jump of 2 orders of magnitude as compared with the LRDOS for frequencies in the gap.

Given the inherent strong modulation of the dielectric constant in a PC structure, it is particularly interesting to examine the lateral dependence of the spontaneous emission rate. Figure 2(A) shows a contour plot of the LRDOS modification for an x-oriented dipole midway in the slab depth versus emission frequency and for lateral locations along a trajectory that traces the irreducible part of the unit cell [Fig. 2(D)]. The emission is inhibited in the bandgap at all positions, whereas outside the gap we observe Fabry–Perot modulations together with enhancement at the low- and high-frequency edges. The enhancement of the emission occurs especially on the high-frequency edge of the gap (the air band) for dipoles in air holes and predominantly at the low-frequency edge (the dielectric band) for dipoles in the dielectric.

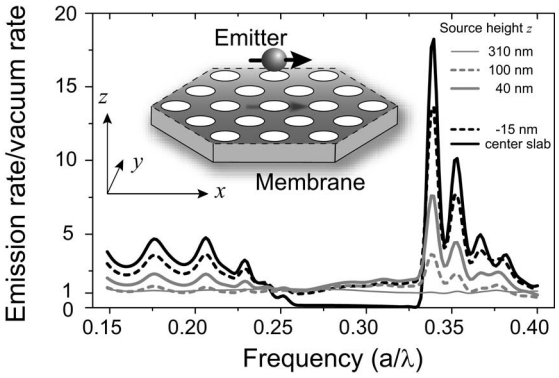

Fig. 1. Emission rate normalized to the vacuum rate versus frequency for an x-oriented dipole in the central hole of a PC membrane (details in text). Black spectra correspond to dipoles in the slab ($z<0$) and gray to dipoles above the slab ($z>0$), as listed in the legend.

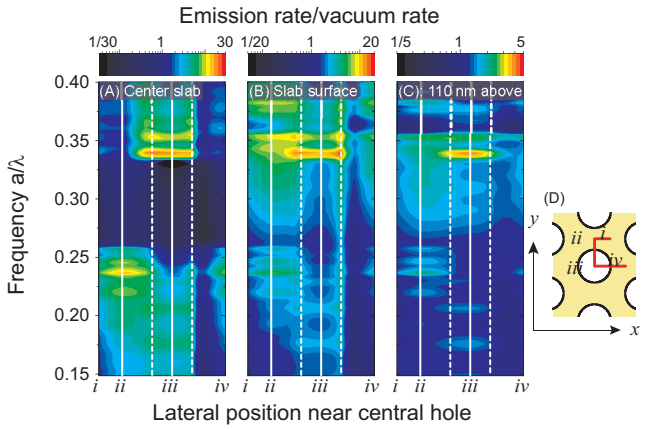

Fig. 2. Emission rate normalized to the vacuum rate for an x-oriented dipole (A) in the mid-depth, (B) on the surface, and (C) 110 nm above the PC membrane as a function of frequency and position along the trajectory indicated by the red line in (D). The trajectory traces the edges of the irreducible part of the unit cell. The dashed lines mark the borders between air hole and dielectric. The logarithmic color scales are shown on top.

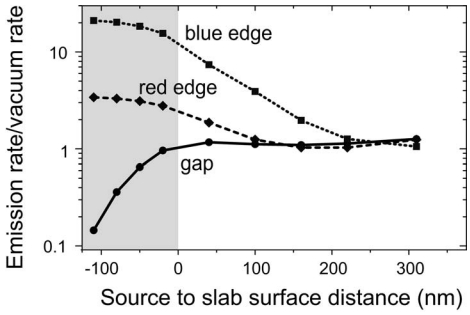

Fig. 3. Emission rate modification as a function of the height of a dipole above the PC membrane. Diamonds, circles, and squares show data for $a/\lambda=0.23,0.28,0.34$, corresponding to frequencies below, in, and above the gap, respectively. The shaded region shows the range of positions $-125<z<0$ nm in the membrane.

Next, we ask whether it is possible to capture these effects by scanning an emitter just above the PC slab. Different spectra in Fig. 1 show the LRDOS modification of a dipole laterally centered in the structure but at various heights z above the membrane. In addition, Figs. 2(B) and 2(C) display the modification of the LRDOS for the dipole right at the crystal–air interface and at 110 nm above this plane. These data reveal that, as z increases, the inhibition and enhancement are reduced in size. To examine this distance dependence more closely, in Fig. 3 we plot the normalized emission rate as a function of the distance between the dipole and the membrane surface for three key frequencies $a/\lambda=0.23,0.28,0.34$, just below, in, and just above the bandgap, respectively. Evidently the inhibition diminishes for emitters located above the slab. In contrast, enhancements persist at the blue edge of the gap even if the dipole is lifted into air above the membrane. Figures 2 and 3 let us conclude that it is possible to enhance the spontaneous emission rate by a factor of 5 to 10 if the emitter position is controlled to within 50 nm above the PC membrane. Note that the emission of a dipole

near a simple homogeneous dielectric slab is also enhanced due to coupling to the guided modes. However, at the gap edges the PC causes a further strong enhancement of the LRDOS.[17]

The required resolution and control for mapping the modification of the emission rate can be achieved by scanning a subwavelength emitter attached to the end of a sharp tip.[10–12] A crucial question arises as to the effect of the tip on the LRDOS. To estimate this effect, we have calculated the LRDOS for sources embedded inside cylindrical tips of diameter 125 nm pushed 130 nm into the central hole of the PC structure. We find that the influence of the PC structure on the spectrum of LRDOS enhancement and inhibition is unchanged for the system of emitters embedded in glass ($\epsilon \lesssim 2.25$). In contrast, tips of very high-index material such as silicon fundamentally change the LRDOS spectrum, leading to the creation of a low-Q localized defect mode from the band edge due to the addition of dielectric material.[9] The presence of a silicon tip causes an overall reduction of the gap depth and an increase and redshift of the rate enhancement at the blue edge of the gap. Suitable probes of the LRDOS in PC membranes are therefore emitters inside low-index tips.

Although optical detection of single emitters has become possible for some systems, many applications such as the realization of a nanolaser would benefit from coupling an ensemble of emitters to a PC. Furthermore, nanoscopic ensembles are more readily available than single emitter systems. Thus we have also considered the modification of the spontaneous emission rate for a subwavelength ensemble of randomly oriented dipoles. Note that, because the LRDOS is essentially unchanged for the TM polarization, dipolar components normal to the slab reduce the visibility of lifetime effects. We have considered over 25 symmetry inequivalent dipole orientations (corresponding to over 300 orientations in a 2π solid angle) in the central air hole and have calculated the corresponding LRDOS and luminescence extraction efficiency.[13,17,18] We find that in general the time-resolved flux of fluorescence photons extracted from the slab follows a significantly nonsingle exponential decay behavior. Nonetheless, the mean decay constant reveals inhibition by a factor of 3, and enhancement by a factor of 5, compared with vacuum. The observation of inhibition is facilitated by the increase of the emission extraction efficiency for in-plane dipoles from $\sim$20% for frequencies below the gap to $>$80% in the gap.[13,18]

In conclusion, we have shown that strong inhibition and enhancement of emission can be achieved for emitters well inside photonic crystal membranes, while a significant level of enhancement persists even in the near field above the structures. Since these results also hold for emitters embedded in nanoscopic dielectric probes, scanning probe technologies can be promising for on-command spontaneous emission control. An important advantage of emitters inside such probes is that they are shielded from unwanted interactions and can be calibrated by simply retracting the probe from the structure.

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