

Comment on “Spontaneous Emission of Organic Molecules Embedded in a Photonic Crystal”

A recent Letter of Petrov *et al.* [1] addresses the modification of the spontaneous emission of luminescent molecules embedded in a photonic crystal. They report both accelerated and inhibited spontaneous emission, with a factor of ~ 2 difference evident between the decay components. The central point of their Letter is that the photonic crystal (i.e., its dielectric contrast) is responsible for this effect. We show below why this “photonic band gap effect” cannot be responsible for their observations.

The photonic crystal of Petrov *et al.* was formed by impregnating an artificial opal of porous silica globules with a polymer (PMMA) containing a fluorescent dye. The ratio of the dielectric constants of the porous silica and the PMMA is 1.3, indicative of a photonic crystal that weakly interacts with light. Indeed, a narrow stop band in the fluorescence emission spectrum of about 6% relative width (FWHM) is observed [their Fig. 3(b)]. The inhibition of spontaneous emission should then be at most $\sim 10\%$, as estimated by the fraction of solid angle covered by such stop bands—but only for measurements performed at the blue edge of the stop band. The solid angle covered is largest here. This may be seen using a simple Bragg diffraction analogy, that is independent of the network topology: as the wavelength decreases from the red to the blue edge of the stop band, the Bragg condition is satisfied for increasingly larger cones of angles about normal incidence. (A further decrease in wavelength opens up a hole in the middle of the cone because the Bragg condition is no longer satisfied at normal incidence.) Surprisingly, Petrov *et al.* report measurements at a wavelength of 510 nm, which is at the red edge of the stop band, where they should not see any inhibition.

A more sophisticated estimate with both accelerated (at the red edge) and inhibited (at the blue edge) spontaneous emission follows from photon density of states (DOS) calculations, such as those by Busch and John [2]. Calculations for an opal with a much greater dielectric contrast (2.1 instead of 1.3; see Fig. 4 of Ref. [2]) give a maximum difference between accelerated and inhibited decay components of only 16%, not the near factor of 2 difference demonstrated in Fig. 4 of Ref. [1].

It is not necessary to consider the more refined local DOS [3] since its variation in these weakly photonic crystals is negligible. The results presented by Petrov *et al.* would require an inconsistently large variation in the LDOS, such as is found for strongly photonic crystals of dielectric contrast ~ 12 [2,4].

We have studied photonic crystals similar to that investigated by Petrov *et al.* and have observed no such photonic effects, as anticipated. Our system consists of dye-doped silica spheres in water, and has a dielectric contrast of 1.2.

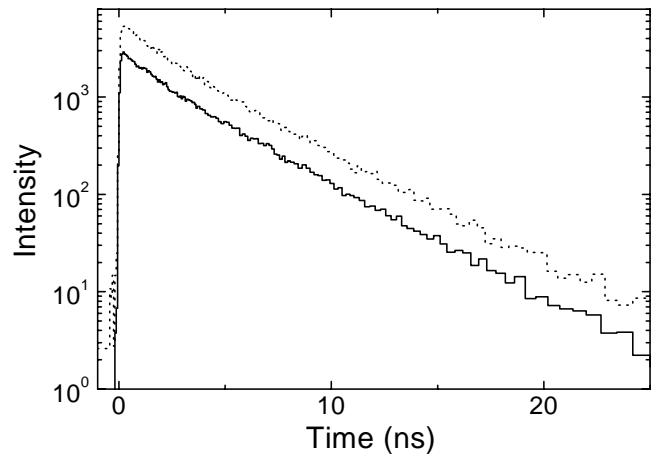


FIG. 1. Time-resolved fluorescence of dye in a photonic crystal (solid curve, average decay time $\tau = 3.50 \pm 0.03$ ns) and in an equivalent reference system (dashed curve, $\tau = 3.57$ ns). The similarity of the two curves, offset for clarity, demonstrates their experimentally indistinguishable decay kinetics.

The dye is covalently attached to the silica spheres and covered by a silica layer. The chief advantage of our system is the ability to confidently compare the crystal with a reference system consisting of the same spheres in a random arrangement. This means that we can distinguish photonic effects on the emission rate from other effects, such as chemical interactions between the dye and the environment. In Fig. 1, we show the time-resolved fluorescence of dye molecules within the photonic crystal and the reference, taken at wavelengths near the blue edge of a stop band. Regrettably, similar raw data are not shown in Ref. [1]. Observe the close similarity between the decay kinetics of the two curves in Fig. 1, extending over more than three decades in intensity. We must conclude, both for theoretical and experimental reasons, that the modification of fluorescence decay times reported by Petrov *et al.* cannot be attributed to photonic effects.

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Received 30 December 1998

PACS numbers: 42.70.Qs, 42.50.-p

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