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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 \( \sim 2 \) 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 PMMA is observed [their Fig. 3(b)]. The inhibition of 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 evident between the decay components of only 16%, not the near factor of 2 difference reported by Petrov et al. [1].

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 \( \sim 12 \) [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.

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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