Controlling spontaneous emission with metamaterials

M. A. Noginov,[1,*] H. Li,[1] Yu. A. Barnakov,[1] D. Dryden,[2] G. Nataraj,[2] G. Zhu,[1]
C. E. Bonner,[1] M. Mayy,[1] Z. Jacob,[3] and E. E. Narimanov[3]

[1]*Center for Materials Research, Norfolk State University, Norfolk, Virginia 23504, USA*

[2]*Summer Research Program, Center for Materials Research, Norfolk State University, Norfolk, Virginia 23504, USA*

[3]*School of Electrical Engineering, Purdue University, West Lafayette, Indiana 47907, USA*

**Corresponding author: mnoginov@nsu.edu*

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We have observed, in metamaterial with hyperbolic dispersion (an array of silver nanowires in alumina membrane), a sixfold reduction of the emission lifetime of dye deposited onto the metamaterial's surface. This serves as evidence of an anomalously high density of photonic states in hyperbolic metamaterials, demonstrates the feasibility of an earlier-predicted single-photon gun, and paves the road for the use of metamaterials in quantum optics.

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Metamaterials are engineered composite materials containing subwavelength inclusions (meta-"atoms") that allow for custom-tailored electromagnetic response of the medium, leading to an entirely new class of phenomena, ranging form negative index and high-resolution imaging to enhanced quantum-electrodynamic effects [1,2] and "cloaked" systems with reduced visibility [3,4].

In particular, metamaterials with hyperbolic dispersion [5–7], in which real parts of the elements of the dielectric tensor have different signs, enable the negative refraction and hyperbolic [8,9]. These hyperbolic materials are furthermore predicted to have broadband "supersingularity" [10] of photonic density of states, which causes enhanced and highly directional spontaneous emission and enables a variety of devices with new functionalities, including a single-photon gun [10].

In this Letter, we experimentally study the effect of the hyperbolic supersingularity on the spontaneous emission lifetime of dye deposited on a hyperbolic metamaterial. The experimental results described below are found to be in qualitative agreement with the theoretical predictions [10]. The enhancement of the spontaneous emission rates that we observe is consistent with the general concept of ultrahigh Purcell factors in metamaterials, originally introduced in [1,2], where it was illustrated on the example of slow light waveguides.

Alumina membranes with the dimensions 1 cm× 1 cm × 51 μm, acquired from Synkera, Inc., had 35 nm channels (voids) extending through the whole thickness of the membrane perpendicular to its surface. The surface filling factor of the voids was ~15%. The membranes were filled with silver, following the method described in [7]. The film of polymethyl methacrylate (PMMA) doped with IR-140 laser dye at concentration 0.013 M has been deposited on the top of the silver-filled membrane; see Fig. 1.

To determine the values of the electric permittivity in the directions parallel, $\varepsilon_{\|}$, and perpendicular, $\varepsilon_{\perp}$, to the membrane's surface, we have experimentally measured angular dependences of the reflectance in both p and s polarizations [at $\lambda = 873$ nm—Fig. 2] and fitted them with the known formulas [7]. The details of the experimental method are discussed in [5,7]. The material has exhibited a hyperbolic dispersion with the effective values of permittivity ($\varepsilon_{\|} = 4.99 + i0.22$, $\varepsilon_{\perp} = -0.15 + i1.07$) close to those in [7], confirming a good reproducibility of the fabrication technique. The experimental results are in agreement with the model predictions [11] for the silver filling factor ~8%, which is smaller than the nominal filling factor by ~15%. The latter discrepancy can be explained by imperfect electroplating or the difference between the properties of bulk silver [12] and silver deposited into the membrane. The same model predicts that, at the wavelength of the maximum emission of dye (described below), the real part of $\varepsilon_{\perp}$ is equal to −0.3. (Other real and imaginary components of $\varepsilon_{\|}$ and $\varepsilon_{\perp}$ exhibit smaller dispersion.) Note that the low value of the imaginary part of $\varepsilon_{\|}$ is in agreement with the sample's relatively high transmittance at normal incidence, $T = 0.12$.

The emission band of the IR-140 laser dye has its maximum at 892 nm and a FWHM equal to 37 nm [13]. The absorption band has the peak at 820 nm and an FWHM equal to 90 nm. Control samples included dye-doped polymeric films deposited onto pure glass, a neutral-density glass filter with the absorption coefficient ~50 cm^{-1}, a pure

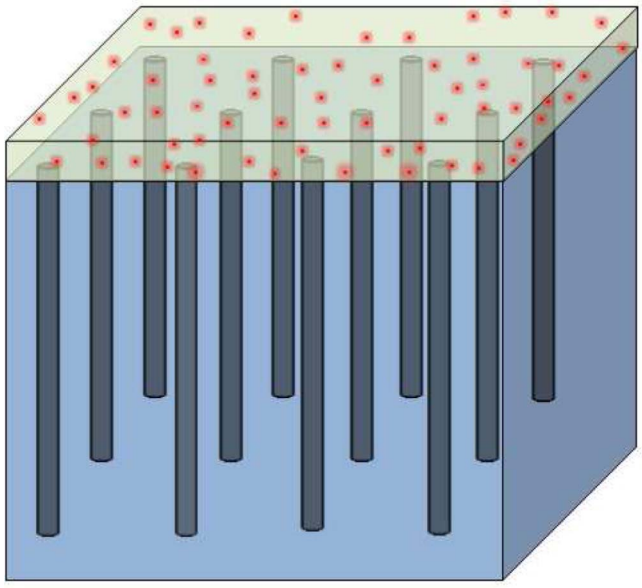

Fig. 1. (Color online) Schematic of the sample: alumina membrane with circular silver nanowires (a hyperbolic metamaterial) coated with dye-doped polymeric film.

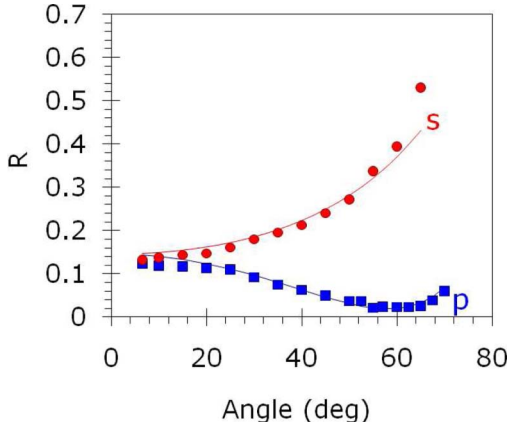

Fig. 2. (Color online) Angular dependence of the reflectance R in s and p polarizations at $\lambda = 873$ nm. Solid lines—calculation according to Eq. (1) of [6].

(unfilled) membrane, and silver and gold thin films on glass. The thickness of the dye-doped polymer film on the silver-filled membrane, measured with the Dektak 6 profilometer, was equal to ~81 nm. The samples were excited at $\lambda = 800$ nm with 100 fs pulses of the mode-locked Ti-sapphire laser (Mira-900), and the emission was detected with the C5680 streak camera. Because of the wide-open entrance slit, the time resolution of the streak camera was ~30 ps. The emission was separated from pumping by the 810 nm long-pass filter.

The emission kinetics in the dye-doped film deposited onto the unfilled membrane was a nearly single exponential, with the decay-time equal to 760 ps (Fig. 3). Similar exponential decays were observed in all other control samples. The emission kinetics in some of the control samples, e.g., dye-doped PMMA film deposited on gold, had a short peak overlapping with the pumping pulse; see Fig. 3. This peak (also observed in some substrates without dye-doped films) is a combination of the pumping light leaking through the filter as well as the luminescence and nonlinear optical response of the substrate to 100 fs laser pulses. This short pulse is not of interest to the present study, and we shall concentrate on the emission kinetics, which follow it. In the film deposited onto the silver-filled membrane, the emission decay time of dye (averaged over many measurements taken in different parts of the sample) was as short as ~125 ps (Fig. 3) in qualitative agreement with the theoretical predictions [10].

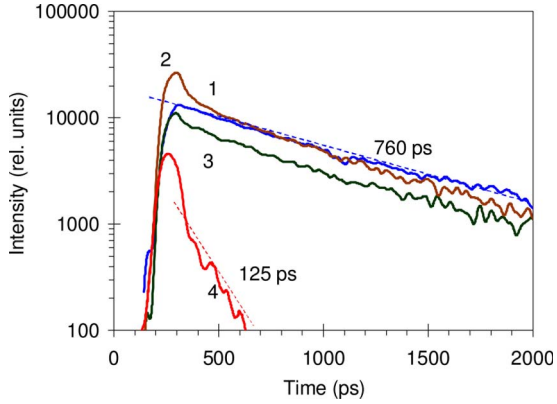

Fig. 3. (Color online) Emission kinetics in the IR140/PMMA films deposited on (1) the top of pure alumina membrane, (2) gold film on glass, (3) silver film on glass, and (4) silver-filled alumina membrane.

The shortening of the emission decay-time could not be due to nonradiative luminescence quenching on silver inclusions (the surface filling factor was ~15%), because no quenching was observed in the dye-doped sample deposited onto the silver film; see Fig. 3.

The high photonic density of states in a hyperbolic metamaterial gives rise to preferential and enhanced spontaneous emission into the membrane. At the same time, the probability of photon emission to an opposite hemisphere is being reduced [10]. Correspondingly, the emission intensity, detected in our experiment from the same side of the sample from which it is illuminated, is expected to be reduced. This prediction is in an agreement with the experimental observation; see Fig. 3.

A rigorous quantum-electrodynamic approach for analyzing the problem of spontaneous emission in the vicinity of lossy, dispersive metamaterial structures was developed in [1,2]. A related semiclassical model was subsequently used in the case of hyperbolic metamaterials, as described in [10]. The experimentally observed lifetime shortening is stronger than the one calculated based on this semiclassical approach, which predicts a 1.8-fold reduction of the effective spontaneous emission lifetime (Fig. 4). In the simulation, we used the effective parameters of the system described above and calculated emission lifetimes of dipoles randomly oriented and uniformly distributed within 80-nm-thick PMMA film.

The discrepancy can be even larger if the dye in the polymer (without metal or metamaterial) has a quantum yield significantly smaller than unity. The difference can be explained by a variety of reasons (which will be the subject of an elaborate future study), including the strong dispersion of the metamaterial within the bandwidth of the dye, overestimation of the thickness of the dye-doped polymeric film, and nonlocal response of the nanorod metamaterial, which leads to additional waves [14].

To summarize, in the example of alumina membrane embedded with silver nanowires, we have proven the

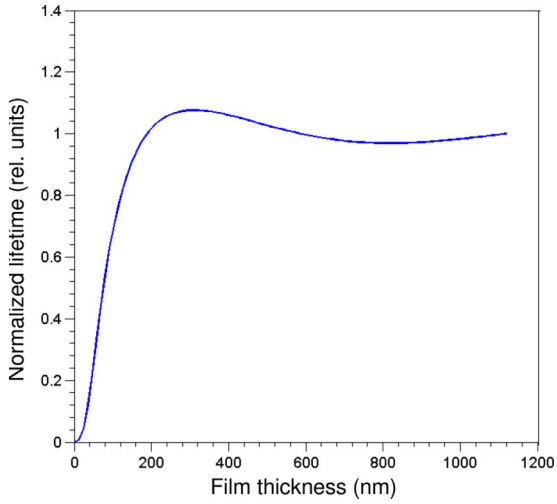

Fig. 4. (Color online) Calculated effective emission lifetimes of dipoles randomly oriented and uniformly distributed within PMMA film on the top of the silver-filled alumina membrane (normalized to the emission lifetime in pure PMMA).

theoretically predicted anomalously high photonic density of states in hyperbolic metamaterials. The demonstrated possibility to control the spontaneous emission with metamaterials paves the road for many exciting applications, including a single-photon gun needed in quantum optics and information technology [10].

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