Control of Emission and Reflectance with Metamaterials

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Abstract

We experimentally show that a broad-band singularity in the density of photonic states in metamaterials with hyperbolic dispersion enables two classes of interesting physical phenomena: (i) enhancement of spontaneous emission radiative rate, quantum yield and directionality, and (ii) strong reduction of reflectance off corrugated metamaterials surfaces.

1. Introduction

Metamaterials – engineered composite materials with rationally designed subwavelength inclusions – have much stronger responses to electromagnetic waves than any conventional media. Fascinating theoretical predictions and experimental realizations of metamaterials' applications and devices include negative index media, optical cloaking, and sub-diffraction imaging and focusing. Metamaterials with hyperbolic dispersion [1-3], in which different elements of the dielectric tensor have different signs, are predicted to have broad-band singularity of photonic density of states, which causes enhanced and highly directional spontaneous emission [4] as well as strong increase of light scattering from defects and metamaterials' corrugated surfaces, with nearly all incident light "sucked" into propagating modes of a hyperbolic medium [5].

In this work, the two classes of phenomena have been studied experimentally. The control of spontaneous emission with metamaterials has been demonstrated in arrays of silver nanowires embedded in alumina membrane and in lamellar metal-dielectric multilayered structures. In another particular experiment, a strong reduction of reflectance off corrugated metamaterials surfaces has been demonstrated in silver-filled alumina membranes.

2. Emission control in arrays of silver nanowires

Alumina membranes with the dimensions 1cm x 1cm x 51μm had 35 nm channels (voids) extending through the whole thickness of the membrane perpendicular to its surface. The membranes were filled with silver, following the method described in Ref. [3]. This synthesis technique results in a metamaterial with hyperbolic dispersion (negative electric permittivity in the direction perpendicular to the membrane's surface, $\varepsilon_\perp$, and positive electric permittivity in the direction parallel to the membrane, $\varepsilon_\parallel$) in the near-infrared range of the spectrum [3]. From the measurements of the angular dependence of reflectance in s and p polarizations (similar to traces 1 and 2 in Fig. 1a) we have deduced the following materials parameters at λ=873 nm: $\varepsilon_\perp'$=-0.15, $\varepsilon_\perp''$=1.07, $\varepsilon_\parallel'$=4.99, and $\varepsilon_\parallel''$=0.02.

In the experiments, IR 140 laser dye-doped PMMA polymeric films have been deposited on the top of silver-filled membranes. The emission band of the IR140 dye overlaps with the spectral range of the hyperbolic dispersion of the metamaterial. Control samples included dye-doped polymeric films deposited onto pure (unfilled) membrane, and silver and gold thin films on glass. The film thickness was approximately equal to 130 nm. The samples were excited at λ=800 nm with 100 fs pulses of the

mode-locked Ti-sapphire laser and the emission was detected with the IR streak camera. The emission was separated from pumping by the 810 nm long-pass filter.
The emission kinetics in the dye-doped film deposited onto unfilled membrane was nearly single-exponential, with the decay-time equal to 760 ps, Fig. 1b. The emission kinetics in some of the other control samples, *e.g.* gold film, were shortened to ~700 ps. In the film deposited onto the silver-filled membrane, the emission decay-time of dye was as short as ~125 ps, Fig. 1b, in a qualitative agreement with the theoretical predictions [4]. The shortening of the emission decay-time could not be due to non-radiative luminescence quenching on silver inclusions, since no quenching of comparable magnitude was observed in the samples deposited onto metallic films. Since the emission kinetics shortening is explained by the enhancement of emission irradiated *inside* the metamaterial sample, the emission intensity, detected from the same side of the sample from which it is illuminated, is expected to get reduced. This prediction is in an agreement with the experimental observation, Fig. 1b.

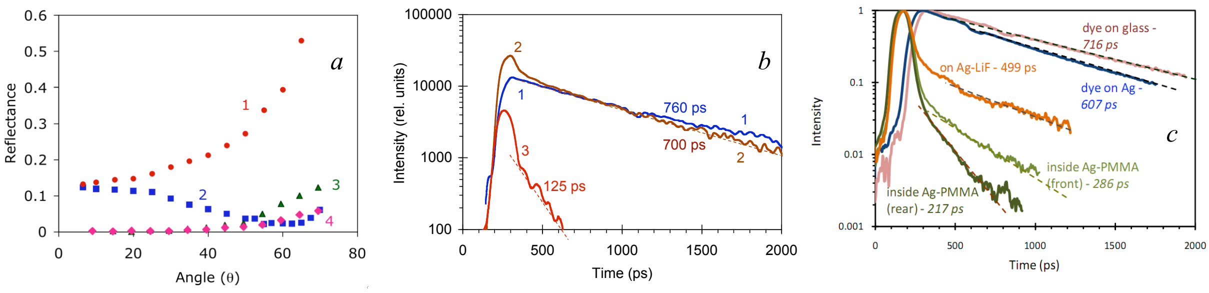

Fig. 1. (a) Angular reflectance profiles measured on untreated (1,2) and roughened (3,4) parts of the same membrane sample in s-polarization (1,3) and p-polarization (2,4). (b) Emission kinetics in the IR140/PMMA films deposited on the (1) top of pure alumina membrane, (2) gold film on glass and (3) silver-filled alumina membrane. (c) Emission kinetics of the IR-140 dye in the silver-PMMA lamellar metamaterial, compared with the emission kinetics of the same dye deposited on the top of Ag-LiF metamaterial (14 alternating layers) and on the top of glass and silver substrates.

3. Emission control in lamellar structures

Three different types of metal-dielectric lamellar structures, consisting of organic and inorganic dielectric components have been fabricated. The first type consisted of 20 alternating layers of silver and IR-140 dye-doped PMMA thin films, deposited using thermal evaporation and spin coating respectively. For comparison, we have fabricated a similar Ag-PMMA structure without dye doping. Alternative all-inorganic lamellar structures (Ag-LiF and Ag-MgF_2) have been produced by thermal evaporation. A layer of dye-doped PMMA (30 nm) was deposited on top of undoped metamaterial samples. All multilayered structures have demonstrated hyperbolic dispersion, which was evaluated based on the measurements of angular reflectance profiles [3].
In qualitative agreement with the theoretical predictions [4], Ag/dye-doped PMMA multilayered structure showed a strong reduction of the fluorescence lifetime, down to 286 ps when the sample was illuminated from the front and 217 ps when it was illuminated from the rear. For comparison, the emission life-times of the same dye deposited onto a glass substrate and silver film were equal 716 ps and 607 ps, respectively (Fig. 1c). Expectedly, the fluorescence lifetime reduction in the films deposited on the top of *undoped* metamaterials (including Ag-LiF, Ag-MgF_2 and Ag-PMMA), was considerably smaller, as in the case of Ag-LiF, which had a fluorescence lifetime equal to 499 ps (Fig. 1c).

4. Reduction of reflectance in corrugated silver-filled alumina membranes

The hyperbolic metamaterial used in this study (silver-filled alumina membrane) was the same as described in Section 2 above. The vertical roughness of untreated sample was measured with the atomic force microscope (AFM) to be equal to rms=40 nm and the lateral correlation length was equal to ~100 nm.

Then, the surface of the sample was corrugated by grinding it with Al_2O_3 polishing powder, with the vertical roughness of the corrugated sample equal to 600 nm and the lateral correlation length equal to ~1.17 μm. The reflectance measurements were repeated in the corrugated sample, resulting in much smaller intensities of reflected light, especially at small incidence angles, traces 3 and 4 in Fig. 1a.
As corrugated samples produce not only specular reflection but also diffused scattering of an incident light, we have evaluated the ratio of the reflected and scattered light intensities by measuring optical signal with a large-aperture detector (1 inch diameter) in the direction of the reflected beam at different distances from the sample. The most conservative estimate (assuming Lambertian scattering distribution diagram) have shown that the ratio of scattering and specular light intensities is much smaller than the difference between the reflectance in corrugated and not corrugated samples, Fig.1a.

5. Conclusions

To summarize, we have proven that spontaneous emission of dye molecules can be controlled by metamaterials with hyperbolic dispersion, which possess broad-band singularities in the density of photonic states. Both the reduction of radiative live-times (and corresponding increase of the luminescence quantum yield) and the directionality of emission (evaluated indirectly based on the emission intensities), which have been demonstrated experimentally, pave the road to many exciting applications including a single photon gun needed for quantum optics and information technology [4]. We have also demonstrated that the reflectance of a hyperbolic metamaterial is significantly reduced upon corrugation of its surface, in an agreement with the theoretical prediction [5]. With the original concept equally applicable to all parts of the electromagnetic spectrum, our result thus opens an entirely new route towards radiation-absorbing materials and surfaces, with wide range of applications from solar light harvesting to radar stealth technology.

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