Radiative Decay Engineering with Hyperbolic Metamaterials

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Abstract: We demonstrate the decrease in the spontaneous emission lifetime of dye molecules due to the enhanced photonic density of states (PDOS) of a hyperbolic metamaterial (HMM), opening the route to PDOS engineered HMM devices.

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The photonic density of states (PDOS), like its electronic counterpart, is the key physical quantity governing a variety of phenomena and hence PDOS manipulation is the route to new photonic devices. The DOS can be altered significantly by using microcavities [1], photonic crystals [2], surface plasmons [3] or slow light waveguides [4]. Here, we demonstrate how a unique dielectric response can enhance PDOS for bulk propagating waves within a medium, opening new vistas in PDOS engineering based on metamaterials.

Metamaterials with hyperbolic dispersion, were recently shown to have a unique broadband singularity in the photonic density of states, unlike any other conventional photonic system [5]. The HMM allows propagating high wavevector spatial modes which decay exponentially in an isotropic medium (eg:vacuum) and these modes contribute to the density of states causing a divergence [5,6]. Spontanous emission which depends markedly on the available photonic density of states was predicted to be enhanced in the vicinity of the metamaterial with hyperbolic dispersion [5]. Here we demonstrate a hyperbolic metamaterial substrate that can alter the radiative decay rate and radically modify the spatial distribution of the spontaneous emission from emitters placed above it. The spontaneous emission is enhanced in the vicinity of the hyperbolic metamaterial due to the larger photonic density of states compared to vacuum, causing a decrease in the radiative lifetime of emitters such as quantum dots or molecules. Such a substrate can lead to devices for increased sensitivity fluorescence detection and other biochemical applications.

The hyperbolic metamaterial is designed (Fig. 1(a)) to operate at the wavelength of emission of the chosen dye, Rhodamine 800. Twelve alternating subwavelength layers of alumina and gold (filling factor = 0.5) are deposited using an electron beam evaporation technique with careful crystal monitor control of the individual layer thicknesses. Effective medium theory predicts that the dielectric permittivity of this metamaterial structure, in the direction parallel and perpendicular to the layers are of opposite signs achieving hyperbolic dispersion in a broad bandwidth [6,7,8].

To confirm the effective medium calculations and hyperbolic dispersion at the wavelength of interest, the effective dielectric tensor is extracted from the reflectivity data for s and p polarized incident light (Fig. 1(b)) using a $\chi 2$ fitting method . The result of a multidimensional search ensures hyperbolic dispersion as shown in Fig. 1(c).

Emission characteristics are studied using the Rhodamine 800 ($\lambda_{peak} = 715$ nm) dye in an epoxy matrix (n=1.4) placed over the metamaterial. The thickness of the dye layer is 30 nm ascertained by profilometry using the alpha step profilometer. To characterise the role of quenching and non radiative decay rates, we compare the results with a metal substrate (thick layer of gold) and an alumina substrate. The lifetime measurements are carried out using MicroTime 200 flouorescence lifetime imaging (FLIM) system. The dye is excited using a pump laser (λ= 617 nm) with 100 fs pulses. The dye molecules placed on metamaterial show a distinct decrease in lifetime as compared to either the dielectric or metal.

The lifetime decrease is caused by the continuum of high wavevector spatial modes emitted by the dye in the close vicinity of the metamaterial which cannot propagate in vacuum or dielectric. In the case of metal, these high spatial modes are simply absorbed, giving rise to non radiative decrease in the lifetime (quenching) [8,9]. The hyperbolic dispersion in the metamaterial allows propagating modes with large wavevectors which cause a radiative decrease in the lifetime of the dye. Lifetime simulations using a semiclassical approach based on a many dipole model are in agreement with the experimentally measured lifetimes. Moreover, our simulations show that the imaginary part of the permittivities which have been ascertained with high accuracy do not cause any substantial non radiative decrease as compared to the metal.

In conclusion, we have proposed a metamaterial structure which enhances the spontaneous emission rate of an emitter placed above it. This is in direct agreement to the predicted effect arising due to an enhanced photonic density of states in hyperbolic metamaterials. The road to PDOS engineered devices using hyperbolic metamaterials will require structures to outcouple high-k photons and also optimization to overcome losses for practical device applications.

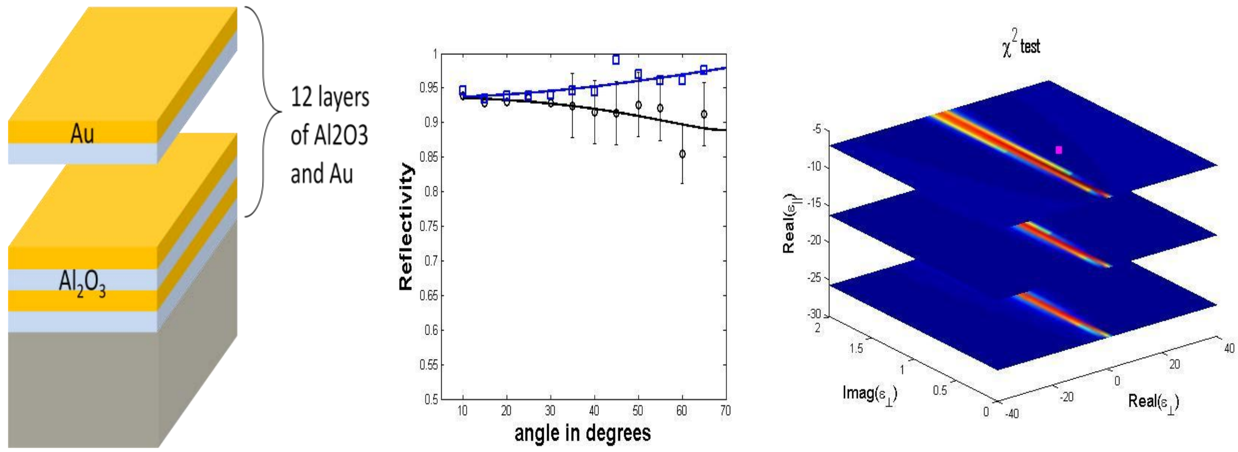

Fig. 1 (a) Metamaterial structure consisting of 12 alternating subwavelength layers of gold and alumina (fill fraction = 0.5) to achieve hyperbolic dispersion at the dye emission wavelength of 715 nm. The epoxy layer mixed with dye (rhodamine) is placed on top of the metamaterial and the light emitted by the dye, reflected from the metamaterial is collected by the FLIM system (b) Reflectivity vs angle for s polarized and p polarized light incident on the metamateria. (blue squares: experimental data for s-pol and black circles: experimental data for p-pol) The solid lines denote the corresponding fit according to the effective medium model. Error bars of 5 % have been shown on only few points for clarity (c) The fitting is done by searching for the effective parameters that minimize χ^2. Blue corresponds to regions with low χ^2 and red corresponds to regions of high χ^2. The pink dot corresponds to the absolute minimum in this multidimensional search. The retrieved parameters are, parallel to the layers $\varepsilon_{parallel}$ = -7+0.7i and perpendicular to the layers $\varepsilon_{perpendicular}$ = 7.3 + 0.9i

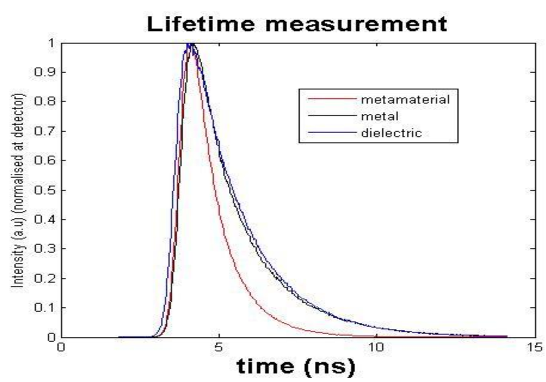

Fig. 2 (a) Lifetime measurement results. The metamaterial decay (red) shows a clear decrease in lifetime by a factor of 1.8 as compared to the metal (black) and the dielectric substrates (blue). Simulation results taking into account the quantum yield of the dye and distributed nature of the dipoles show a factor of ~1.7 . The intensity is normalized at the detector for better collection efficiency of the photons. In the case of the metamaterial, the photons are preferentially emitted into the metamaterial due to the availablility of high 'k' propagating modes.

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