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Broadband Purcell effect: Radiative decay engineering with metamaterials

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We show that metamaterials with hyperbolic dispersion support a large number of electromagnetic states that can couple to quantum emitters leading to a broadband Purcell effect. The proposed approach of radiative decay engineering, useful for applications such as single photon sources, fluorescence imaging, biosensing, and single molecule detection, also opens up the possibility of using hyperbolic metamaterials to probe the spontaneous emission properties of atoms and artificial atoms such as quantum dots. © *2012 American Institute of Physics*.
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Microcavities and photonic crystals are among the most promising systems for enhancing spontaneous emission by the Purcell effect[1] and to simultaneously collect the emitted photon in a given quantum state.[2,4] They form the test bed for cavity quantum electrodynamics experiments and aid the major advances in single photon sources. However, the high quality of the resonance required for the cavity Purcell effect immediately puts a restriction on the spectral width of the emitter and hence on the possible compatible sources. For example, the reduced linewidth of quantum dots at low temperatures which is ideally compatible with a microcavity for the demonstration of the Purcell effect is too wide at room temperatures—making such systems unviable for Purcell enhancement.[3] Moreover, other emitters such as molecules and nitrogen vacancy centers in diamond have broad bandwidth emission not ideally compatible with cavity technology, and therefore an alternative, non-cavity based approach is needed.[4] The resulting recent interest towards systems which show a broadband Purcell effect[5–8] opened up the route to a number of applications from broadband single photon sources[9,10] to strong coupling of emitters to plasmons.[11,12] In particular, we proposed[13] that the large number of electromagnetic states of a hyperbolic metamaterial leads to a divergence in the photonic density of states allowing broadband control over light matter interaction at room temperature. As opposed to conventional methods based on closed cavity Purcell enhancement of spontaneous emission[4] or open cavity systems based on photonic crystal waveguides,[14] our approach relies on a radiative decay engineering approach using metamaterials,[15] engineering the dielectric repsonse of the medium surrounding the emitter to provide extra electromagnetic states for in-coupling. This effect has been demonstrated experimentally,[16,17] and substantial enhancement was observed in the spontaneous emission rates of organic dyes and quantum dots when positioned near the surface of a metamaterial with hyperbolic dispersion. In this paper, we expand on our original theoretical prediction of this effect[13] and present a quantitative description of the spontaneous emission rate enhancement.

Metamaterials with hyperbolic dispersion, also known as indefinite media,[18] lie at the heart of devices such as the hyperlens[19,20] and non-magnetic negative index waveguides.[21] In an isotropic medium, the dispersion relation $\frac{k^2}{\varepsilon} = \frac{\omega^2}{c^2}$ defines a spherical iso-frequency surface in the k-space (see Fig. 1(a)), thus placing an upper cut-off for the wavenumber so that high wavevector modes simply decay away. In contrast to this behavior, a strongly anisotropic metamaterial where the the components of the dielectric permittivity tensor have opposite signs in two ortogonal directions can support bulk propagating waves with unbounded wavevectors. This can be most clearly seen in the case of uniaxial anisotropy ($\varepsilon_z \equiv \varepsilon_\parallel, \varepsilon_x = \varepsilon_y \equiv \varepsilon_\perp$) where the dispersion relation for the extraordinary (TM-polarized) waves $\frac{k_\parallel^2}{\varepsilon_\perp} + \frac{k_\perp^2}{\varepsilon_\parallel} = \frac{\omega^2}{c^2}$ for $\varepsilon_\parallel \varepsilon_\perp < 0$ describes a hyperboloid of revolution around the symmetry axis z (see Fig. 1(b)) and thus does not limit the

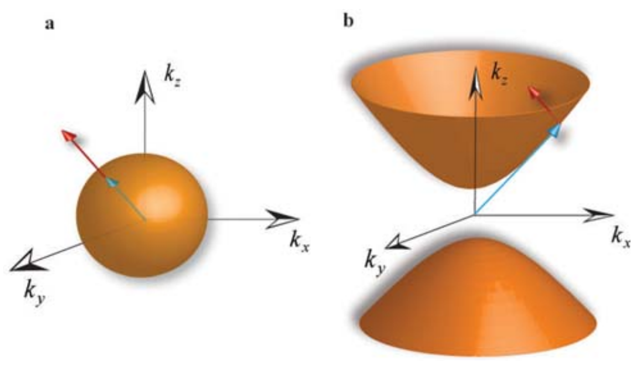

FIG. 1. (a) Dispersion relation for an isotropic medium. The blue arrow denotes an allowed wavevector whereas the normal to the dispersion relation gives the direction of the group velocity (red arrow). (b) Hyperbolic dispersion relation allowing large number of electromagnetic states with unbounded values of the wavevector (blue arrow). The group velocity vectors (red arrow) lie within a cone which implies light propagation in such media is inherently directional.

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magnitude of the wavenumber. Such high-k propagating modes allow for subwavelength imaging[22] and subdiffraction mode confinement.[23] As we demonstrate in the present paper, these high wavenumber spatial modes in hyperbolic metamaterials also have a strong effect on spontaneous emission.

Using the Fermi's golden rule, an increased number of radiative decay channels due to the high-k states in hyperbolic media, available for an excited atom, ensure enhanced spontaneous emission. This can increase the quantum yield by overcoming emission into competing non-radiative decay routes such as phonons. A decrease in lifetime, high quantum yield, and good collection efficiency can lead to extraction of single photons reliably at a high repetition rate from isolated emitters.[4]

We consider the classic example of radiative decay engineering using a substrate which interacts with an emitter placed above it.[24,25] We emphasize that the Purcell effect can be understood from classical dipole radiation theory.[26] In Fig. 2(a) we plot the corresponding emission decay relaxation time as a function of the distance to the sample, for a hyperbolic metamaterial with the dielectric permittivity tensor $\varepsilon_x = 1.2 + 0.1i, \varepsilon_y = 1.2 + 0.1i, \varepsilon_z = -4 + 0.1i$. In agreement with the qualitative arguments above, in the close vicinity of the substrate the availability of the large number of photonic states causes the photons to be preferentially emitted into the metamaterial, and the lifetime decreases considerably (Fig. 2(a) inset shows the coupling efficiency in the near field). Even though the emitter is placed in vacuum and is coupled to the quasi continuum of vacuum states, the large number of states in the metamaterial leads to a Purcell effect without the need for confinement. The decay rate is limited by the patterning scale "a" of the metamaterial. For distances, $d \ll a$ the effective medium approximation is not valid for a dipole in the near field since the interaction is dominated by waves with wavectors $k > (1/a)$.

The available radiative channels for the spontaneous photon emission consist of the propagating waves in vacuum, the plasmon on the metamaterial substrate, and the continuum of high wavevector waves which are evanescent in vacuum but propagating within the metamaterial. The corresponding decay rate into the metamaterial modes when the emitter is at a distance $a < d \ll \lambda$ (where a is the metamaterial patterning scale) is

$$\Gamma_{total} = \Gamma_{rad} + \Gamma_{non-rad} = \Gamma_0 \frac{\sqrt{\varepsilon_x|\varepsilon_z|}}{2(k_0 d)^3(1+\varepsilon_x|\varepsilon_z|)} + \varepsilon''\Gamma_0 \frac{\varepsilon_x - |\varepsilon_z|}{4(k_0 d)^3(1+\varepsilon_x|\varepsilon_z|)^2}$$

where the dielectric constants of the metamaterial are given by $\varepsilon_{||} = \varepsilon_x + i\varepsilon''$, $\varepsilon_\perp = -|\varepsilon_z| + i\varepsilon''$, Γ_0 is the free space decay rate, and k_0 is the free space wavevector.[31]

Along with the reduction in lifetime and high efficiency of emission into the metamaterial, another key feature of the hyperbolic media is the directional nature of light propagation. Fig. 2(b) shows the field along a plane perpendicular to the metamaterial-vacuum interface exhibiting the beamlike radiation from a point dipole. This is advantageous from the point of view of collection efficiency of light since the spontaneous emitted photons lie within a cone.[27] The group velocity vectors in the medium which point in the direction of the Poynting vector are simply normals to the dispersion curve [Fig. 1]. For vacuum, these normals point in all directions, and hence the spontaneous emission is isotropic in nature. In contrast to this behavior, the hyperbolic dispersion medium allows wavevectors only within a narrow region defined by the asymptotes of the hyperbola. Hence the group velocity vectors lie within the resonance cone giving rise to a directional spontaneously emitted photon propagating within the metamaterial. The angle of the beam depends on the dielectric constants of the metamaterial and is given by $\tan(\theta) = \sqrt{\varepsilon_x/|\varepsilon_z|}$. The light can be confined to a subdiffraction beam when $|\varepsilon_z| \gg \varepsilon_x$. The beamlike nature of the photon in the metamaterial arising solely due to the hyperbolic dispersion has to be distinguished from that obtained by the mode properties of a resonant structure such as a micropost microcavity[4] or that of a guided mode in a photonic crystal waveguide.[14,28]

As described in Ref. 29, the actual realization of the hyperbolic metamaterial introduces deviations from the effective medium description. This effect can be significant especially when the dipole is placed at distances closer than the wavelength of emission. The high-k waves predicted by effective medium theory are the bloch modes due to coupled plasmonic states of the multilayer metal-dielectric structure. The equivalence is exact in the limit of a0 (a, unit cell size). Here we consider a practical realization of a hyperbolic metamaterial consisting of alternate layers of silver ($\varepsilon_{Ag} = -1.82 + 0.59i$) and alumina ($\varepsilon_{Al_2O_3} = 2.7$) at a wavelength of $\lambda \approx 350$ nm. The system consists of 8 layers, each

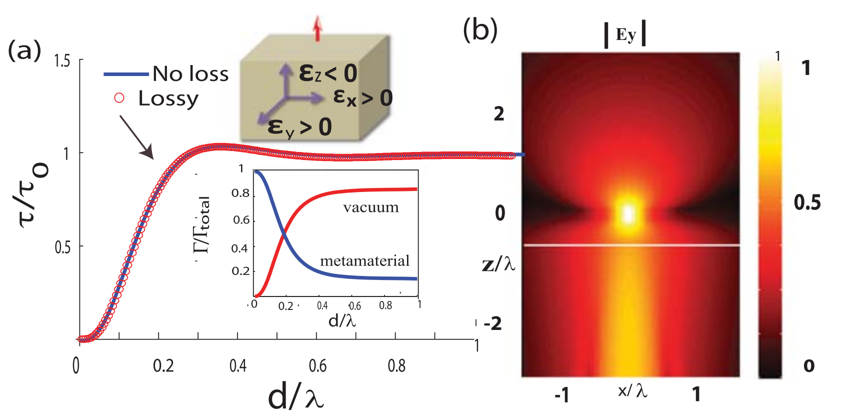

FIG. 2. (a) Spontaneous emission lifetime of a perpendicular dipole above a hyperbolic metamaterial substrate (see inset). Note the substantial lifetime decrease in the close vicinity of the metamaterial. The inset shows the coupling efficiency of the light to the metamaterial modes with distance. Almost all the light couples to the metamaterial modes in the near field (inset). (b) False color plot of the field of the point dipole in a plane perpendicular to the metamaterial-vacuum interface (see inset of (a)) depicting the highly directional nature of the spontaneous emission (resonance cone).

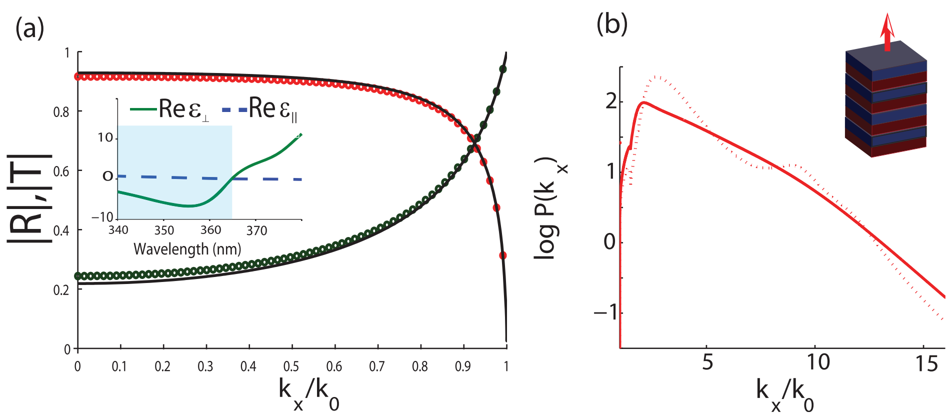

FIG. 3. (a) Comparison of the reflection and transmission amplitudes of plane waves incident on the planar multilayer realization of the hyperbolic metamaterial and effective medium theory. The metamaterial system consists of 8 alternating subwavelength layers of Ag/Al_2O_3. Green and red circles correspond to reflection and transmission computed using transfer matrix methods, and the black superimposed line is calculated from effective medium theory. (inset) Hyperbolic dispersion is achieved in a broad bandwidth around $\lambda = 350$ nm as the dielectric constants are of opposite signs in perpendicular directions. (b) The power spectrum of the dipole at a distance of $d = \lambda/10$ from the layered structure for different absorption in the metal. Most of the spontaneous emission occurs into high wavevector states which propagate within the h-MM. The decay rate and coupling efficiency (related to the area under the curve) varies little with the absorption (dotted curve is for low absorption in the metal).

of thickness $a = 8$ nm which is easily achievable by current fabrication techniques. We compute and compare the propagating wave spectrum which is routinely used in ellipsometric measurements for extraction of effective medium parameters. Fig. 3(a) shows the plane wave reflection and transmission coefficients computed using transfer matrix techniques in the layered realization superimposed on the effective medium prediction. Since $a \ll \lambda$, effective medium theory holds in a broad bandwidth (Fig. 3(a) inset). The effective permittivity of the multilayer structure is $\varepsilon_x = 0.44 + 0.29$ and $\varepsilon_z = -6.03 + 7.66i$. The power spectrum of the waves emitted by the dipole is shown in Fig. 3(b) which elucidates the role of large wavevector waves that carry energy within the metamaterial. In the close vicinity of the hyperbolic metamaterial, the power from the dipole is completely concentrated in the large spatial wavevector channels. The same evanescent wave spectrum when incident on a lossy metal or dielectric would be completely absorbed, causing a non-radiative decrease in the lifetime of an emitter (quenching). On the contrary, the metamaterial converts the evanescent waves to propagating, and the absorption thus affects the outcoupling efficiency of the emitted photons due to a finite propagation length in the metamaterial. The coupling efficiency and Purcell effect is 92% and 10, respectively, at a distance of $\lambda/10$. This is also the case in the low loss limit (dotted line) which shows that absorption mainly affects the propagation length of the metamaterial modes. Near the wavelength region when the parallel component of the dielectric permittivity is zero, an optimum lifetime decrease is obtained which is lower than a thin film of metal by a factor of 2.

Effective medium theory shows that the bandwidth of the silver alumina multilayer system is 20 nm near $\lambda = 360$ nm, whereas the bandwidth of a silver titanium dioxide system will be 40 nm around $\lambda = 400$ nm. A silver nanowire array in a dielectric host also acts as a hyperbolic metamaterial and can have a broad bandwidth of over 200 nm at $\lambda = 750$ nm. The Purcell factor is only limited by how close the dipole can be placed to the metamaterial structure before nonradiative processes dominate or the effective medium theory breaks down. Furthermore, very high Purcell factors can be obtained by placing emitters within the metamaterial due to increased coupling. The high-k metamaterial states do not propagate in vacuum; hence, gratings or a curvilinear metamaterial geometry like the hyperlens would be needed to outcouple the resonance cones.

In conclusion, we have shown that the electromagnetic states of a hyperbolic metamaterial lead to a non-resonant broadband Purcell effect. The proposed device based on hyperbolic metamaterials is compatible with a wide variety of sources and capable of room temperature operation due to the broad bandwidth enhancement of spontaneous emission and directional photon emission. Our work paves the way for using metamaterials for applications in quantum nanophotonics ranging from single photon sources to fluorescence based sensing.[30]

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