Broadband Purcell effect: Radiative decay engineering with metamaterials

Zubin Jacob, 1* Igor Smolyaninov, 2 Evgenii Narimanov 1

1 Birck Nanotechnology Center, School of Electrical and Computer engineering, Purdue University, West Lafayette, IN 47907, U.S.A.,
2 Department of Electrical and Computer Engineering, University of Maryland, College Park, MD 20742, U.S.A.

* To whom correspondence should be addressed; E-mail: zjacob@purdue.edu.

Engineering the photonic density of states (PDOS) using resonant microcavities or periodic dielectric media gives control over a plethora of classical and quantum phenomena associated with light. Here, we show that nanostructured metamaterials with hyperbolic dispersion possess a broad bandwidth singularity in the PDOS, an effect not present in any other photonic system, which allows remarkable control over light-matter interactions. A spectacular manifestation of this non-resonant PDOS alteration is the broadband Purcell effect, an enhancement in the spontaneous emission of a light source, which ultimately leads to a device that can efficiently harness a single photon from an isolated emitter. Our approach differs from conventional resonant Purcell effect routes to single photon sources with a limitation in bandwidth, which places restrictions on the probable use of such methods for practical device applications, especially at room temperature. The proposed ‘metadevice’, useful
for applications from quantum communications to biosensing also opens up the possibility of using metamaterials to probe the quantum electrodynamic properties of atoms and artificial atoms such as quantum dots.
The design and engineering of a single photon state is central to quantum communication networks, quantum key distribution protocols and various cavity QED experiments (1, 2, 6, 7). Semiconductor based single photon sources which efficiently couple to optoelectronic devices can significantly impact existing communication systems. Availability of single photons also helps to shed light on wave-particle dualities as well as fundamental issues of quantum measurement and uncertainty (3). While single emitters such as atoms or artificial atoms like quantum dots show single photon emission, the efficiency and quantum yield need to be improved considerably to construct an efficient single photon device. Moreover, the low light collection efficiency due to the isotropic nature of the spontaneous emission is a serious limitation to construct practical devices.

Increasing the PDOS is the key to harnessing the spontaneous emission from emitters which have a low quantum yield (1). Fermi’s golden rule immediately shows that a higher photonic density of states can overcome emission into competing non-radiative decay routes such as phonons. Apart from increasing the quantum yield, the issue of timing jitter inherent in the decay of the excited state can be reduced significantly by decreasing the spontaneous emission lifetime (4). Achieving a high repetition rate for the single photon device not limited by the intrinsic lifetime also requires enhancement of spontaneous emission (5).

Microcavities are among the most promising approaches for enhancing spontaneous emission by the Purcell effect and to simultaneously collect the emitted photon in a given quantum state (6). They form the test bed for cavity quantum electrodynamics experiments and aid the major advances in single photon sources (7). 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 decreased linewidth of quantum dots at low temperatures which are ideally compatible with microcavities for the demonstration of the Purcell effect are too wide at room temperatures making them unviable for Purcell en-
hancement (8, 9). Moreover, other single photon emitters such as molecules (10) and nitrogen vacancy centers in diamond (11) still await a cavity technology capable of enhancing their broad bandwidth emission.

Apart from isolated emitters for single photon sources, the issue of a broadband Purcell effect is pertinent to emission from a large number of sources as in the case of LEDs. At room temperature, the inhomogeneous broadening of emitters, such as a collection of quantum dots severely limit the cavity approach to Purcell enhancement. Such a broadband Purcell effect can lead to high efficiency, high frequency LEDs (1).

In this paper, we show that metamaterials hold the answer for a broadband Purcell effect which allows control over light matter interaction at room temperature. The proposed meta-device not only enhances the spontaneous emission of an emitter such as a quantum dot, but also causes the photons to be emitted in a preferential direction leading to an efficient single photon gun. As opposed to conventional methods based on closed cavity Purcell enhancement of spontaneous emission (6, 7) or open cavity systems based on photonic crystal waveguides (12), our approach relies on a completely new approach, engineering the dielectric response of the medium surrounding the emitter to provide dramatically increased density of photonic states. A high PDOS immediately translates to a larger number of radiative decay channels available for an excited atom ensuring enhanced spontaneous emission, a necessary quality to construct efficient single photon sources from isolated emitters.

The broad bandwidth effect arises due to the non resonant nature of the singularity in the PDOS of propagating waves in a hyperbolic dispersion metamaterial. This unique form of the dispersion relation occurs in non-magnetic strongly anisotropic metamaterials which have dielectric permittivities of opposite signs in two perpendicular directions (13). These hyperbolic metamaterials, also known as indefinite media (?), can support propagating waves with large wavevectors and lies at the heart of devices such as the hyperlens (14–17) and non-magnetic
negative refraction materials (19, 20). These high wavevector modes would have ordinarily decayed away in vacuum due to the circular dispersion relation which is bounded (14). The unbounded hyperbolic form of the dispersion relation, ensures a large number of available ‘k’ states for the emitted photons from a source. Furthermore, the hyperbolic dispersion in strongly anisotropic materials also gives rise to photons in a preferred direction thus increasing the collection efficiency of the single photon emitter (21).

The photonic density of states is a quantity of interest that controls a variety of phenomena. Using a microcavity can enhance it at resonance frequencies, or a periodic dielectric medium like a photonic crystal can cause a bandgap, totally suppressing the available PDOS (1). In vacuum, the PDOS is simply related to the dispersion relation where the allowed states within the spherical shell lying between frequencies \( \omega \) and \( \omega + d\omega \) determine the DOS (Fig. 1(a)). In contrast, a metamaterial with hyperbolic dispersion allows spatial modes with unbounded wavevectors causing a divergence in the density of states (Fig. 1(b)). Finite losses (\( \text{Im}(\epsilon) = \delta \ll 1 \)) will cut off the singularity at \( k_{\text{max}} \sim k_0/\delta \) leading to a density of states (see the Supplementary Information)

\[
\rho_{\text{meta}}(\omega) \sim \frac{\omega^2}{c^3 \delta^3} \gg \rho_{\text{vacuum}}(\omega)
\]  

Furthermore, hyperbolic dispersion can be achieved in a relatively wide spectral range so the expected divergent behaviour has broad bandwidth. Being able to engineer the PDOS in a broad bandwidth presents a paradigm shift from conventional resonant approaches and can lead to a variety of interesting device applications. We emphasize that the singularity occurs for bulk propagating waves in the metamaterial without the need for coupling to a waveguide mode or a counter propagating wave as in a resonator. As we will show below, this strong singularity in the PDOS circumvents the need for three dimensional confinement of the emitter to achieve the Purcell effect.
This large density of states considerably alters the rate of spontaneous emission. We use the semiclassical approach to calculate the emission rate of a point dipole (dipole moment $\mu$) placed inside the hyperbolic metamaterial with $\epsilon = \text{diag}(\epsilon_x, \epsilon_y, \epsilon_z)$ where $\epsilon_x = \epsilon_y > 0$ and $\epsilon_z < 0$. The decay rate of a point dipole diverges as (see the Supplementary Information)

$$\Gamma^{rad} = \frac{\mu^2 \omega^3 |\epsilon_z|^2}{4 \epsilon^3 \hbar} \int_0^\pi \frac{\sin^3 \theta d\theta}{[(\epsilon_x \cos^2 \theta + \text{Re}(\epsilon_z) \sin^2 \theta)^2 + \delta^2 \sin^4 \theta]^{5/4}} \sim \delta^{-3/2}$$

This divergent behavior of the decay rate is counterintuitive to the conventional response of materials where the decay rate goes to a constant value when losses can be neglected. The nanopatterning scale of the hyperbolic metamaterial or the losses which currently hinder most metamaterial devices will ultimately limit the highest allowed wavevector and finally the decay rate of the emitter.

Though the hyperbolic metamaterial can significantly enhance spontaneous emission, accessing the interior of a bulk hyperbolic dispersion medium would require highly stringent fabrication conditions. We therefore consider the classic example of radiative decay engineering using a substrate which interacts with an emitter placed above it (24). A metamaterial substrate with hyperbolic dispersion can dramatically reduce the spontaneous emission lifetime of an emitter due to the above mentioned singularity in density of states. The calculated spontaneous emission lifetime variation with distance is shown in Fig. 2(a). In the close vicinity of the substrate, the availability of the large number of photonic states causes the photon to be preferentially emitted into the metamaterial and the lifetime decreases considerably. Even though the emitter is placed in vacuum and is coupled to the quasi continuum of vacuum states, the strong PDOS singularity in the metamaterial leads to a Purcell effect without the need for confinement. The available channels for the photon consist of the propagating waves in vacuum, the plasmon on the metamaterial substrate and the the continuum of high wavevector waves which are evanescent in vacuum but propagating within the metamaterial. The anisotropy can
be chosen so as to completely suppress the plasmonic channel (25) and enhance the efficiency of emission into the desired metamaterial modes. The corresponding decay rate into the metamaterial modes when the emitter is at a distance \( d \ll \lambda \) is (see the Supplementary Information)

\[
\Gamma_{\text{meta}} \approx \frac{\mu^2}{8\hbar d^3} \frac{2\sqrt{\left|\epsilon_x\right|\left|\epsilon_z\right|}}{(1 + \epsilon_x\left|\epsilon_z\right|)} \tag{3}
\]

In the close vicinity of the hyperbolic metamaterial, the power from the dipole is completely concentrated in the large spatial wavevector channels (Fig 2(b)). 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 evanscient waves to propagating and the absorption thus affects the outcoupling efficiency of the emitted photons due to a finite propagation length in the metamaterial. Taking into account losses and current metamaterial fabrication technologies the efficiency of emission into the propagating waves within the metamaterial can exceed \( \eta \approx 80\% \), with a Purcell factor of approximately \( F_p = 5 \) (inset of Fig 2(b)).

Another key feature of the hyperbolic metamaterial along with the reduction in lifetime and high efficiency of emission into the metamaterial is the directional nature of light propagation in hyperbolic dispersion media (21). Fig. 2(c) 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 the single photon source since the spontaneous emitted photon lie within a cone (23). The group velocity vectors in the medium which point in the direction of the Poynting vector are simply normals to the dispersion curve. For vacuum, these normals point in all directions and hence the spontaneous emission is isotropic in nature. In contrast, 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 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 property of a resonant structure like a micropost microcavity \((7)\) or that of a guided mode in a photonic crystal waveguide \((12)\).

To extract these photons efficiently it is necessary to construct a metamaterial structure which performs the transformation from high spatial wavectors to those that can propagate in a medium such as vacuum. We achieve an outcoupling efficiency of \(\eta_{\text{out}} \approx 20\%\) by adding curvature to the metamaterial as shown in Fig. 2(d). Conservation of angular momentum in this cylindrical geometry causes the emitted photon within the metamaterial to couple to spatial modes propagating in vacuum. One realisation of the single photon meta-device is by using alternating curved subwavelength layers of metal and dielectric as in the hyperlens \((14)\) (Fig. 2(d)). Current fabrication technologies can achieve this leading to the above mentioned large density of photonic states as well as directional radiation. The broad bandwidth hyperbolic dispersion metamaterial which comprises the metadevice has been demonstrated in bulk with a high figure of merit at various wavelengths of interest and are among the most promising metamaterials for device applications due to their relative ease of fabrication \((17, 19, 20)\).

The metamaterial single photon metadevice as proposed in this paper should find immediate applicability for quantum key distribution protocols such as BB84 \((26, 27)\) wherein the figure of merit is the antibunching of photons and a high efficiency. The ultimate goal of a room temperature, broadband source providing single photons with quantum indistinguishability and high efficiency will require outcoupling structures such as hypergratings \((28)\).

In conclusion, we have uncovered a broadband singularity in the PDOS of hyperbolic metamaterials which is the route to photonic devices with new functionalities. This unique broadband manipulation of the PDOS unlike conventional methods leads to a non-resonant Purcell effect. The proposed single photon 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.

References and Notes

1. B. Lounis and M. Orrit, *Rep. Prog. Phys.*, **68**, 1129 (2005).

2. C.H. Bennet and G. Brassard, *IEEE International Conference on Computers, Systems and Signal Processing* (IEEE, Bangalore, India, 1984).

3. M. Scully, K. Druhl, *Phys. Rev. A.* **25**, 2208, (1982).

4. C. Becher, A. Kiraz, P. Michler, W. V. Schoenfeld, P. M. Petroff, Lidong Zhang, E. Hu, A. Imamoglu, *Physica E*, **13**, 412 (2002).

5. S. Strauf, N. G. Stoltz, M. T. Rakher, L. A. Coldren, P. M. Petroff and D Bouwmeester, *Nat. Photon.*, **1**, 704, (2007).

6. E. Moreau, I. Robert, J. M. Gerard, I. Abram, L. Manin, and V. Thierry-Mieg, *Appl. Phys. Lett.* **79**, 2865 (2001).

7. M. Pelton, C. Santori, J. Vuckovic, B. Zhang, G. Solomon, J. Plant and Y. Yamamato, *Phys. Rev. Lett.*, **89**, 233602 (2002).

8. T. Baba and D. Sano, *IEEE J. Sel. Top. Quantum Electron.*, **9**, 5, (2003).

9. P. Michler, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse and S. K. Buratto, *Nature*, **406**, 968 (2000).

10. Th. Basche, W. E. Moerner, M. Orrit and H. Talon, *Phys. Rev. Lett.*, **69**, 1516, (1992).

11. C. Kurtsiefer, S. Mayer, P. Zarda and H. Weinfurter, *Phys. Rev. Lett.*, **85**, 290, (2000).
12. S. Hughes, *Opt. Lett.*, 29, 2659 (2004).

13. V. A. Podolskiy, and E. E. Narimanov, *Phys. Rev. B* 71, 201101 (2005).

14. Z. Jacob, L. V. Alekseyev, and E. Narimanov, *Opt. Exp.* 14, 8247-8256 (2006)

15. A. Salandrino, N. Engheta, *Phys. Rev. B* 74, 075103 (2006).

16. I. Smolyaninov, Y. Hung, and C. Davis, *Science* 315, 1699-1701, (2007).

17. Z. Liu, H. Lee, Y. Xiong, C. Sun, X. Zhang, *Science* 315, 1686, (2007).

18. Z. Jacob and E. E. Narimanov, *Appl. Phys. Lett.*, 93, 221109 (2008).

19. A. J. Hoffman, L. Alekseyev, S. S. Howard, K. J. Franz, D. Wasserman, V. A. Podolskiy, E. E. Narimanov, D. L. Sivco and C. Gmachl, *Nat. Mat.*, 6, 946 (2007).

20. M. A. Noginov, Yu. A. Barnakov, G. Zhu, T. Tumkur, H. Li, and E. E. Narimanov, *Appl. Phys. Lett.*, 94, 151105 (2009).

21. Z. Jacob, L. V. Alekseyev, and E. Narimanov, *J. Opt. Soc. Am. A* 24, A52-A59(2007).

22. G. W. Ford and W. H. Weber, *Phys. Reports*, 113, 197, (1984).

23. E. Arbel and L. B. Felsen in *Electromagnetic Theory and Antennas* edited by E. C. Jordan (Pergamon Press, New York, 1963).

24. K. H. Drexhage, *J. Lumin* 693, (1970).

25. A. V. Akimov, A. Mukherjee, C. L. Yu, D. E. Chang, A. S. Zibrov, P. R. Hemmer, H. Park, and M. D. Lukin, *Nature*, 450, 402, (2007)

26. J. Vuckovic, D. Fattal, C. Santori, G. Solomon and Y. Yamamoto, *Appl. Phys. Lett.*, 82, 3596, (2003).
27. E. Waks, C. Santori and Y. Yamamoto, *Phys. Rev. A*. **66**, 042315 (2002).

28. S. Thongrattanasiri and V. A. Podolskiy, *Opt. Lett.*, **34**, 890 (2009).
Figure 1: (A) Dispersion relation for an isotropic medium such as vacuum. The blue arrow denotes an allowed wavevector, whereas the normal to the dispersion relation gives the direction of the group velocity (red arrow). The photonic density of states is related to the volume of a shell bounded between two such spheres. (B) Hyperbolic dispersion relation allowing unbounded values of the wavevector (blue arrow) due to which the photonic density of states diverges. The group velocity vectors (red arrow) lie within a cone which implies light propagation in such media is inherently directional.
Figure 2: (A) Spontaneous emission lifetime of a perpendicular dipole above a metamaterial substrate (see inset). Note the lifetime goes to zero in the close vicinity of the metamaterial as the photons are emitted instantly. (B) Most of the power emitted by the dipole is concentrated in the large spatial modes which are converted to propagating waves within the metamaterial. (inset) Contribution of the vacuum spatial modes and metamaterial spatial modes to the lifetime of the dipole. (C) Plot of the field in a plane perpendicular to the metamaterial vacuum interface depicting the highly directional nature of the spontaneous emission. (D) Curved structure consisting of alternating subwavelength layers of metal and dielectric to achieve hyperbolic dispersion and simultaneously outcouple the photon with large spatial wavevectors into vacuum. (inset) Outcoupled field for the cylindrical hyperlens consisting of alternate layers of silver and alumina (14).