Broadband enhancement of spontaneous emission from nitrogen-vacancy centers in nanodiamonds by hyperbolic metamaterials

M. Y. Shalaginov[1], S. Ishii[1,2], J. Liu[1], A. V. Kildishev[1], and V. M. Shalaev[1]
[1]*School of Electrical and Computer Engineering, Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47907, USA*
[2]*Advanced ICT Research Institute, National Institute of Information and Communications Technology, 651-2401, Kobe, Japan*
Author e-mail address: shalaginov@purdue.edu

Abstract: We experimentally demonstrate a broadband enhancement of emission from nitrogen-vacancy centers in nanodiamonds. The enhancement is achieved by using a multilayer metamaterial with hyperbolic dispersion.
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Generation of single photons is of vital importance for the development of quantum computation and communication technologies which will allow information to be transmitted and processed much faster and more securely [1, 2]. Stable and bright single photon sources hold the key to the development of these technologies. Though there have been many single photon sources reported, nitrogen-vacancy (NV) centers in diamonds have gained significant attention in the recent past due to their stability, broad-bandwidth, and room temperature operation [3]. The major bottleneck with diamond NV centres is that the photon emission rate is low. Many techniques have been proposed in the past to enhance the emission rate of diamond NV centers using resonant photonic structures [4, 5, 6]. Unfortunately, these structures are typically narrowband and counteract the broadband advantage of diamond NV centers. Coupling NV centers to metamaterials with hyperbolic dispersion opens up a possibility to engineer spontaneous emission efficiency of single-photon emitters in a broad spectral range.

According to Fermi's golden rule, the rate of spontaneous emission is directly related to photonic density of states (PDOS). The anomalously large PDOS in a broad spectral range is a unique property of hyperbolic metamaterials (HMMs) that can make a difference in emission efficiency of quantum emitters [7, 8]. In this work, multilayered HMM was fabricated as a stack of 16 alternating layers of gold (Au) and alumina (Al_2O_3) with each being 19 nm thick. The films were deposited on a coverslip using electron-beam evaporation. Effective dielectric functions (Fig. 1a) retrieved from spectroscopic ellipsometry (V-VASE, J.A. Woollam Co.), show opposite signs for perpendicular ($\mathrm{Re}\,\varepsilon_{\perp}$) and parallel ($\mathrm{Re}\,\varepsilon_{\parallel}$) components in the range of wavelengths longer than 550 nm, leading to hyperbolic dispersion.

Nanodiamonds (NDs) with median size of 35 nm in highly diluted aqueous suspension were spin-coated onto the surface of the HMM covered with a 30-nm thick SU-8 spacer layer (Fig. 1b). The spacer layer was introduced to avoid photoluminescence quenching by a metallic surface. The average number of NV centers per crystal is around ten [9]. Figure 1c shows the broadband emission from NV centers in the vicinity of the metamaterial surface.

The lifetimes of NV centers in NDs deposited on a glass substrate (Fig. 1d) and HMM (Fig. 1e) were measured using time-resolved confocal microscopy (time-correlated single photon counting method). The mean value of lifetime (inverse of the total decay rate) for the HMM sample and the glass sample is 1.55 ns and 20.89 ns, respectively, which is a factor of 13 reduction. However, the ratio of the radiative decay rates for the same samples has changed only by a factor of 3. Relative change in the radiative decay rates was evaluated using the reference method [8]. It can be considered that the non-radiative decay rates are substantially increased when a ND is placed in the vicinity of the HMM. One of the solutions to overcome this problem is to build an HMM using plasmonic materials with lower losses.

In summary, we have experimentally demonstrated the enhancement of spontaneous emission from NV centers over a broad range of wavelengths by using gold-alumina multilayer HMM. The achieved enhancement of the radiative decay rate on HMM substrate is about 3 times higher compared to the reference sample with a bare glass substrate.

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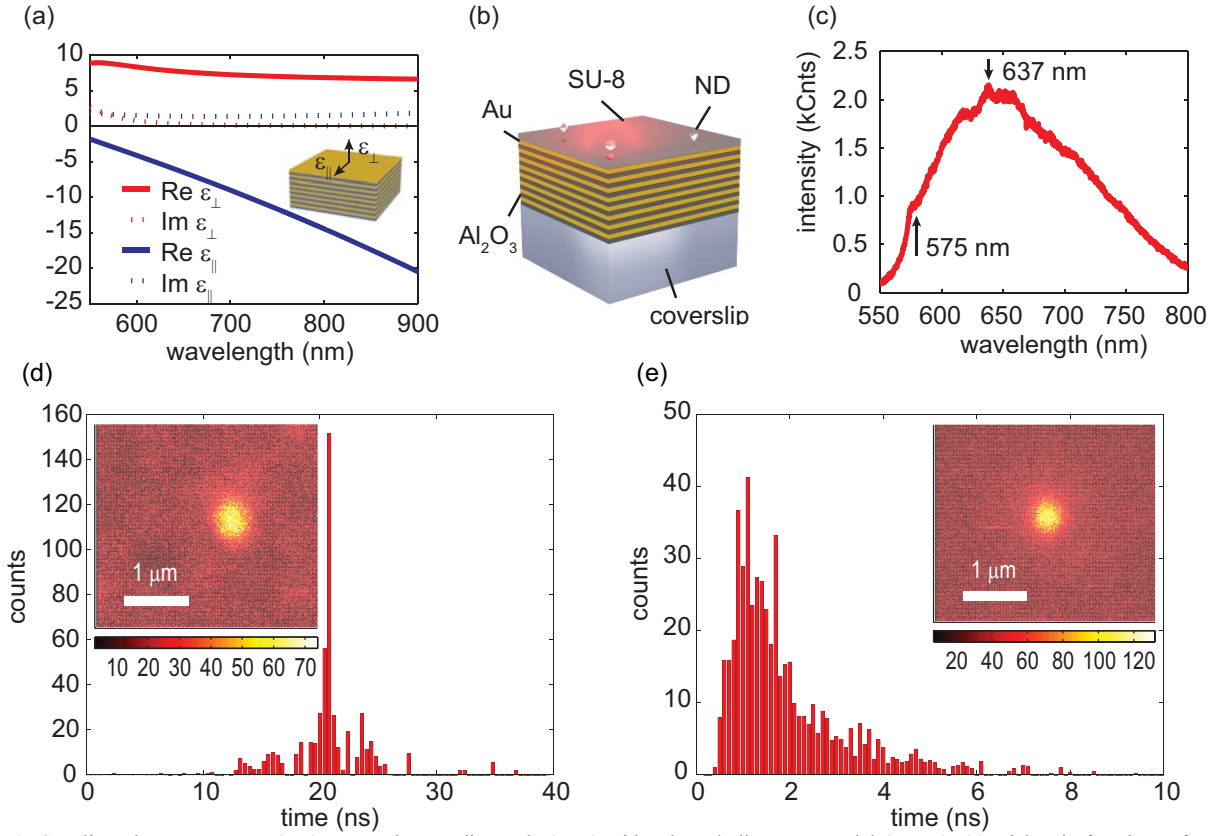

Fig. 1. Coupling nitrogen-vacancy (NV) centers in nanodiamonds (NDs) with a hyperbolic metamaterial (HMM). (a) Dielectric functions of fabricated HMM (16 alternating layers of Au and Al_2O_3, thickness of each layer – 19 nm) retrieved by spectroscopic ellipsometry measurements, within the range of the plot (550 – 900 nm) $\mathrm{Re}\,\varepsilon_\perp > 0$, $\mathrm{Re}\,\varepsilon_\parallel < 0$, hence leading to hyperbolic dispersion. (b) Schematic of the experimental sample consisting of the HMM, 30-nm-thick spacer layer composed of SU-8 photoresist, and NDs with NV centers inside. (c) Emission spectra of NV centers when NDs are placed on HMM. Experimentally measured spontaneous emission lifetimes of the NV centers on (d) coverslip, (e) HMM. Corresponding mean values (standard deviations) of the lifetime distributions are: (d) 20.89 ns (1.15 ns), (e) 1.55 ns (0.95 ns).

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