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Enhancement of luminescence of quantum emitters in epsilon-near-zero waveguides

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ABSTRACT

We report a resonant enhancement of luminescence intensity from an ensemble of CdSe/ZnS quantum dots embedded in a nanoscale rectangular photonic waveguide operating in the cutoff regime and therefore experimentally demonstrate the recently predicted phenomenon of increase in the emission rate of an ensemble of quantum emitters in the epsilon-near-zero environment.

Enhancement of light emission from quantum emitters is one of the main goals of nanophotonics. The increase in a quantum system's spontaneous emission rate by its environment and, in particular, by confinement in a resonant cavity known as the Purcell effect[1,2] is widely used: multifold enhancements of the emission rate have been demonstrated in emitters embedded in plasmonic[3] and dielectric metamaterials.[4] A spectacular increase in the spontaneous emission rate has been observed in quantum light emitters placed in nanoscale plasmonic resonators.[5,6] An array of coupled plasmonic-enhanced emitters can be forced into a collective mode of coherent emission by coupling between the resonators (lasing spaser[7,8]). The extraordinary electromagnetic field with no spatial phase change and extremely large phase velocity offered by near-zero-index (NZI) media[9] suggests that such collective coherent emission of an ensemble of quantum emitters can also be achieved in NZI media.[10,11]

As the index of refraction, $n(\omega) = \sqrt{\varepsilon(\omega)\mu(\omega)}$, is determined by the relative permittivity, $\varepsilon(\omega)$, and the relative permeability, $\mu(\omega)$, there are three ways to achieve the zero index: epsilon-near-zero (ENZ),[10,12–14] mu-near-zero (MNZ), and epsilon-and-mu-near-zero (EMNZ).[15] As the Einstein A_{21} coefficient for spontaneous emission is proportional to the refractive index of the surrounding medium, it is expected that spontaneous emission is strongly suppressed in NZI media. However, a recent theoretical study[16] revealed that the spontaneous emission rate can be enhanced even in NZI media if the impedance is large enough to compensate the depletion of the modes at the ENZ point. 1D ENZ[10,12–14,17] media are predicted to show divergent enhancement due to the divergent impedance at the ENZ point, while 1D EMNZ and 2D ENZ[18] media are to show finite rate enhancement.

1D ENZ waveguides[10,12,14,17] are of particular interest as they are compatible with integrated photonic circuits. It has been shown that 1D ENZ waveguides can support phase-free propagation[17] and local density of optical state (LDOS) enhancement[14] at the ENZ point, but the behavior of quantum emitters embedded in 1D ENZ waveguides has not been studied/characterized experimentally. Here, we report the experimental demonstration of resonant enhancement of the intensity of emission of an ensemble of quantum emitters embedded in a 1D ENZ media, a nanoscale photonic waveguide operating at the cutoff.

A rectangular waveguide is widely used as a microwave component that supports transverse electric (TE) and transverse magnetic (TM) modes for the wave transmission. When it is scaled down to nano-scale in the form of a dielectric core surrounded by metallic sidewalls, it supports the dominant quasi-TE mode, which shows cutoff behavior, and the position of this cutoff can be easily tuned with the refractive index, n, and width, w, of the dielectric core. This type of waveguide can serve as an epsilon-near-zero medium near the cutoff frequency and exhibits the enhanced LDOS near such cutoff.[14,19] Enhanced luminescence of quantum emitters is expected when they are embedded in such waveguides whose cutoff is properly tuned to the emission wavelength of the emitters.

To study the luminescence of quantum emitters in the ENZ regime, we constructed a series of optical waveguides with different cutoff frequencies in the optical part of the spectrum. The waveguides had rectangular cross sections, a poly(methyl methacrylate) (PMMA)

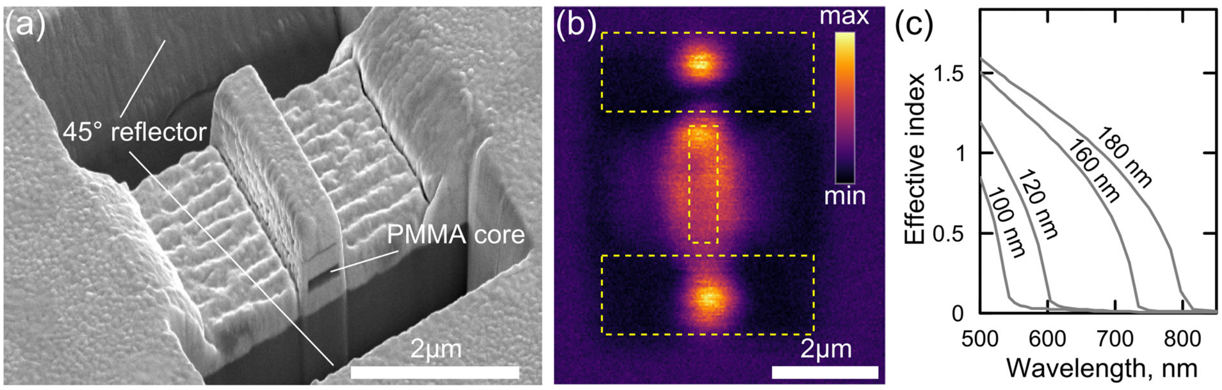

FIG. 1. Quantum emitters in the ENZ waveguide. (a) Scanning electron microscopy image of a QD-embedded waveguide with two 45° reflectors. (b) Scanned photoluminescence intensity map. The top and bottom bright spots in the map indicate the positions on the 45° reflectors for in- and outcoupling of the pump and emitted light. (c) Effective refractive index of quasi-TE mode in the waveguides of different widths, *w*.

dielectric core, and silver cladding [see Fig. 1(a)]. The waveguides were embedded with CdSe/ZnS quantum dots (QDs) (size, ~9 nm; diluted to 1.5 nmol/ml; NN-labs) with a central emission wavelength of ~630 nm. To manufacture the waveguides, we thermally evaporated a silver film on the silicon substrate and then spin-coated on the silver layer a 100 nm-thick layer of a mixture of PMMA and QDs with an area density of 90 QDs/μm^2. After another layer of thick silver film was deposited, the film was milled by focused ion beam to define nine waveguides with the width from 100 nm to 180 nm, with 10 nm steps. All waveguides had the same length of 700 nm. A subsequent deposition of the silver film by thermal evaporation was followed to cover the exposed sidewalls. We also manufactured control waveguides by skipping the silver deposition on the sidewalls. As the parallel plate waveguides support TEM modes, the control waveguides do not show any cutoff or strong resonance behavior near the spectral region of QD emission wavelength, ~630 nm. Two 45° mirror facets for in- and outcoupling of the pump laser and luminescence were fabricated on each waveguide by focused ion beam milling. Figure 1(c) shows the effective index of quasi-TE mode for the waveguides with $w = 100$, 120, 160, and 180 nm. The effective index is given by the ratio of the propagation constants of the guided mode and the electromagnetic wave in the free-space, respectively.

The photoluminescence of the QDs embedded in the waveguides was characterized by observing the emission of the waveguide pumped by a laser at the wavelength of 405 nm. The pump was focused, and luminescence was collected using the same high-numerical aperture objective (Nikon CFI LU Plan FLUOR Epi 100X, NA = 0.9). To identify optimal conditions for coupling the pump into the waveguides, we first recorded the intensity maps of integrated photoluminescence by scanning the sample with a piezo-stage [Fig. 1(b)]. The two bright spots in the map indicate positions where the pump laser is efficiently coupled into the waveguide via the 45° mirrors. The spectra of the QD luminescence signal at the outcoupling mirror were, then, detected for each waveguide using an imaging spectrometer equipped with a thermoelectrically cooled CCD.

In the control waveguides without sidewalls, the spectra of QD luminescence do not depend on the waveguide width [Fig. 2(a)]. In the rectangular waveguides that show ENZ behavior at the cutoff, the QD luminescence spectra strongly depend on the waveguide width. Figure 2(b) shows the PL spectra from QD-embedded waveguides where the spectra are normalized by matching the background luminescence level at 570 nm to unity. The luminescence from QDs in a 100 nm-wide waveguide is suppressed, and the spectrum is similar to that from the unstructured sample. However, as the width is gradually increased, the luminescence shows a sudden jump in intensity in the ENZ regime, for $w = 160$ nm. The peak of emission is centered at the wavelength of 650 nm that is in between the cutoff wavelengths of 670 nm and 630 nm and the emission maximum wavelength of free QDs [Fig. 2(b)]. The strong luminescence peak disappears in the waveguides with the width exceeding 160 nm. The reduced intensity of luminescence in these waveguides compared to the control waveguides in Fig. 2(a) is attributed to the increased impedance mismatch at the pump wavelength, 405 nm, since TEM mode with much smaller characteristic impedance is not supported in these rectangular waveguides. Although the increased impedance of rectangular waveguides reduces the pump coupling efficiency, it also boosts the otherwise negligible spontaneous emission probability due to the negligible number of

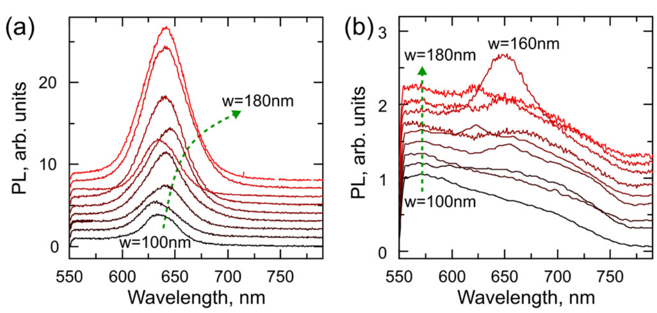

FIG. 2. Luminescence of QDs in waveguides. Photoluminescence spectra of QDs in waveguides (a) without and (b) with sidewalls with the width from 100 nm to 180 nm in 10 nm steps. Note the steady increase in intensity of luminescence in open-sided waveguides that show no ENZ behavior and abrupt appearance of the strong luminescence peak in the ENZ regime for a waveguide with a width of 160 nm.

optical modes at the cutoff.[16] With a further increase in the waveguide width, QD luminescence exhibits a series of weaker maxima related to the Fabry–Pérot resonances along the waveguide axis, which is consistent with the theoretical prediction in Ref. 10. While Fabry–Pérot resonances depend on the waveguide length due to the dispersive nature of the modes, the resonance at the ENZ point does not depend on the waveguide length and the position of the emitters within the waveguide.[10]

In an ideal 1D ENZ medium without loss, the Purcell effect is manifested as enhancement above cutoff and complete inhibition below cutoff, which produces an asymmetric line shape, unlike the curve in Fig. 2(b) for $w = 160$ nm. We attribute the symmetric line shape of the emission observed experimentally to the material loss and the use of finite waveguides, which can soften the asymmetric details by allowing the emitters near the waveguide openings to radiate into the farfield.

Superradiance[20] from ensembles of quantum emitters is one of the exciting phenomena that are sought after in ENZ media. This collective emission of radiation from an ensemble of quantum emitters requires the emitters to experience the same radiation field: the electromagnetic field with no phase advancement at the ENZ point can relax the spatial restriction to achieve superradiance.[10,21,22] Although it is not part of our present work, it would be interesting to study the effect of the waveguide length on the luminescence intensity and the bunching of photons at the cutoff to verify the superradiance concept in ENZ media.

In conclusion, we have demonstrated experimentally the recently predicted increase in the emission rate of the ensemble of quantum emitters in the epsilon-near-zero environment. We observed the phenomenon in the luminescent spectra of QDs embedded in a nanoscale rectangular waveguide at the cutoff wavelength where the waveguide exhibits the epsilon-near-zero behavior. The phenomenon offers interesting opportunities for developing high brightness coherent quantum sources.

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DATA AVAILABILITY

The data that support the findings of this study are available from the NTU research data repository at https://doi.org/10.21979/N9/YOSN8Z.

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