

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 at the cut-off regime and therefore experimentally demonstrate the recently predicted phenomenon of increase of 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 of 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 plasmonic³ and dielectric metamaterials⁴. Spectacular increase of 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) media⁹ 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’s 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, recent theoretical study¹⁶ revealed that 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 ENZ¹⁸ 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 propagation¹⁷ and local density of optical states (LDOS) enhancement¹⁴ at 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 cut-off behavior and the position of this cut-off can be easily tuned with the refractive index, n , and width, w , of the dielectric core. This type of waveguides can serve as an epsilon-near-zero medium near the cut-off frequency and exhibit the enhanced LDOS near such cut-off^{14, 19}. Enhanced luminescence of quantum emitters is expected when they are embedded in such waveguides whose cut-off is properly tuned to the emission wavelength of the emitters.

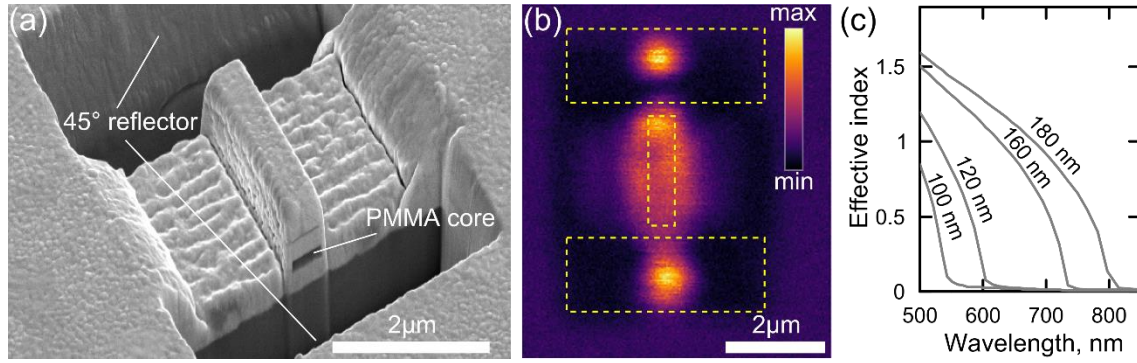


Figure 1. **Quantum emitters in the ENZ waveguide.** (a) Scanning electron microscope image of a QD-embedded waveguide with two 45° reflectors; (b) Scanned photoluminescence intensity map. The top and bottom bright spots in the map indicate the positions on the 45° reflectors for in- and out-coupling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

To study 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, poly(methyl methacrylate) (PMMA) 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 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 area density of $90 \text{ QDs}/\mu\text{m}^2$. 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 step. All waveguides had the same length of 700 nm. A subsequent deposition of 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 cut-off or strong resonance behavior near the spectral region of QD emission wavelength, ~ 630 nm. Two 45° mirrors facets for in- and out-coupling of 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 ratio of the propagation constants of the guided mode and the electromagnetic wave in the free-space, respectively.

Photoluminescence of the QDs embedded in the waveguides was characterized by observing emission of the waveguide pumped by a laser at the wavelength of 405 nm. The pump was focused and luminescence was collected by the same high-numerical aperture objective (Nikon CFI LU Plan FLUOR Epi 100X, NA=0.9). To identify optimal condition 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 out-coupling mirror were then detected for each waveguide by an imaging spectrometer equipped with a thermoelectrically-cooled CCD.

In the control waveguides without sidewalls the spectra of QD luminescence does 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 QDs-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 unstructured sample. However, as the width is gradually increased, the luminescence shows a sudden jump in intensity

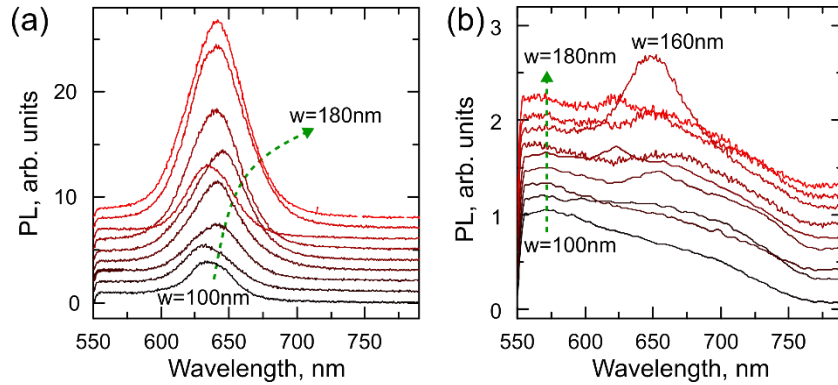


Figure 2. **Luminescence of QDs in waveguides.** Photoluminescence spectra of QDs in waveguides (a) without and (b) with sidewalls with the width from 100 nm to 180 nm in 10 nm steps. Note steady increase of intensity of luminescence in open-sided waveguides that show no ENZ behavior and abrupt appearance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

at 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 wavelength of 670 nm and 630 nm, the emission maximum wavelength of free QDs (Fig. 2(b)). The strong luminescence peak disappears in the waveguides with 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 negligible number of optical modes at the cut-off¹⁶. With the further increase of 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 ENZ point does not depend on the waveguide length and the position of the emitters within the waveguide¹⁰.

In an ideal 1D ENZ medium without loss, the Purcell effect is manifested as enhancement above cut-off and complete inhibition below cut-off, which produces an asymmetric lineshape, unlike the curve in Fig. 2(b) for $w = 160$ nm. We attribute the symmetric lineshape 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.

Superradiance²⁰ 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 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 waveguide length on the luminescence intensity and the bunching of photons at the cut-off to verify the superradiance concept in ENZ media.

In conclusion, we have demonstrated experimentally the recently predicted increase of emission rate of 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 cut-off 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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