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Tailoring Magnetic Dipole Emission by Broken-Symmetry TiO_2 Metasurfaces

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Abstract. Strong magnetic dipole emission is offered by rare earth ions such as trivalent lanthanides, due to selection rule forbidden electric dipole (ED) transitions. This stimulates the study of optical nanostructures, which efficiently tailor magnetic dipole emission. High refractive index all dielectric nanostructures are promising candidates in this regard due to their strong magnetic response and negligible absorption loss in the visible spectral range. Here, we design and experimentally realize a broken-symmetry titanium dioxide (TiO_2) metasurface supporting an out-of-plane magnetic dipole (MD) resonance at 590 nm wavelength, corresponding to the MD transition of trivalent Europium ions (Eu^{3+}). A strong photoluminescence (PL) enhancement of the MD transition up to a factor of 15.5 is observed.

1 Introduction

High index all-dielectric metasurfaces Offer many important opportunities for the realization of highly efficient flat optical devices [1-3]. In particular, by coupling to their multipolar electric and magnetic Mie-resonances, they can efficiently manipulate spontaneous emission of coupled quantum emitters [4].

A particularly interesting class of all-dielectric metasurfaces for spontaneous emission control are those supporting high quality (Q) factor bound states in the continuum (BIC)**.** Lying inside the continuum while being isolated from the radiation loss, BIC states are characterized by a strong energy confinement and consequently field enhancement localized to the structure supporting them [5]. In a typical symmetric system, the coupling between bound state and continuum is forbidden by symmetry. Through breaking the in-plane symmetry of the nanostructure, a weak coupling between its bright in-plane and dark out-of-plane modes can occur which leads to emergence of high Q- factor sharp resonances since in this case, the radiation loss is suppressed, and the energy is stored inside the nanostructure. These high Q-factor resonances can provide a strong PL enhancement and emission directionality [6].

In optical frequency range, MD transitions in a quantum system are forbidden since their transition rates are usually 4-5 orders of magnitude weaker than allowed ED transition rates [7]. However, MD transitions with a comparable strength as ED transitions exist in lanthanide ions, such as trivalent europium (Eu^{3+}), which makes them a proper candidate for magnetic light matter interaction studies. Eu^{3+} emitters possess a strong MD transition at 590 nm ($^5D_0 \rightarrow {}^7F_1$) and several ED transitions in visible range including the most dominant one at 610 nm ($^5D_0 \rightarrow {}^7F_2$) [8,9].

2 Methods and Results

In this work, we designed Mie-resonant metasurfaces composed of broken symmetry TiO_2 nanoparticles, that support high Q-factor out-of-plane magnetic dipole resonances at 590 nm, corresponding to the MD transition of Eu^{3+}. We fabricated the metasurfaces using electron-beam lithography (EBL) in combination with reactive ion

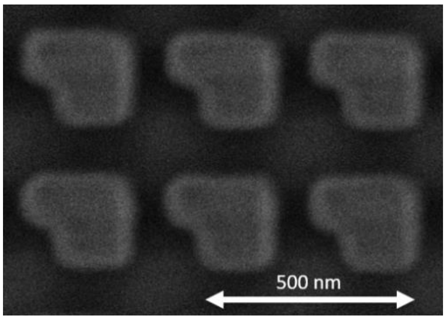

Fig. 1. Top-view SEM image of a fabricated broken-symmetry TiO_2 metasurface.

etching . We then spin-coated the fabricated metasurfaces with a polymethyl methacrylate (PMMA) resist film containing Europium(III) thenoyltrifluoroacetone. Figure 1 shows a top-view scanning electron microscope (SEM) image of a typical fabricated sample.
Next, using a commercially available confocal laser scanning microscope setup (PicoQuant, MicroTime200), we performed PL spectroscopy to investigate PL emission enhancement of Eu^{3+} provided by the broken-symmetry

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metasurface. Measured PL emission spectra for both the coated metasurface and an unstructured coated reference area is displayed in the Figure 2.

Using a 4 × /0.1NA collective objective, we achieved a brightness enhancement of up to 15.5 for the MD transition at 590 nm. The ED transition of Eu^{3+} at 610 nm

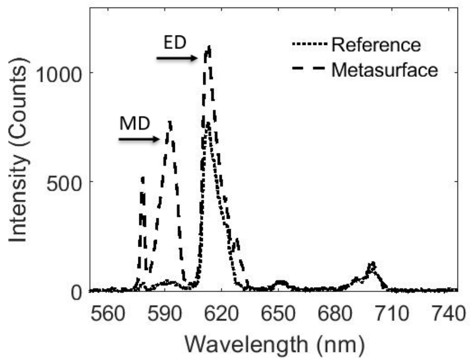
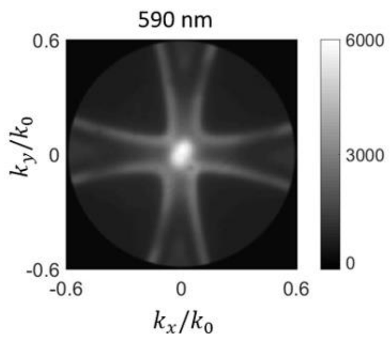


Fig. 2. The PL emission spectra of the Eu^{3+} doped PMMA covered sample.

is also enhanced, but to a much lower degree. The enhancement ratio of the MD transition at 590 nm to the ED transition is ~8.7.

Furthermore, to investigate the spatial emission characteristics of the sample, we performed back focal plane (BFP) imaging of the emission from the coated metasurface at the two wavelengths of 590 nm and 610 nm using a 0.6 NA objective. The result is shown in Figure 3.



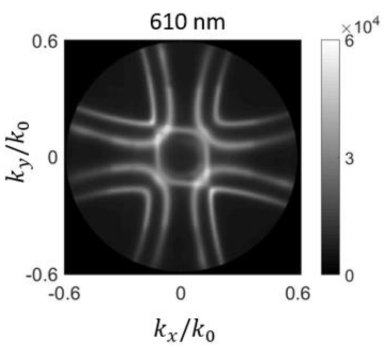


Fig. 3. Measured BFP images of the sample at 590 nm and 610 nm wavelength, corresponding to the electric and magnetic dipole transition, respectively.

The BFP images of the sample show a bright emission for the near-zero polar angles, i.e., a strong forward emission normally out of the sample plane, for the MD at 590. For the ED at 610 nm, instead, forward emission is strongly suppressed. As such, the strong PL emission enhancement for the MD observed while measuring with a low NA collective objective can be explained by the dramatically different emission patterns observed for the two different transitions. From this we can conclude the obtained selective MD enhancement is mainly due to an improved collection efficiency for that transition. Our results offer a novel possibility toward tailoring the directionality of MD-dominated spontaneous emission. It is of a great importance for the research and application exploiting magnetic nature of light. Furthermore, controlling the directionality of the emission is strongly demanded to realize optical devices with high efficiency.

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