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Shaping Emission Spectra of Quantum Dots by All-dielectric Metasurfaces

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Abstract: Silicon nanodisks support both electric and magnetic resonances, which can be tuned independently *via* their geometry. We utilize these engineered resonances and demonstrate dielectric metasurfaces for efficient shaping of the emission spectra of quantum dots.

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Driven by their huge potential for a range of important potential applications such as plasmonic metamaterials, metallic nanostructures have been extensively studied in the last decade and remain an active field of research. However, plasmonic nanostructures inherently suffer from strong dissipative losses of metals at optical frequencies, thereby critically hampering their performance and practical applicability in nanophotonic devices. A new route to overcome this persistent problem has been opened by the recent experimental observations of electric and magnetic Mie-type resonances in high-permittivity all-dielectric nanoparticles. Such all-dielectric nanoparticles exhibit very low losses at optical frequencies and their resonances can be utilized, in a direct analogy to plasmonic resonances, to engineer both magnetic and electric optical response. When combined together, such all-dielectric nanoparticles can form a new class of metamaterials and metasurfaces with unique lossless properties, thus opening novel applications in nanophotonics, including wavefront engineering or advanced light sources.

Of particular interest for all-dielectric optical nanoantennas is the controlled interference of the electric and magnetic resonances. The case of close proximity of these modes enables the precise tailoring of directional scattering [1-4], including suppression of resonant backward scattering in the case of a spectral mode overlap [3] and reversal of the scattering direction depending on the incident wavelength [4]. However, to date most experiments on scattering by all-dielectric nanoparticles have been focused on geometries like spheres or cubes, which do not allow for the individual spectral tuning of the magnetic and electric resonances [5]. This limitation has been recently overcome by the use of all-dielectric silicon nanodisks. Nanodisks enable the individual spectral control over the induced electric and magnetic resonances of the particles, including the case of a spectral mode overlap [6]. Using this spectral overlap here we demonstrate functional metasurfaces, which can strongly enhance and shape the spectrum of emission of quantum dots, through close coupling to the metasurface.

In our experiments, we fabricate silicon nanodisk arrays on SiO_2 substrate using electron-beam lithography on silicon-on-insulator wafers, in combination with reactive ion etching. Next, the structures are covered with a thin polymer layer containing spectrally matched PbS QDs. as schematically illustrated in Fig. 1(a). A monotonous variation of the nanodisk diameter d (samples #1-6: d = 450, 475, 500, 550, 575, and 600 nm, respectively) at constant nanodisk height (220 nm) allows us to tune the spectral positions of the nanodisks' electric and magnetic resonances with respect to each other and to bring them into a spectral overlap [6]. Fig. 1(b) shows the near-normal-incidence linear-optical transmittance spectra of the fabricated nanodisk array metasurface, measured using white-light spectroscopy. In order to compare our experimental results with theory we also calculate the corresponding normal-incidence transmittance spectra for the experimental sample parameters using CST Microwave Studio. We obtain a very good agreement with experimental data [see Fig. 1(c)]. Our calculations furthermore reveal that near-unity transmittance is obtained for a perfect mode-overlap in the absence of Fabry-Perot resonances that originate from the layered silicon-on-insulator wafer structure.

For the largest nanodisk diameter, two clearly separate resonances are observed [see sample #6 in Figs. 1(b,c)], which are characterized by low transmittance (transmittance spectral dips). These dips correspond to the fundamental electric and magnetic dipolar resonances of the nanodisks, which positions were also calculated independently and indicated by the cyan and red lines in Figs. 1(b,c), respectively. As the disk diameter is reduced, the two resonances move closer together until they overlap spectrally, before they start separating again. Importantly, the interference of electric and magnetic modes of the individual subwavelength silicon nanodisks leads to suppression of resonant reflection from the metasurface. This results in higher transmittance levels (instead of transmittance dips) for the case of overlapping magnetic and electric resonances. Furthermore, highly directional forward scattering can also be obtained for optimized silicon nanodisks under excitation by a localized dipole source, demonstrating the great potential of silicon nanodisk resonators for control of the directivity and emission spectrum of localized emitters [6]. However, it remains an open question if silicon nanodisks metasurfaces can also facilitate the energy extraction from nanoemitters, for their use as emission control of quantum dots, atoms or molecules.

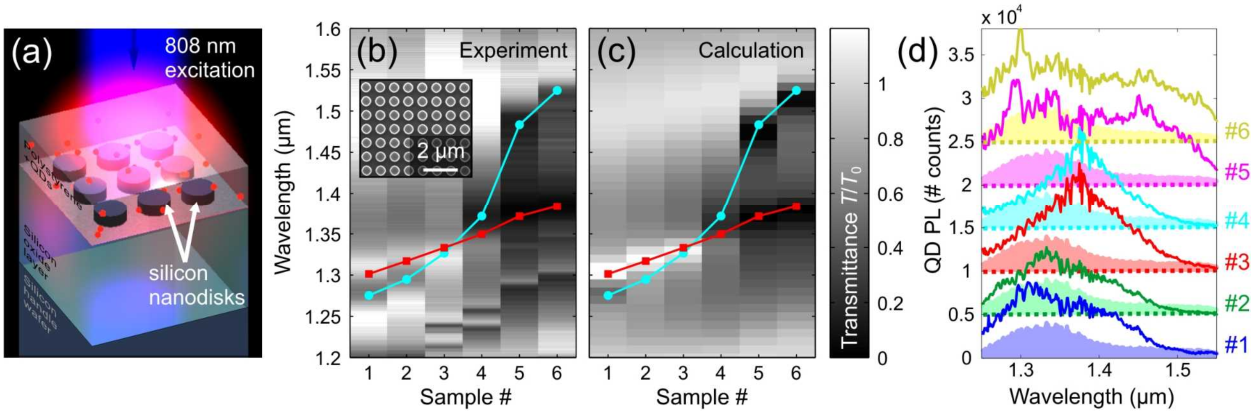

Fig. 1: (a) Sketch (not to scale) of PbS QDs coupled to silicon nanodisks. (b) Experimentally measured and (c) numerically calculated linear-optical transmittance spectra of silicon nanodisk metasurface with systematically increasing disk diameters from sample #1 to sample #6. The inset in (b) shows an SEM image of a typical sample. Red and cyan lines indicate the numerically extracted spectral positions of the magnetic and electric dipole modes, respectively. (d) Measured micro-PL spectra of PbS QDs coupled to the silicon nanodisk samples. The shaded areas indicate the emission of the QDs on a bare wafer for reference.

In order to gain an insight into the interplay between nanoemitters and silicon nanodisk resonators, we investigate experimentally how the spontaneous emission properties of near-infrared PbS QDs are influenced by coupling to the nanodisks exhibiting closely spaced electric and magnetic Mie-type modes. We then perform micro-photoluminescence (PL) spectroscopy measurements of the coupled system for various nanodisk diameters.

Our experimental results are shown in Fig. 1(d), and they clearly show that the spectral QD PL line shape is strongly dependent on the disk aspect ratio. In particular, the spectral positions of the local maxima correlate with the electric and magnetic resonant frequencies in different structures, indicating a good coupling of the QDs to the Mie-type resonances of the silicon nanodisks forming the metasurface. This correlation is preserved for the overlap of resonances, indicating that a high photonic local density of states exists in the metasurface despite a lack of a pronounced spectral dip signature in the transmittance spectra due to the suppression of the backward scattering and predominant forward scattering. Such a behavior cannot be achieved by any isolated optical resonance in any type of nanoparticle arrays.

Our results demonstrate that metasurfaces formed by a lattice of high-index all-dielectric nanodisks are not only able to tailor directional scattering, but can also influence the process of the energy extraction from a localized emitter. Based on these key functionalities, we expect silicon metasurfaces to play an important role in the key applications, including directional nanoantennas, display technologies and functional lossless metasurfaces.

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References

[1] M. Kerker, D.-S. Wang, and C. L. Giles, "Electromagnetic scattering by magnetic spheres," J. Opt. Soc. Am. **73**, 765-767 (1983).

[2] S. Person, M. Jain, Z. Lapin, J. J. Saenz, G. Wicks, and L. Novotny, "Demonstration of Zero Optical Backscattering from Single Nanoparticles", Nano Lett. **13,** 1806-1809 (2013).

[3] W. Liu, A. E. Miroshnichenko, D. N. Neshev, and Yu. S. Kivshar, "Broadband Unidirectional Scattering by Magneto-Electric Core-Shell Nanoparticles", ACS Nano **6,** 5489-5497 (2012).

[4] Y.-H. Fu, A. I. Kutznetsov, A. E. Miroshnichenko, Y. F. Yu, and B. Luk'yanchuk, "Directional Visible Light Scattering by Silicon Nanoparticles", Nat. Commun. **4,** 1527-1- 1527-6 (2013).

[5] A. B. Evlyukhin, C. Reinhardt, and B. N. Chichkov, "Multipole Light Scattering by Nonspherical Particles in the Discrete Dipole Approximation", Phys. Rev. B **84,** 235429-1-235429-8 (2011).

[6] I. Staude, A. E. Miroshnichenko, M. Decker, N. T. Fofang, S. Liu, E. Gonzales, J. Dominguez, T. S. Luk, D. N. Neshev, I. Brener, and Yu. Kivshar, ACS Nano **7,** 7824-7832 (2013).