Control of single-photon emitters in two-dimensional materials using dielectric nanoantennas

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Abstract: We show that dielectric nanoantennas are capable of inducing very high Purcell enhancement up to factors > 45 for defect-based single-quantum emitters in atomically thin layered materials, enabling bright single-photon emission with polarization control. © 2022 The Author(s)

1. Introduction

Sub-wavelength dielectric resonators supporting Mie resonances have attracted significant interest due to their ability to control light-matter interaction without material loss. Dielectric nanoantennas have found numerous applications in directional light emission and nonlinear light-matter interaction enhancement [1]. Leveraging the advantages and favorable optical properties of dielectric nanoantennas to the enhancement of quantum light generation is a very exciting direction. Recently, strain and defect-activated single-photon emitters in WSe2 have shown great advantages in highly pure, high yield, and site controllable quantum light generation. Using engineered substrates for strain modulation is a widely used method to deterministically create single-photon emitters (SPEs) in two-dimensional (2D) materials such as transition metal dichalcogenides (TMDs) [2], as shown in Fig. 1(a); however, the majority of such studies use low refractive index materials such as silicon dioxide, which lacks the optical response that can enhance the brightness of the SPEs. The integration of high refractive index subwavelength resonators supporting Mie resonances with 2D materials to produce and engineer single-photon emitters promises a straightforward and highly compact scheme for inducing the strain leading to confinement of SPEs as well as an ultra-high Purcell enhancement leading to brighter single-photon emission. Additionally, engineering the geometries of sub-wavelength antennas can lead to many other advantages, including directional emission of single photons, polarization control, and chirality. Recent reports have shown that this integration can significantly improve an emitter's brightness and quantum efficiency with dielectric dimers [3]. Here we present new designs of subwavelength dielectric antennas and verify bright emission from single-quantum emitters in the monolayer TMD WSe2. In addition, we study different regimes of operation for such antennas that can be used to enhance light scattering vs. the emitters' brightness. Our designs show great versatility in combining strain engineering, Purcell enhancement, and polarization control of defect-based single-quantum emitters in atomically thin layered materials.

2. Results and discussion

In this work, we investigate four different structures of sub-wavelength antennas and their effects on the singlephoton emission from a monolayer WSe2 transferred onto a dimer sample. All designs here are made of crystalline silicon with a height of 220 nm. The structures under investigation include a square-shaped monomer (Fig. 1(c)), a disk monomer (Fig. 1(d)), a square dimer (Fig. 1(e)), and a disk dimer (Fig. 1(f)). Simulation results show that all geometries can be optimized to exhibit significant Purcell enhancement (F_P) . Figure 1(h) shows the simulation results of Purcell enhancement of the square dimer structure. Side length dimensions can be selected for high F_P depending on the emitter's wavelength. For example, for the wavelength of interest for SPEs in WSe₂, 800 nm, we can theoretically achieve F_P as high as 35 with a dimer side length of 450 nm and a gap width of 50 nm; however, the maximum scattering cross-section does not necessarily coincide with the highest F_P enhancement at a given wavelength. As shown in Fig. 1(i and j), we can obtain the highest Purcell enhancement for an emitter around 800 nm at a dimer side length of 450 nm; however, this doesn't correspond to the scattering peak for these dimensions, and side lengths of 325 nm or 400 nm will lead to higher scattering of the single photons with slightly lower F_P enhancement, which provides flexibility in choosing the dimer geometry to optimize emission versus Purcell enhancement. The choice of the design is highly dependent on the type of response one wishes to obtain. For example, a monomer disk antenna can reach F_P of 15, while a dimer of disks with a 50 nm gap can reach up to $F_P = 45$ on top of the gap; however, the strain is more likely to to be stronger at the edges, which is important for SPE creation [2]. The square geometries have very similar advantages with the additional benefit of being able to control the polarization of the emitted single photon, through engineering a uni-directional strain field in the layered material, which is the reason we focus on this geometry going forward.

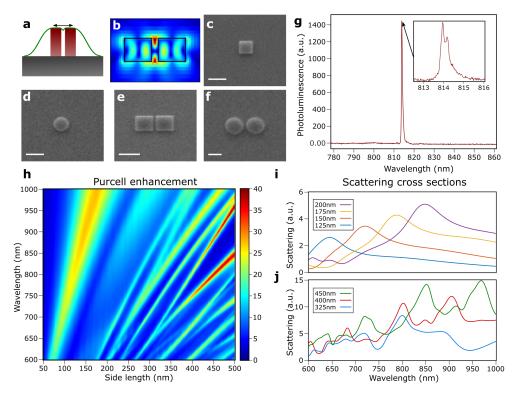


Fig. 1. Engineering single-photon emission using dielectric nanoantennas. (a) A schematic of 2D flake on top of a dimer nanoantenna. (b) Electric field enhancement at wavelength 800nm in a square-shaped dimer with a side length of 325 nm and gap width of 50 nm. (c-f) Scanning electron microscope images of the different dielectric antennas. Scale bar in (c-f) is 500 nm. (g) Photoluminescence of a single-photon source in WSe₂ on top of a dimer measured at 4.2K. Inset shows a zoomed-in image revealing a doublet with a fine structure splitting of 0.3948 meV. (h) Simulated Purcell enhancement of a dipole emitter 1 nm on top of a square-shaped dimer nanoantenna (shown in (e)) as a function of the dimer side length. (i,j) Scattering cross-sections of the square dimer with different side lengths. The same normalization is used for both (i) and (j) for comparison.

To demonstrate bright emission from the dimer, we used a dry transfer method to place a monolayer WSe₂ flake on top of the dimer array. The sample is cooled down in a closed-cycle cryostat to \sim 4.2 K. Photoluminescence spectrum is taken using a continuous 532 nm laser (10 uW excitation power and long-working distance objective with 0.55 numerical aperture). Scanning the laser spot across the sample, we observe bright SPEs in nearly all nanoantennas where the flake coincides. Nanoantenna sites display bright emission lines with linewidth as narrow as \sim 0.2 meV, which is comparable with linewidths observed in hBN encapsulated monolayer WSe₂. An example of a measured SPE spectrum (Fig. 1(g)), exhibits a doublet with fine structure splitting of 0.395 meV, which is consistent with what had been reported for WSe₂ emitters.

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