monocrystalline gold flakes [25, 26] are quite promising for resolving this fundamental problem, and our measurements on similar gold flakes support the suppressed luminescence (not shown). In this work, we choose to focus on utilizing the plasmon-coupled gold luminescence as a method to measure the dependence of the plasmon mode spectrum on the dielectric environment.

Figure 3b presents normalised DF and PL spectra along with the simulated scattering spectra for the five positions of the NC, discussed in Fig. 2. Each spectrum is fit with a Voigt function to determine the peak wavelength. While the peak wavelength depends significantly on the NC distance to the centre of the antenna gap, the plasmon resonance lineshape is essentially unchanged by the presence of the NC. The FDTD simulations of the plasmon lineshape (top set of curves) confirm that the lineshape is unaffected by the presence of the NC. Figure 3c shows the dependence of the resonance wavelength on the NC distance to the gap centre. As the overlap between the NC and the near-field profile of the plasmon mode increases, the plasmon mode samples more of the higher index dielectric and the observed resonance exhibits a nonlinear shift to longer wavelengths. Figure 3c also shows a few-nm offset between DF and PL spectra at each NC position. DF and PL are two fundamentally different mechanisms and are susceptible to wavelength-dependent coupling efficiency of the two light sources. Identifying the exact cause of the offset will require further study.

The measured dependence of the wavelength on the NC position shows reasonable agreement with our simulations. Finally we position the NC next to one of the outer corners of the BT antenna and measure the spectrum of the fundamental plasmon mode. Figure 3c shows that the peak wavelength for this configuration is nearly identical to that of the BT without the NC (Fig. 2f). Figure 3b also confirms that the plasmon mode lineshape is unaffected by the NC at the outer corner. Figure 2 panels f-i show that the field is still strong at the outer corners at the substrate level, while it is strongly reduced at 17.5 nm above the substrate (Figs. 2j-m). Therefore, strong spectral shift would be observed if the NC had overlap with the mode at the substrate level. The absence of any spectral shift is the experimental verification that the NC we are positioning has a smaller footprint at the substrate level than its waist and that the spectral tuning of the mode is achieved by accessing the field well above the substrate.

4. Conclusion

Here we show that a dielectric nanocrystal can be used for tuning the resonance wavelength of a plasmonic BT antenna on the order of a linewidth. This ability is essential for controlling the spectral overlap of a quantum emitter, such as a single molecule, a quantum dot, or a diamond color center, with a plasmonic cavity mode. The spectral tuning is achieved by modifying the mode sufficiently above the substrate level in our experiments, and therefore, the anticipated spatial overlap of a quantum emitter with the mode distribution would be unaffected at the substrate level. We further show that gold luminescence is enhanced by the fundamental plasmon mode of a BT antenna by more than two orders of magnitude. The degree of coupling of the gold luminescence to the BT antenna is a roadblock for applications requiring coherent (strong) emitter-cavity coupling, but it can be used as a tool for characterizing the spectral properties of gold-based plasmonic nanostructures.

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