Supporting Information

Nanoassembly of quantum emitters in hexagonal boron nitride and gold nanospheres

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Here, we present additional data on AFM, simulations, optical and physical characterizations of our materials.

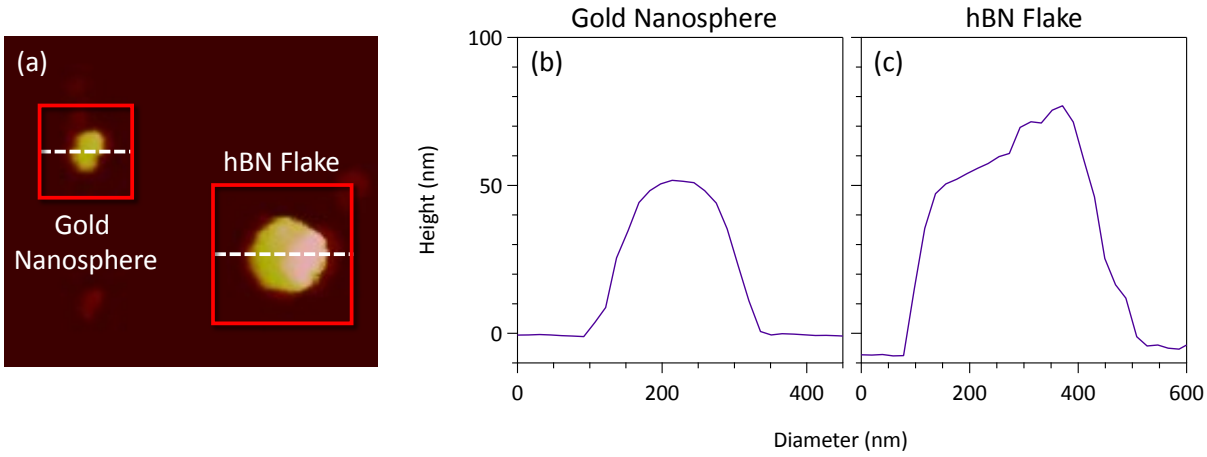

Figure S1. AFM images of the flake and gold NP with cross-sectional analysis. (a) AFM image of the labelled flake and a nanosphere. Cross sectional analysis is performed along the broken

white lines. (b) Cross-sectional analysis of the nanospheres showing a height of ~52 nm. (c) Cross-sectional analysis of the hBN flake showing a maximum vertical thickness of ~84 nm.

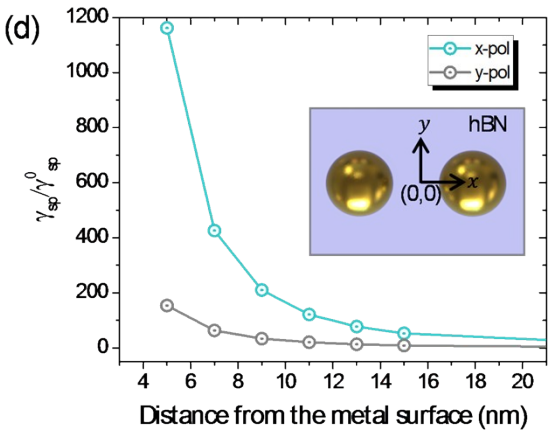

Figure S2. Purcell factor for both x (teal trace) and y-polarized (grey trace) light as a function of the gap distance between two nanospheres. The emitter dipole is assumed to be at the origin and is equidistant from both gold nanospheres. The gap plasmon effect on the dipole source from the two particles diminishes drastically as the metal gap size increases.

Absorption spectrum. Gold nanosphere solution was purchased from nanoComposix. The nanospheres were coated with approximately 5-6 nm thick sodium citrate and were stabilized in aqueous 2 mM citrate solution. The nanoparticles used during this experiment are shown to have maximum absorption at 535 nm as measured with an Agilent Cary-60 UV-Vis spectrometer.

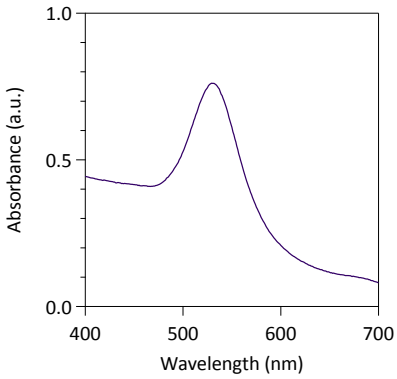

Figure S3. Absorption spectrum of the gold nanospheres solution with absorption peak occuring at 535 nm.

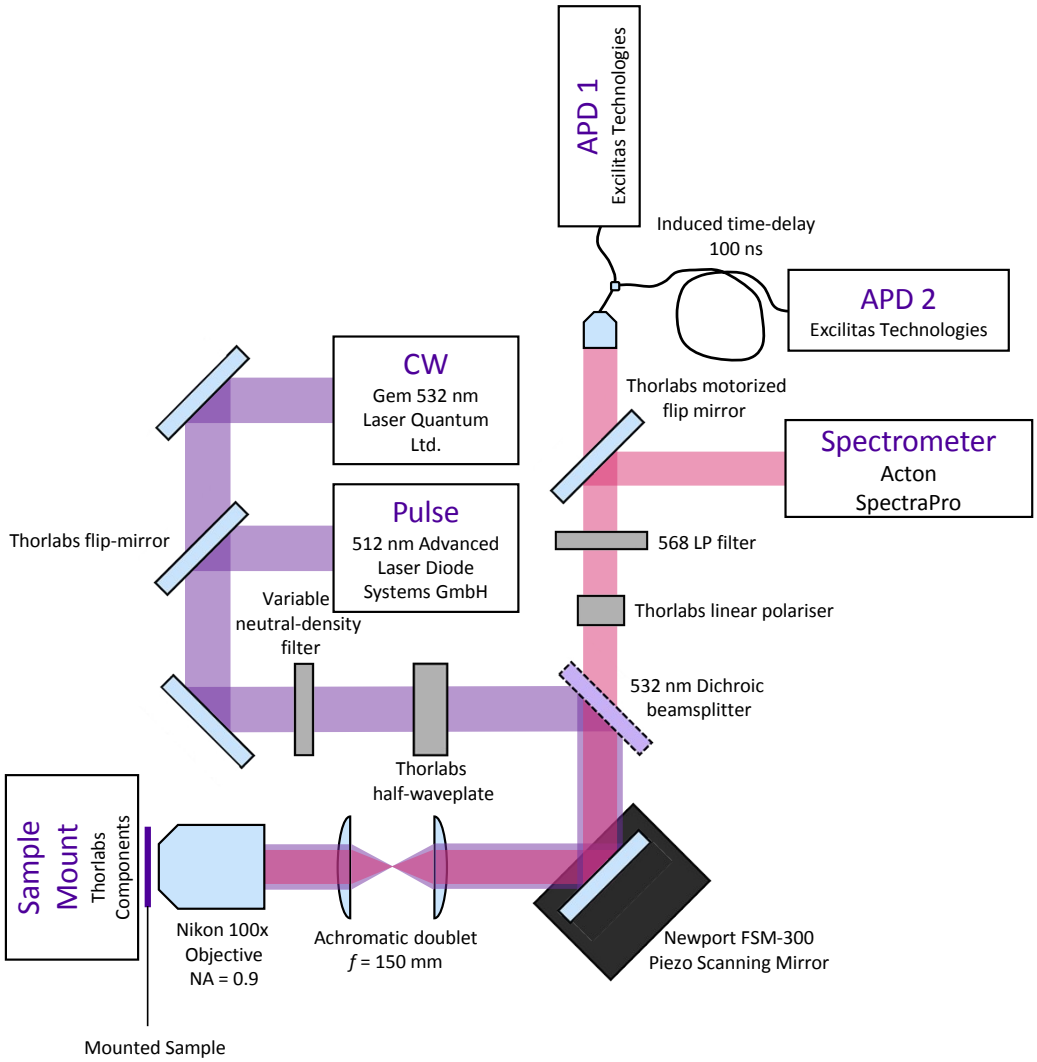

Figure S4. Schematic diagram of the custom-built confocal microscope used for all optical characterizations. Purple path indicates the excitation pathway, whilst the pink path indicates the collection pathway.

Photophysics. The ZPL of the emitter fluorescence was fitted using a Gaussian function. Normalized plots of PL intensity shows the preservation of the spectral signature throughout all coupling processes (Figure S5).

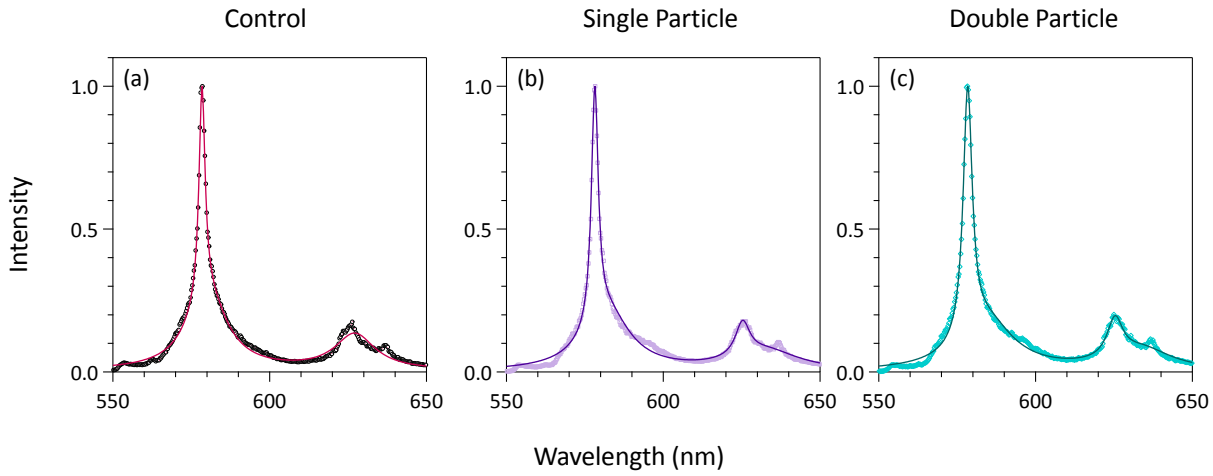

Figure S5. Normalized Gaussian fits of the emitter ZPL with raw fluorescence data showing no deviation of spectral signature during the (a) pristine flake, (b) single particle, and (c) double particle coupling arrangements.

For analysis of the quantum nature of the luminescent defect, the second-order autocorrelation function was used. The framework of emitter analyzed was that of a three level energy system. The antibunched emission was fit to the following equation:

$$g^{(2)}(\tau) = 1 - (1 + a)e^{\frac{-|\tau|}{\tau_1}} + ae^{\frac{-|\tau|}{\tau_2}}$$

where τ_1 and τ_2 designate the radiative transition and metastable state lifetimes, respectively, and a is *a* bunching factor. Pulsing of the laser allows us to examine the radiative emission rate of the emitter. Radiative lifetime measurements were fitted with a biexponential decay function:

$$f(x) = y_0 + A_1 exp\left\{\frac{-(x - x_0)}{\tau_1}\right\} + A_2 exp\left\{\frac{-(x - x_0)}{\tau_1}\right\}$$

where τ_1 and τ_2 represent the instrument response and the emitter lifetime, respectively.