Supporting Information for:

Picosecond Lifetimes with High Quantum Yields from Single-Photon Emitting Colloidal Nanostructures at Room Temperature

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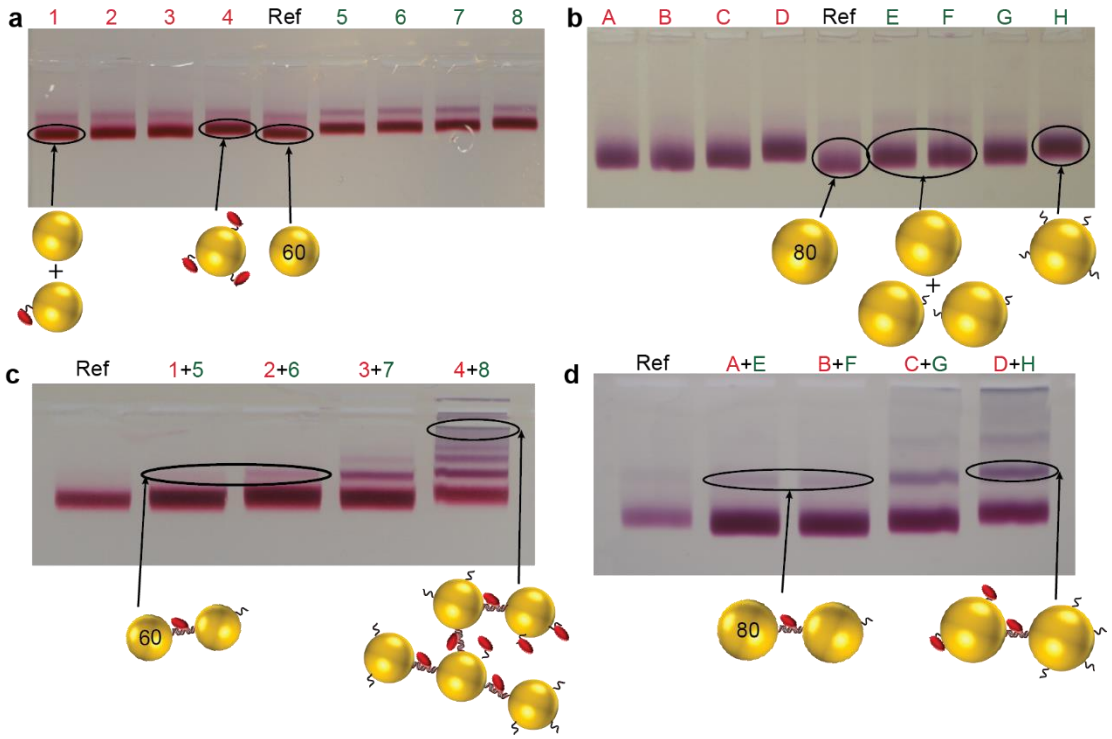

Figure S1. Gold nanoparticle dimer purification. Agarose gel electrophoresis of DNA-functionalized 60 nm (a) and 80 nm (b) AuNPs. The central reference lanes correspond to unconjugated particles incubated with the same amount of NaCl and BSPP before passivation with thiolated / methyl terminated ethylene glycol hexamers. The DNA concentration is progressively increased by a factor of 2 between lanes 2 (resp. 5) and 4 (resp. 8) as well as between lanes B (resp. F) and D (resp. H). The DNA concentration is the same in lanes A and B (resp. E and F). Agarose gel electrophoretic purification of DNA-templated 60 nm (c) and 80 nm (d) AuNP dimers with the different lanes corresponding to the stoichiometric mix of the recovered DNA functionalized particles (as shown in the labels). Schematic representations of the samples corresponding to specific bands are given for clarity.

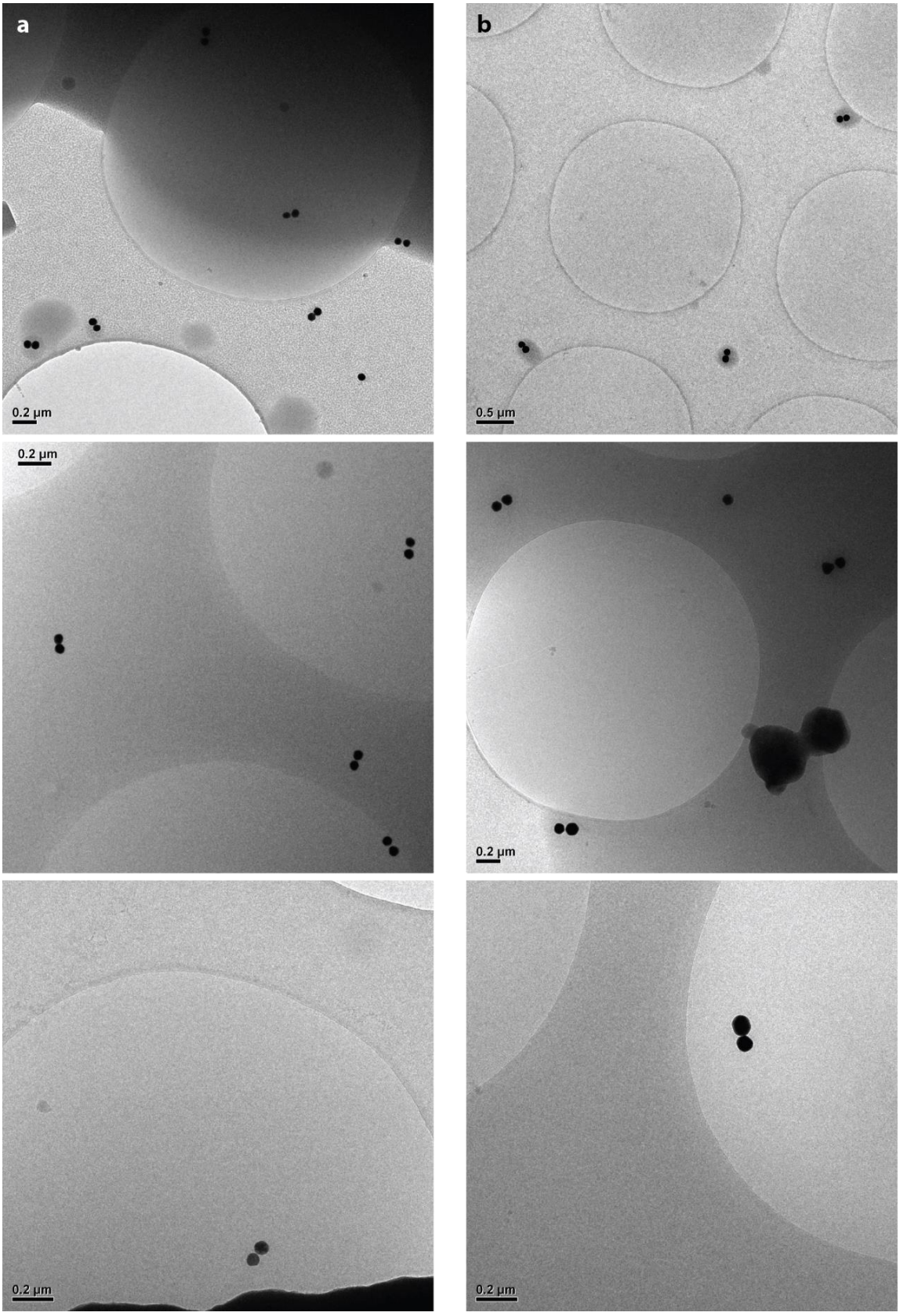

Figure S2. Cryo-EM images of DNA-templated 60 nm (a) and 80 nm (b) diameter AuNP dimers. While several images exhibit three or more dimers and some leftover single particles, most areas of the sample only feature one isolated grouping (bottom images), making the estimation of the sample purity difficult with electron microscopy.

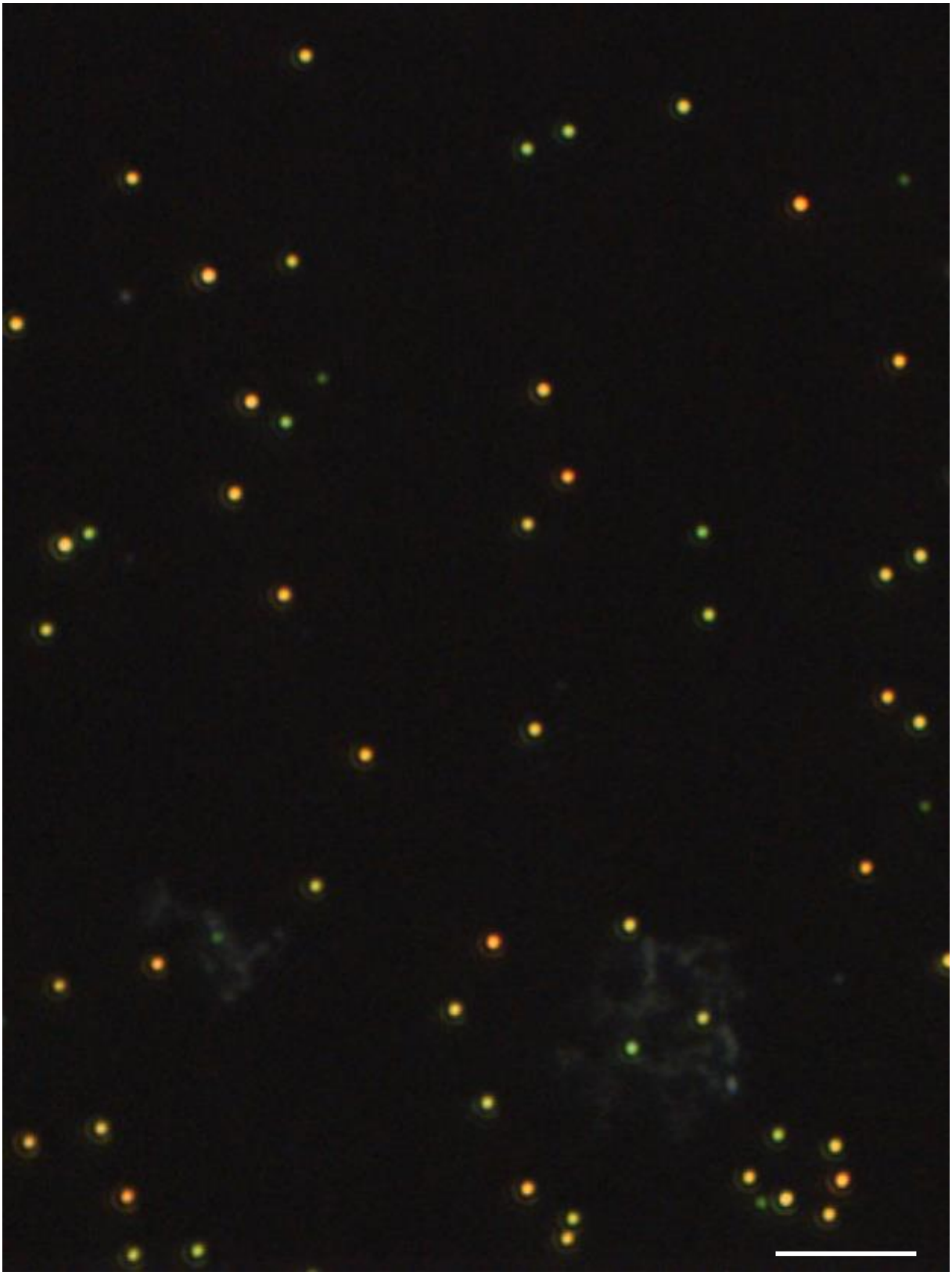

Figure S3. Large-scale darkfield image of 60 nm dimers (bar is 10 µm). On this image, 52 dimers and 5 leftover single particles are visible, corresponding to a dimer purity of about 90 %.

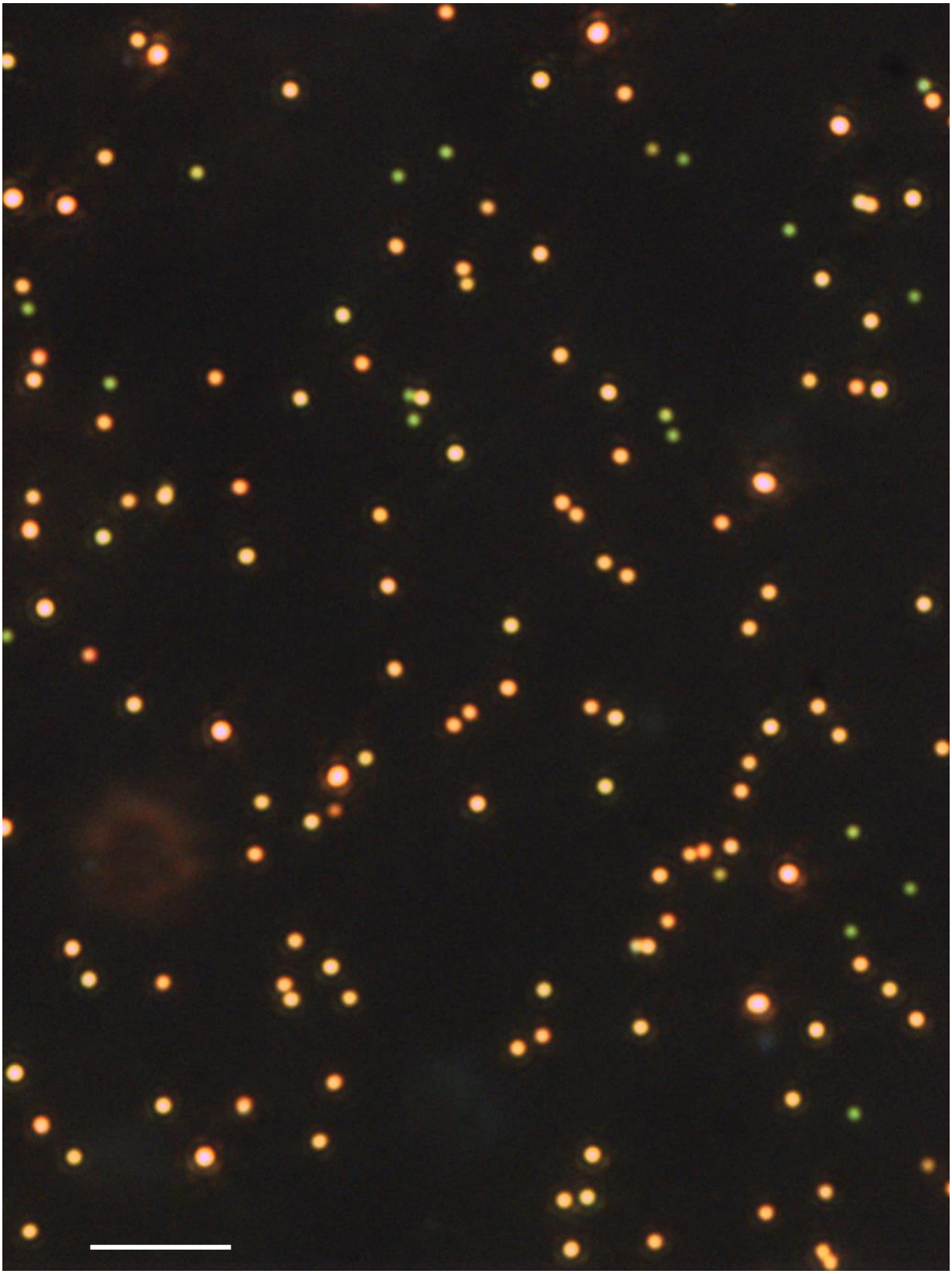

Figure S4. Large-scale darkfield image of 80 nm dimers (bar is 10 µm). On this image, about 115 dimers, 22 leftover single particles and 12 aggregates are visible, corresponding to a dimer purity of about 75 %.

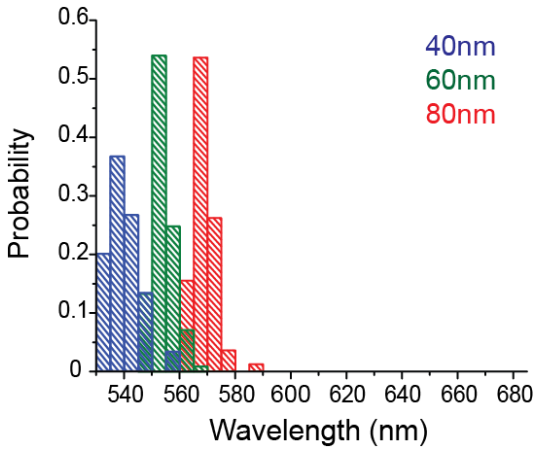

Figure S5. Distributions of plasmon resonance wavelengths for single AuNPs: 40 nm (blue), 60 nm (green) and 80 nm (red) diameters.

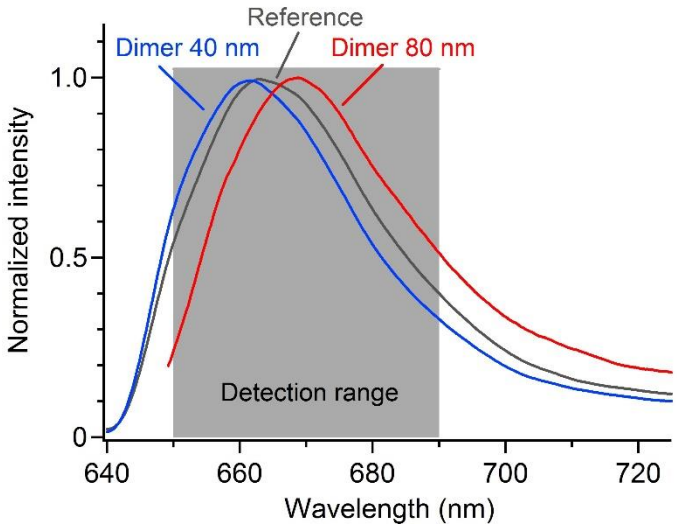

Figure S6. Fluorescence spectra measured with the ATTO647N dye (solid grey line) and the DNA-templated dimer samples with 40 nm (solid blue line) or 80 nm (solid red line) diameter AuNPs. The grey area corresponds to the bandpass filter used for the measurements on freely diffusing emitters of Fig. 2-3 and S5-S6.

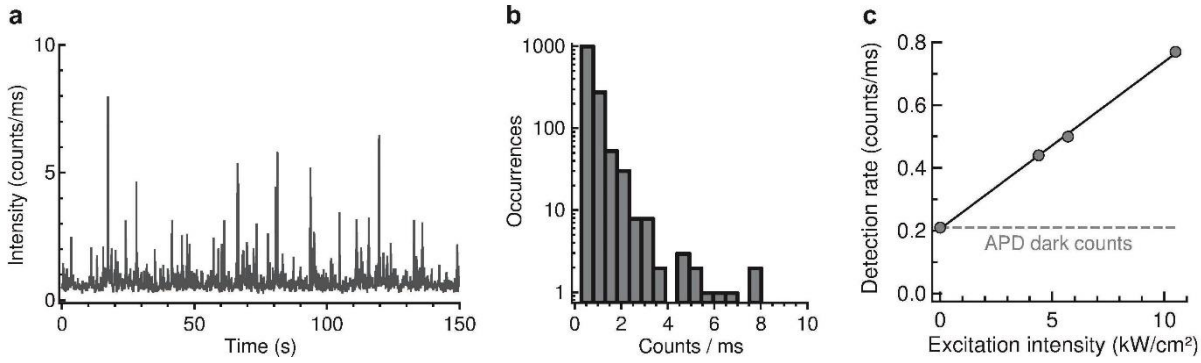

Figure S7. Gold luminescence background in the absence of fluorescent dye for 80 nm dimers. (a) Fluorescence intensity time trace for the dimer antennas of 80 nm diameter without the Atto647N dye (10.5 kW/cm2 excitation intensity at 633 nm, 40 ms binning time). As compared to the conditions of Fig. 3, the excitation intensity has been increased by 3 fold to ease viewing the luminescence bursts. (b) Fluorescence photon count histogram (PCH) deduced from the trace in (a). (c) Evolution of the average luminescence background as a function of the excitation intensity. Contrarily to the fluorescence signals in Fig. 3c, no saturation is observed at high intensities. In all cases, the luminescence background remains negligible (less than 1 %) as compared to the fluorescence signal observed in Fig. 3.

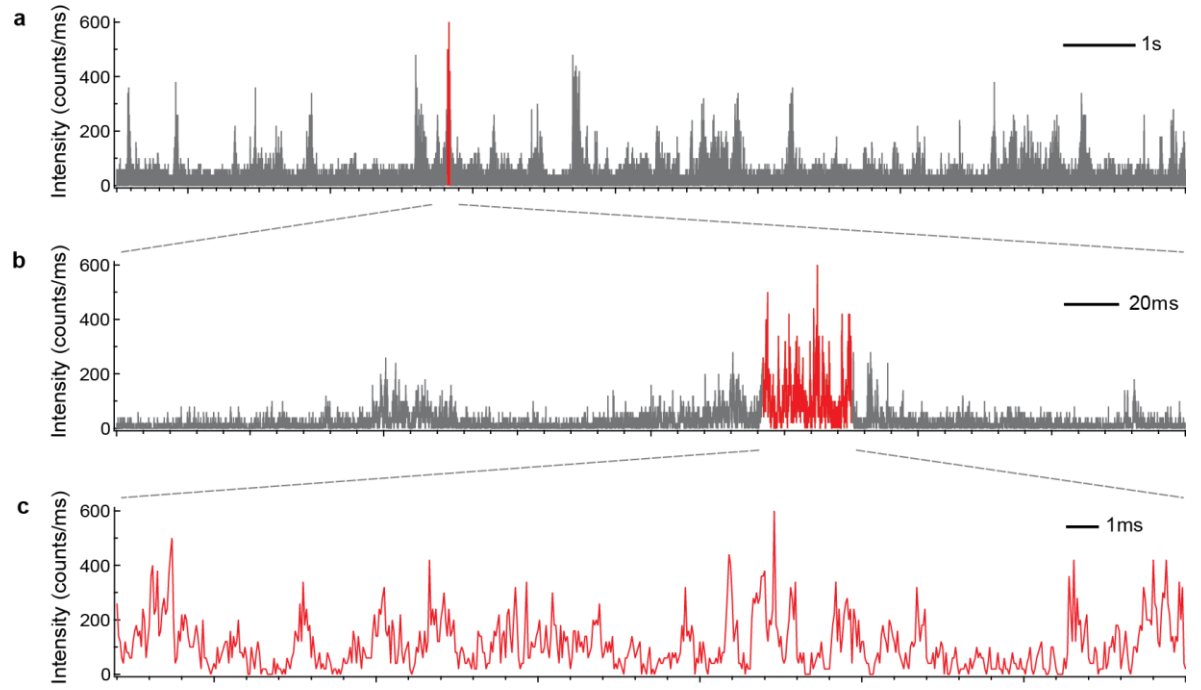

Figure S8. Typical fluorescence intensity time trace for freely moving 80 nm AuNP dimers (a-c). All binning times are set to 50 µs. The excitation intensity is 1.75 kW.cm^{-2}.

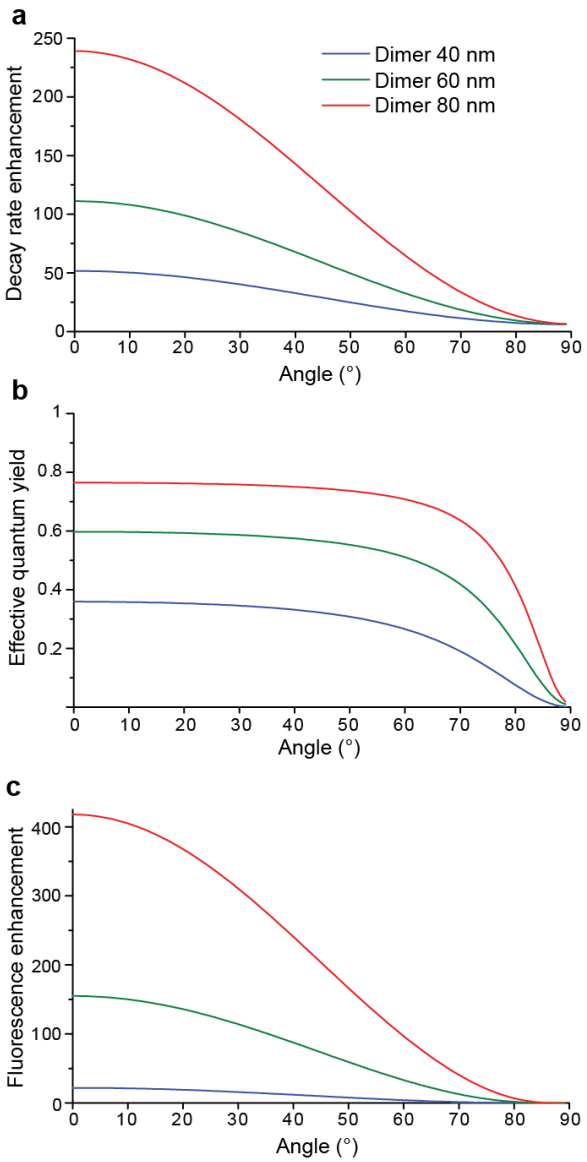

Figure S9. Mie theory calculations: total decay rate enhancement Γ/Γ_0 (a), effective quantum yield of the dye-antenna hybrid emitter (b) and orientation-averaged fluorescence intensity enhancement η_{fluo} (c) as a function of the angle between the fixed transition dipole of the molecule and the axis of the AuNP dimer (considering a 65 % quantum yield and 3.8 ns initial fluorescence lifetime for the ATTO647N dye)**.**

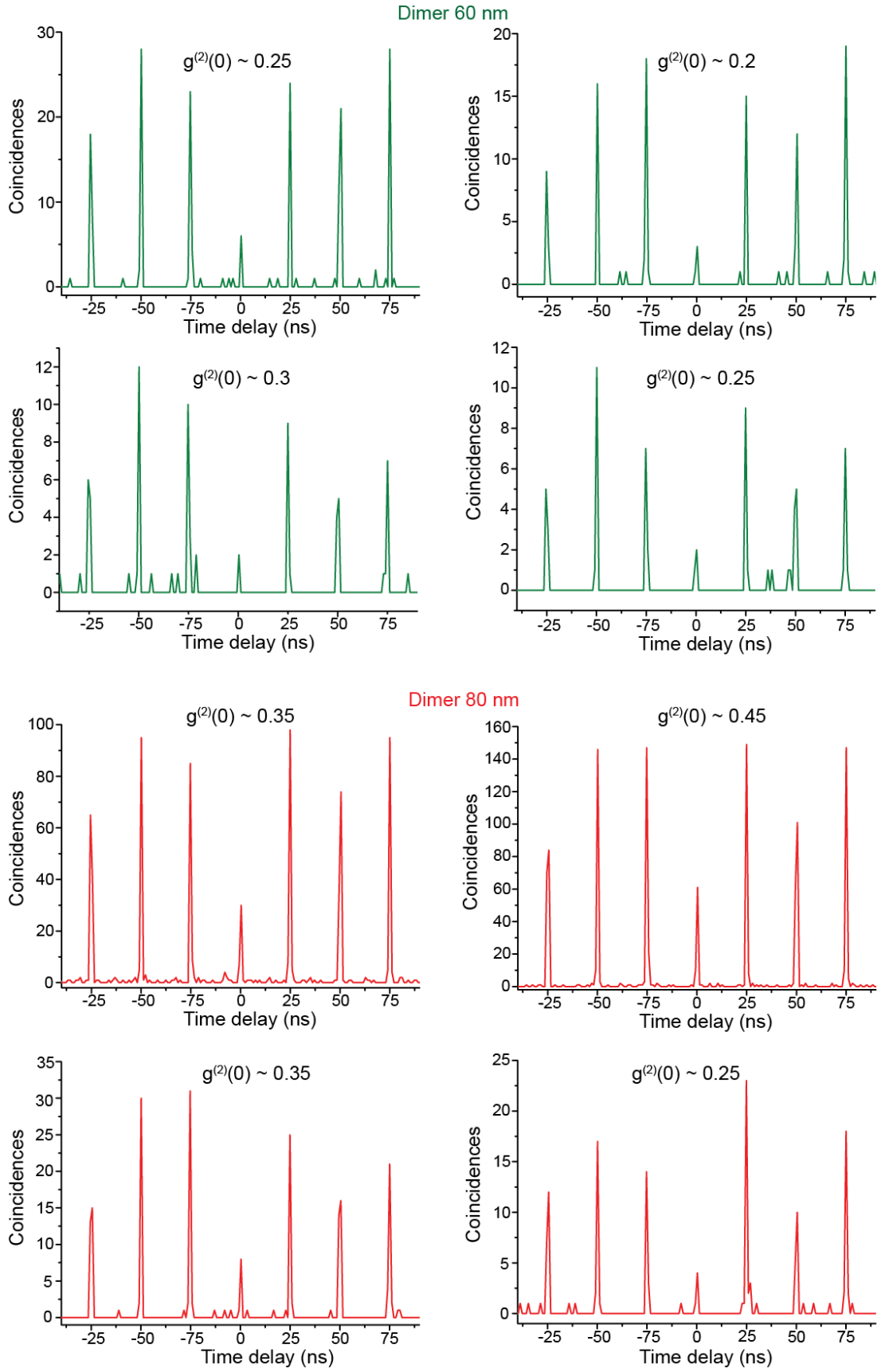

Figure S10. Photon antibunching. Examples of photon emission statistics from single 60 nm and 80 nm dimers with the estimated second order correlation values at zero time delay with respect to the normalized peak heights of consecutive laser pulses.

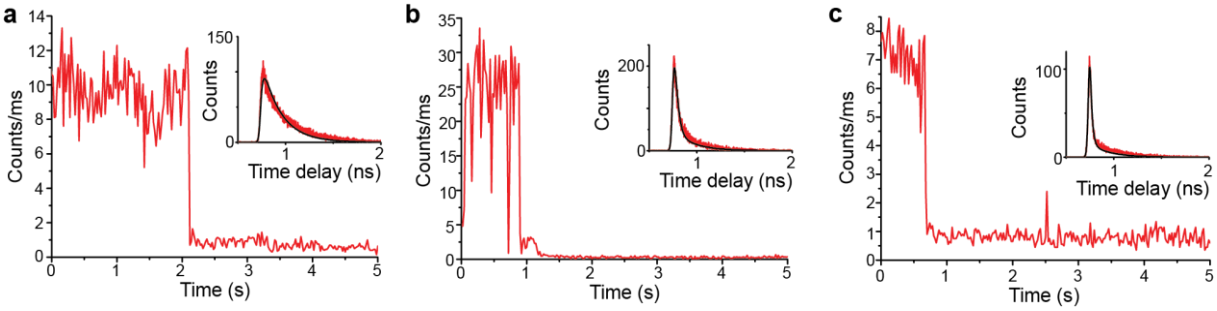

Figure S11. Examples of fluorescence time-traces and lifetimes for individual 80 nm AuNP dimers. The estimated lifetimes are 120 ±15 ps (a), 25 ± 5 ps (b) and 5 ± 5 ps (c).

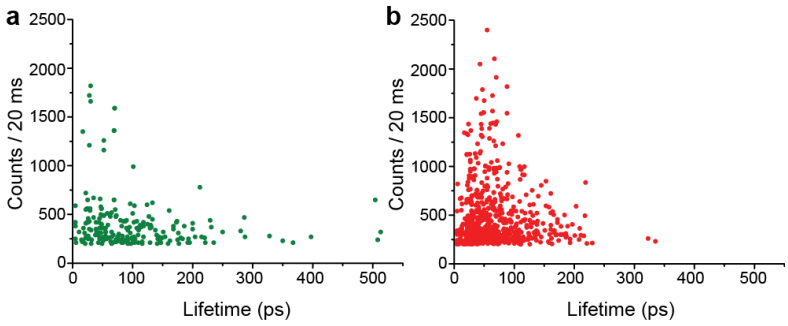

Figure S12. Distributions of fluorescence count rates as a function of the estimated lifetime: for the 60 nm (a) and 80 nm (b) AuNP samples analysed in Figure 4-c of the manuscript (20 ms integration times).