Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information

Single Molecule Confocal Fluorescence Lifetime Correlation Spectroscopy for Accurate Nanoparticle Size Determination†

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Figure S1. Advantage of the FLCS measurement. Comparison of the autocorrelation function calculated by either (a) conventional FCS or (b) FLCS. Two autocorrelation functions are fit by the 3D diffusion model without considering the triplet state, as the excitation power is at or under 4.5 μW and the concentration of Alexa 488 dissolved in water is 5.4 nM. The autocorrelation function from the conventional FCS is not well fit, with χ^2=45.351, whereas the one from the FLCS is well fit, with χ^2=1.006. The corresponding residual curves are shown in the lower panels. The autocorrelation function calculated from the FLCS data effectively rejects measurement artifacts such as detector after-pulsing, especially in the early-microsecond time interval.

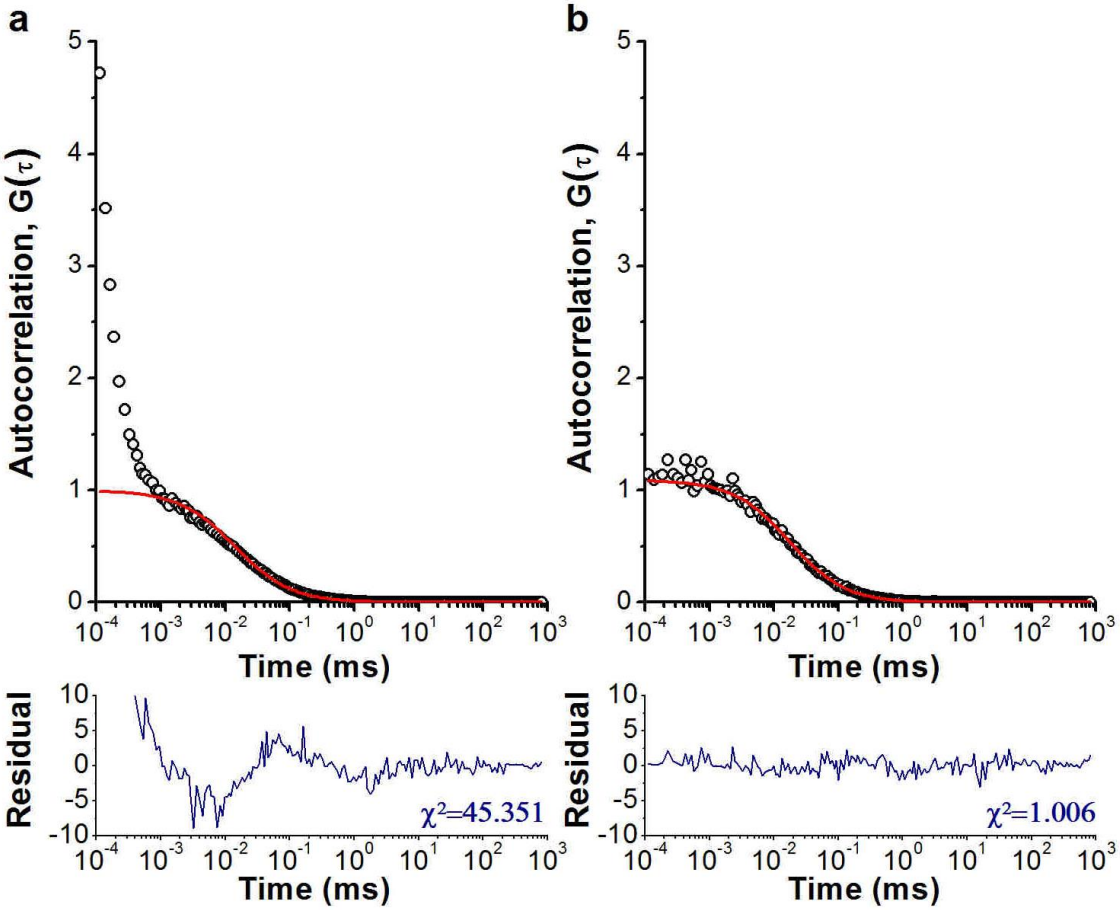

Figure S1

Figure S2. Laser exciation power dependence of fluorescence lifetimes of alexa 488 dyes. The panels from (a) to (f) are results from the laser power (4.5, 11.8, 20.3, 35.1, 39.5, and 41.7) uW, respectively. The black circles are the raw fluorescence lifetime data from FLCS measurements, and the blue lines are the results of the single exponential fit. The resulting fluorescecne lifetimes from the fit are the same for all the excitation intensities within 1 % error, indicative of long lifetime photons possibly involving non-radiative decays were not detected during the bin time of TCSPC acquisition. Corresponding time-dependent residual chi-squared values calculated from each appropriate exponential fit are presented below each lifetime decay plot. The amplitude of the small humps at around 7 ns changed concomittantly with the excitation laser intensity, indicating that they are background excitation photons leaking through a series of fluorescence filters.

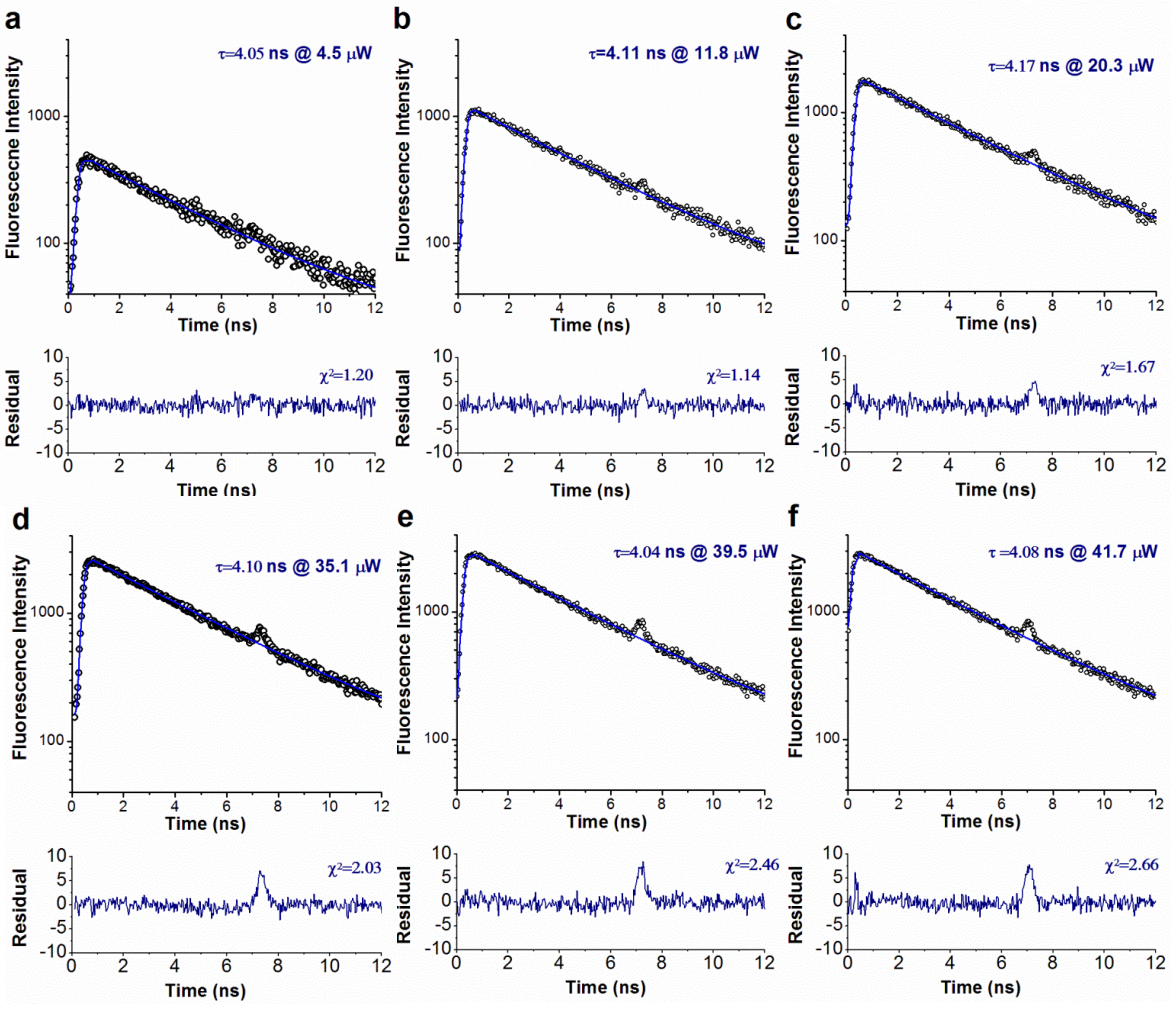

Figure S3. Scanning electron microscope images of nanoparticles of different sizes used in this study.