Supporting Information for: Nondestructive Encapsulation of CdSe/CdS Quantum Dots in an Inorganic Matrix by Pulsed Laser Deposition

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This Supporting Information provides further experimental details on our setups for confocal microscopy and M-line spectroscopy.

Contents

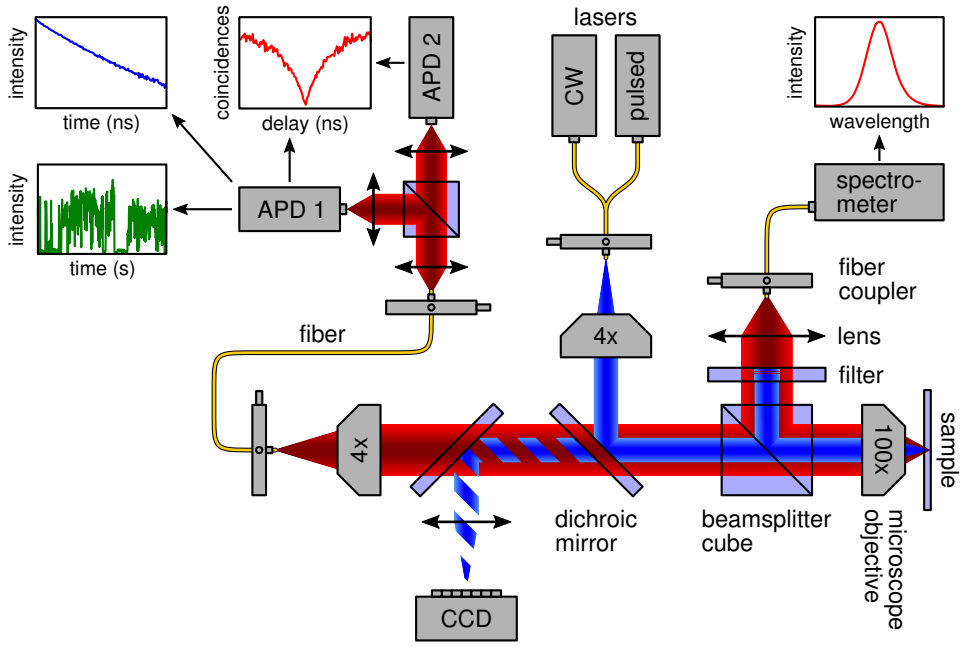

Figure S1: Schematic of the confocal microscope used to acquire luminescence intensity images, emission timetraces, luminescence spectra, fluorescence decay curves, and photon-coincidence histograms of single quantum dots. (CW: continuous-wave, CCD: charge-coupled device camera, APD: avalanche photodiode)

1 Confocal microscopy

The fluorescence properties of single colloidal QDs were studied using the home-made confocal microscope shown schematically in Fig. S1. The excitation laser beam (continuous-wave or pulsed, see below) was focused into a single-mode optical fiber (Thorlabs, P3-460B-FC5), whose output beam was recollimated by a 4× microscope objective and sent into a high-NA oil-immersion objective (Zeiss, Objective A-Plan 100×/1.25 Oil) via a 505 nm dichroic mirror (Thorlabs, DMLP505T). The high-NA objective focused the excitation laser on the far side of a microscopy cover slip, where the QD samples (free-standing or encapsulated) were deposited. 2D confocal images were recorded by scanning the substrate using a closed-loop stabilized XYZ piezostage (PI, P-517-2CD, P-721.CDQ, E725 XYZ-piezostage controller) with nanometer precision. The QD luminescence (centered around 597 nm) was collected through the excitation objective and transmitted through the dichroic mirrors before being focused onto a single-mode fiber (Thorlabs, P3-460B-FC5) for intensity images, or a multimode fiber (Thorlabs, M42L02) for all other experiments; the choice of fiber corresponds to selecting the size of the confocal pinhole, thus allowing to adapt the trade-off between detection efficiency and resolution to experimental requirements. At the output of the detection fibern, the collected light was recollimated and focused with achromatic lenses (Thorlabs, AC254-040-A-ML) on the active surfaces of single-photon counting avalanche photodiodes (APD1: Perkin Elmer, SPCM-AQRH-14; APD2: Laser Components, COUNT-20C). The combination of the two photodiodes with a 50/50 beamsplitter allows to short-time second order correlation function of the QD emission (Hanbury Brown and Twiss interferometer). Residual excitation light passing through the dichroic beamsplitter was imaged onto a CCD camera to monitor the focusing of the excitation laser onto the sample.

Photoluminescence intensity images were recorded under continuous-wave (CW) excitation at 475 nm (argon ion laser, Spectra-Physics, Series 2000) The excitation beam was conditioned with an additional illumination lens (Thorlabs, LA1172-ML) such that a Gaussian profile of

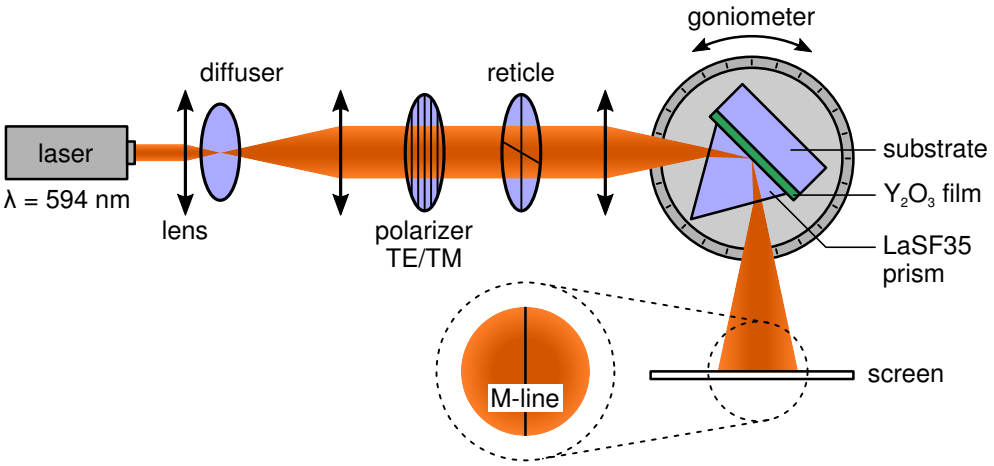

Figure S2: Schematic of our setup for M-line spectroscopy (see text for details).

20 µm full width at half maximum (FWHM) was illuminated on the sample to reduce sample-to-sample variations of the excitation intensity ($\approx 40\,\mathrm{W/cm^2}$, well below saturation of the QDs). Image analysis was performed with a homemade Mathematica program [S1]; the average emission intensity per QD was calculated by summing the detected counts over a square of 5×5 pixels ($500\times500\,\mathrm{nm^2}$ on the sample) around the center of the corresponding luminescence spot.

Fluorescence lifetime measurements were carried out with a picosecond pulsed laser diode (Hamamatsu - C10196, $\lambda = 444\,\mathrm{nm}$, pulse duration 80 ps) using a repetition rate of 2 MHz to accommodate the relaxation of the excited state of the QDs between successive pulses. Time-correlated single photon counting (TCSPC) electronics (Picoquant, TimeHarp 200) allowed to record the arrival times of all detected photons relative to the laser pulses with an internal temporal resolution better than 40 ps. The overall instrumental response function (IRF) was measured at 700 ps. The illumination lens was taken off for TCSPC measurements, and all experiments were performed well below saturation to reduce photobleaching.

Luminescence spectra of single QDs were acquired by inserting a beamsplitter (Thorlabs, BS016) between the objective and the dichroic mirror to send ≈ 50% of the collected light into a mutlimode fiber (Thorlabs, M42L02), which relayed the emission to the entrance slit of a spectrometer (Andor Technology, Shamrock 163). All QD luminescence spectra were recorded with pulsed excitation.

2 M-line spectroscopy

The refractive index and thickness of the Y_2O_3 films deposited under different oxygen pressures were determined with M-line spectroscopy [S2–S4] as illustrated in Fig. S2: An equilateral LaSF35 prism is placed on top of the sample such that a thin air gap remains; this assembly is mounted mounted on a goniometer to allow for precise angle measurements. The 594.1 nm emission of a helium-neon laser is sent through a multimode fiber and a depolarization disc to enhance M-line contrast, then focused onto the base of the LaSF35 prism. The reflected light is observed on a screen, where guided modes launched into the Y_2O_3 film manifest themselves as black lines. Observation of several orders of such modes allowed to determine the refractive index with an absolute precision of 10^{-2} refractive index units and the thicknesses of the film with a relative precision of 2%.

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