Photoluminescence imaging based nano-positioning of single quantum dots for high-performance single-photon generation

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Abstract: We present a wide-field, high-throughput optical technique for locating solid-state quanutm emitters with <10 nm accuracy, and apply it in the creation of micropillar sources with near-optimal single-photon emission.

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Solid-state quantum emitters, especially epitaxial quantum dots (QDs) with large optical oscillator strength, are a promising candidate for future on-chip quantum devices. However, the deterministic creation and eventual scalability of single QD devices greatly suffers from the random nature of the QD positions produced in their self-assembled growth. To address this, a variety of approaches for locating such QDs prior to device fabrication have been reported, and mostly rely on scanning techniques (such as confocal microscopy [1] or cathodoluminescence [2]), resulting in relatively low throughput and long acquisition time. Among different positioning methods, photoluminescence imaging is particularly appealing as it can combine high accuracy, short integration time, and wide-field capability in a simple setup [3, 4]. Here, we present a new high-performance system for nanoscale location of QDs based on photoluminescence imaging. This system exhibits improved positioning uncertainty and shorter acquisition time (3× lower and 100× shorter, respectively) than the first generation setup reported in Ref. [4]. We demonstrate its use in the creation of state-of-the-art single-photon sources, based on micropillar cavities, that emit bright, pure, and Purcell-enhanced indistinguishable photons [5].

In our approach, depicted in Fig. 1(a), a 630 nm LED is used to excite all of the QDs within the system's field of view (typically ≈ 60 μm × 60 μm), while a long wavelength LED simultaneously illuminates the sample. Emitted light from the QDs and reflected light off the sample are directed through filters to reject light from the short wavelength

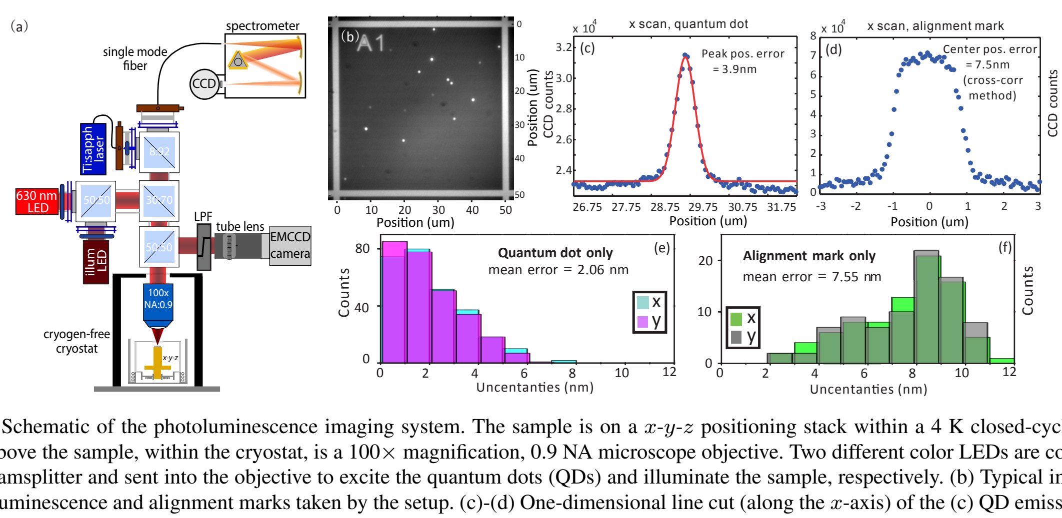

Fig. 1. (a) Schematic of the photoluminescence imaging system. The sample is on a x-y-z positioning stack within a 4 K closed-cycle cryostat. Hanging above the sample, within the cryostat, is a 100× magnification, 0.9 NA microscope objective. Two different color LEDs are combined on a 50:50 beamsplitter and sent into the objective to excite the quantum dots (QDs) and illuminate the sample, respectively. (b) Typical image of the QD photoluminescence and alignment marks taken by the setup. (c)-(d) One-dimensional line cut (along the x-axis) of the (c) QD emission and (d) light reflected off an alignment mark. (e),(f) Histograms of the uncertainties in the QD and alignment mark locations across multiple samples.

LED before going into a sensitive electron-multiplied (EM) CCD camera, or are coupled into a single mode fiber and sent to a spectrometer for spectral analysis. Placing the objective in close proximity to the sample allows for the use of a high NA (0.9), which both increases the solid angle over which emitted photons are collected, and also increases the LED intensity at the sample, ensuring saturation of all QDs within the field of view. Furthermore, the absence of optical windows between the objective and the sample leads to higher quality imaging. A representative image used for positioning is shown in Fig. 1(b), where the acquisition was taken without EM gain over a time of 1 s. Figures 1(c)-(d) show horizontal line cuts through the QD and alignment mark from the image. Through maximum likelihood estimation and a cross-correlation method, uncertainties of 3.9 nm and 7.5 nm in the center positions of the QD and alignment mark have been obtained, respectively. We also show the statistics of our positioning technique in Fig 1(e)-(f), with mean uncertainties for the QDs and alignment marks of 2.06 nm and 7.55 nm respectively.

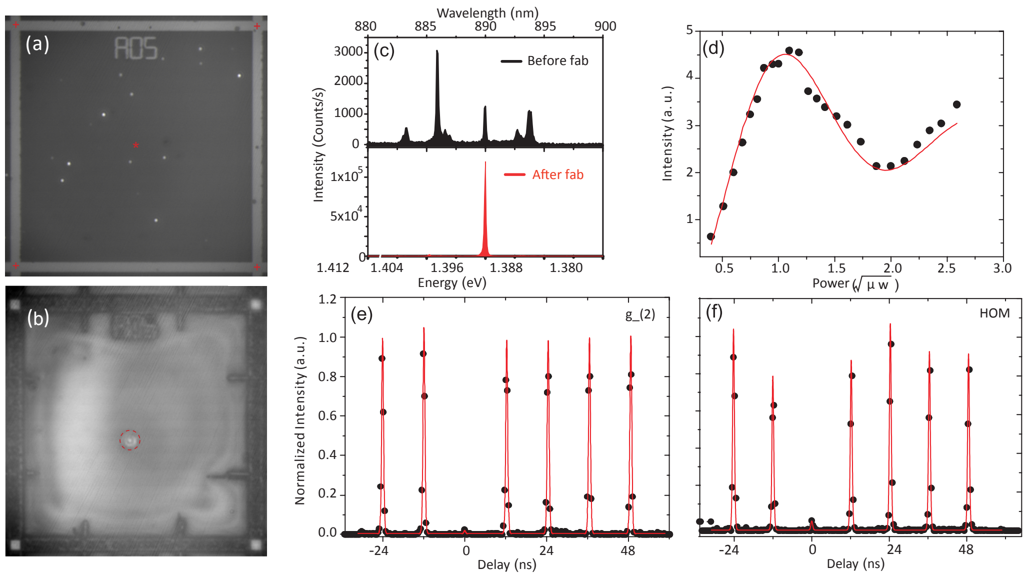

Fig. 2. (a) Photoluminescence image for quantum dot (QD) position extraction. The four alignment mark centers and center of the field are denoted by the red crosses and star, respectively. (b) Photoluminescence image of a micropillar highlighted by the red circle with a single QD in the center. (c) Spectra of the selected QD before (above) and after (below) fabrication of the micropillar. (d) Measured count rate on the spectrometer versus the pulse area of the resonant driving laser field. (e) 2^{nd} order autocorrelation histogram for pulsed resonant excitation with a π-pulse. (f) Histogram of HOM interference with the 12.2 ns delay time. Photons with parallel polarization are prepared here.

We apply our approach to the creation of micropillar cavities that contain a single QD in their centers, for use in triggered single-photon generation. Figure 2(a)-(b) shows the optical images of our device before and after the micropillar etching process. Before fabrication, individual QDs can be clearly identified while only the selected QD is located in the center of the pillar after the dry etching process (this QD was chosen based on its spectral location corresponding to the expected resonance location of the micropillar cavity). A direct comparison of micro-photoluminescence spectra of the positioned QD before and after fabrication suggests that the targeted exciton emission is greatly enhanced by the micropillar cavity structure, as shown in Fig. 2(c). Lifetime measurements (not shown) further support this point, with a radiative lifetime of $\approx$ 100 ps corresponding to a Purcell factor of $\approx$ 7.8. Resonantly exciting the QD with a picosecond pulse, we observe characteristic Rabi-oscillation behavior as a function of the square root of the pump power, which is the key signature for the pulsed coherent driving of the two-level system, shown in Fig. 2(d). Figures 2(e)-(f) present the second-order correlation and Hong-Ou-Mandel (HOM) interference measurements of the emitted single-photon pulses from the fully population-inverted QD excited by π-pulses at a repetition rate of 83 MHz. $g^{(2)}(0) = 0.015 \pm 0.009$, indistinguishability of (98.5 ± 3.2) %, and an extraction efficiency of (49 ± 4) % within the same device confirms an accurate placement of the QD in the center of the micropillar.

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