

# High-throughput nanoscale optical positioning of single quantum dots for high-performance single-photon generation

Jin Liu<sup>1,2</sup>, Yu-ming He<sup>3</sup>, Luca Sapienza<sup>4</sup>, Kumarasiri Konthasinghe<sup>5</sup>, Sebastian Maier<sup>3</sup>, Monika Emmerling<sup>3</sup>, Stefan Gerhardt<sup>3</sup>, José Vinícius De Miranda Cardoso<sup>1</sup>, Jin Dong Song<sup>6</sup>, Antonio Badolato<sup>7</sup>, Christian Schneider<sup>3</sup>, Sven Höfling<sup>3</sup>, Marcelo Davanço<sup>1</sup>, and Kartik Srinivasan<sup>1</sup>

<sup>1</sup>Center for Nanoscale Science and Technology, National Institute of Standards and Technology, Gaithersburg, MD, USA 20899

<sup>2</sup>Maryland Nanocenter, University of Maryland, College Park, MD, USA 20742

<sup>3</sup>Technische Physik and Wilhelm Conrad Röntgen Research Center for Complex Material Systems, Physikalisches Institut, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

<sup>4</sup>Department of Physics and Astronomy, University of Southampton, Southampton SO17 1BJ, UK

<sup>5</sup>Department of Physics, University of South Florida, Tampa, Florida 33620, USA

<sup>6</sup>Center for Opto-Electronic Convergence Systems, Korea Institute of Science and Technology, Seoul 136-791, South Korea

<sup>7</sup>Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627, USA

liujin23@mail.sysu.edu.cn

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

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 ( $3\times$  lower and  $100\times$  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, 6].

In our approach, schematically 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  $\approx 60 \mu\text{m} \times 60 \mu\text{m}$ ), while a long wavelength LED simultaneously illuminates the sample. Emitted light from the QDs and reflected light off the sample are directed through one or more filters to reject light from the short wavelength LED before going into a sensitive electron-multiplied charge-coupled device (EMCCD) camera, or are coupled into a single mode fiber and sent to a grating 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 that

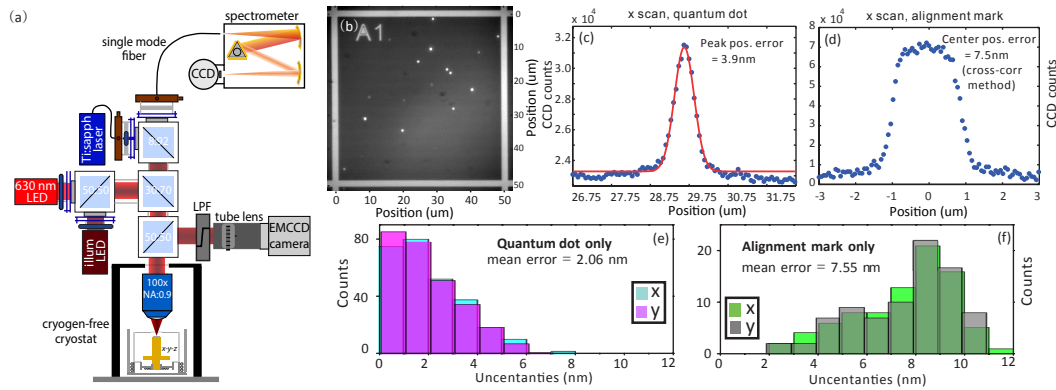


Fig. 1. (a) Schematic of the photoluminescence imaging system. The sample is on a  $x$ - $y$ - $z$  positioning stack within a 4 K closed-cycle cryostat. Hanging above the sample, within the cryostat, is a  $100\times$  magnification, 0.9 NA microscope objective. Two different color LEDs are combined 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.

saturation of all QDs within the field of view can be achieved. Furthermore, the absence of optical windows between the objective and the sample leads to higher quality imaging. One example of an optical image used for positioning is shown in Fig. 1(b), where the image is taken within 1s and without EM gain. 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), where the mean uncertainties for QD and alignment mark are 2.06 nm and 7.55 nm respectively.

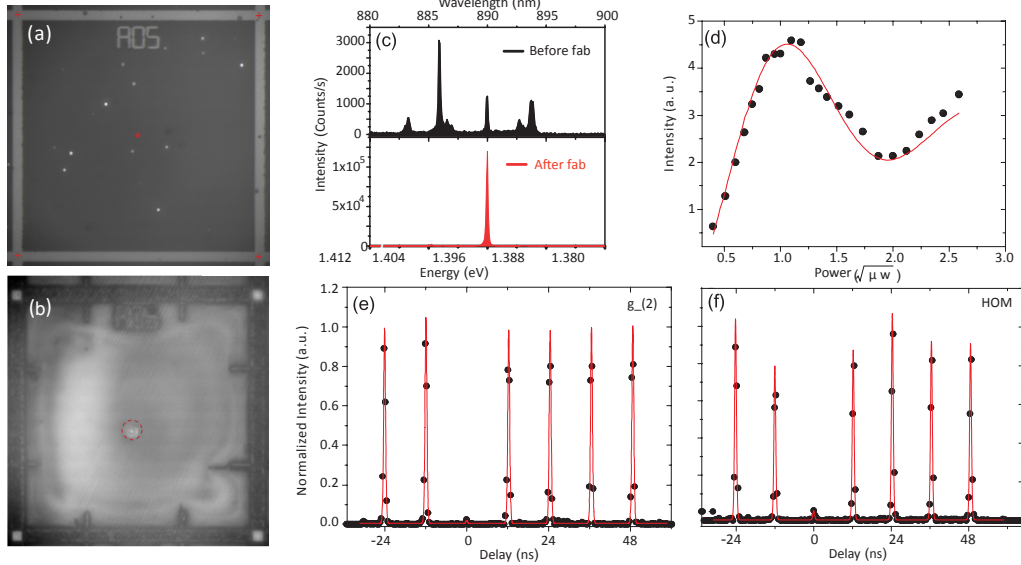


Fig. 2. (a) Photoluminescence image for quantum dot (QD) position extraction. The four alignment mark centers and center of the field are denoted by the red crosses and star, respectively. (b) Photoluminescence image of a micropillar highlighted by the red circle with a single QD in the center. (c) Spectra 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  $\pi$ -pulse. (f) Histogram of HOM interference with the 12.2 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 micro-pillar 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 dot 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  $\pi$ -pulses at a repetition rate of 83 MHz.  $g^{(2)}(0) = 0.015 \pm 0.009$ , indistinguishability of  $(98.5 \pm 3.2) \%$ , and an extraction efficiency of  $(49 \pm 4) \%$  within the same device confirms an accurate placement of the QD in the center of the micropillar.

- [1] A. Dousse, et al., Phys. Rev. Lett. **101**, 267404 (2008).
- [2] M. Gschrey, et al., Nat. Comm. **101**, **6**, 7662 (2015).
- [3] T. Kojima, K. Kojima, T. Asano, and S. Noda, Appl. Phys. Lett. **102**, 011110 (2013).
- [4] L. Sapienza, M. Davanco, A. Badolato, and K. Srinivasan, Nat. Comm. **6**, 7833 (2015).
- [5] X. Ding, et al., Phys. Rev. Lett. **116**, 020401 (2016).
- [6] N. Somaschi, et al., Nat. Photon. **10**, 340-345 (2016).