

# (111)-oriented Stranski-Krastanov Quantum Dots Optimized for Entangled Photon Emission

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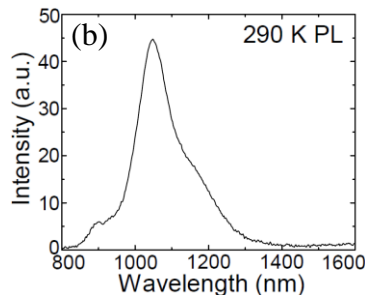
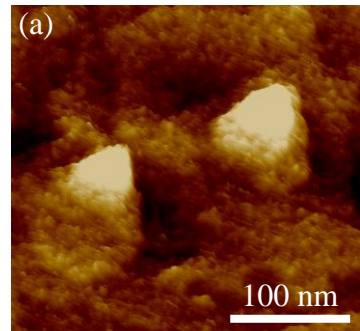
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(111)-oriented quantum dots (QDs) are a promising source for entangled photons due to their high symmetry and the low fine-structure splitting (FSS) between their bright exciton states.[1,2] Therefore, they are of great interest for developing compact, scalable quantum information devices for quantum computing and quantum encryption applications.[1]

We have previously presented results showing the Stranski-Krastanov (SK) growth of (111)-oriented tensile-strained GaAs/InAlAs QDs (TSQDs).[3-5] These TSQDs form as highly-symmetric tetrahedra (Fig. 1(a)) with very low FSS.[4] The use of tensile strain allows for their dislocation-free formation, and reduces their bandgap toward the infrared.[3] Further, TSQDs exhibit clear processing-property correlations, whereby adjusting deposition amount, growth temperature, growth rate, and V/III flux ratio allows us to control QD size, shape, and spectral emission. The resulting roadmap now allows us to optimize TSQDs for specific applications.[5]

Building on that work, here we describe the growth conditions and resultant structural and optical properties of TSQDs optimized specifically for entangled photon emission. We will present a detailed analysis of the structure of individual GaAs TSQDs; power-dependent, temperature-dependent, and time-resolved photoluminescence; and island scaling statistics. We also present experimental results of the reconstruction of the InAlAs(111)A buffer surface. Finally, we will show that we can transform the in-plane shape of the GaAs TSQDs from equilateral triangles to regular hexagons, simply by switching from  $As_2$  to  $As_4$ . The hexagonal TSQDs exhibit improved optical quality and higher symmetry, properties that we expect to be critical for robust quantum entanglement.

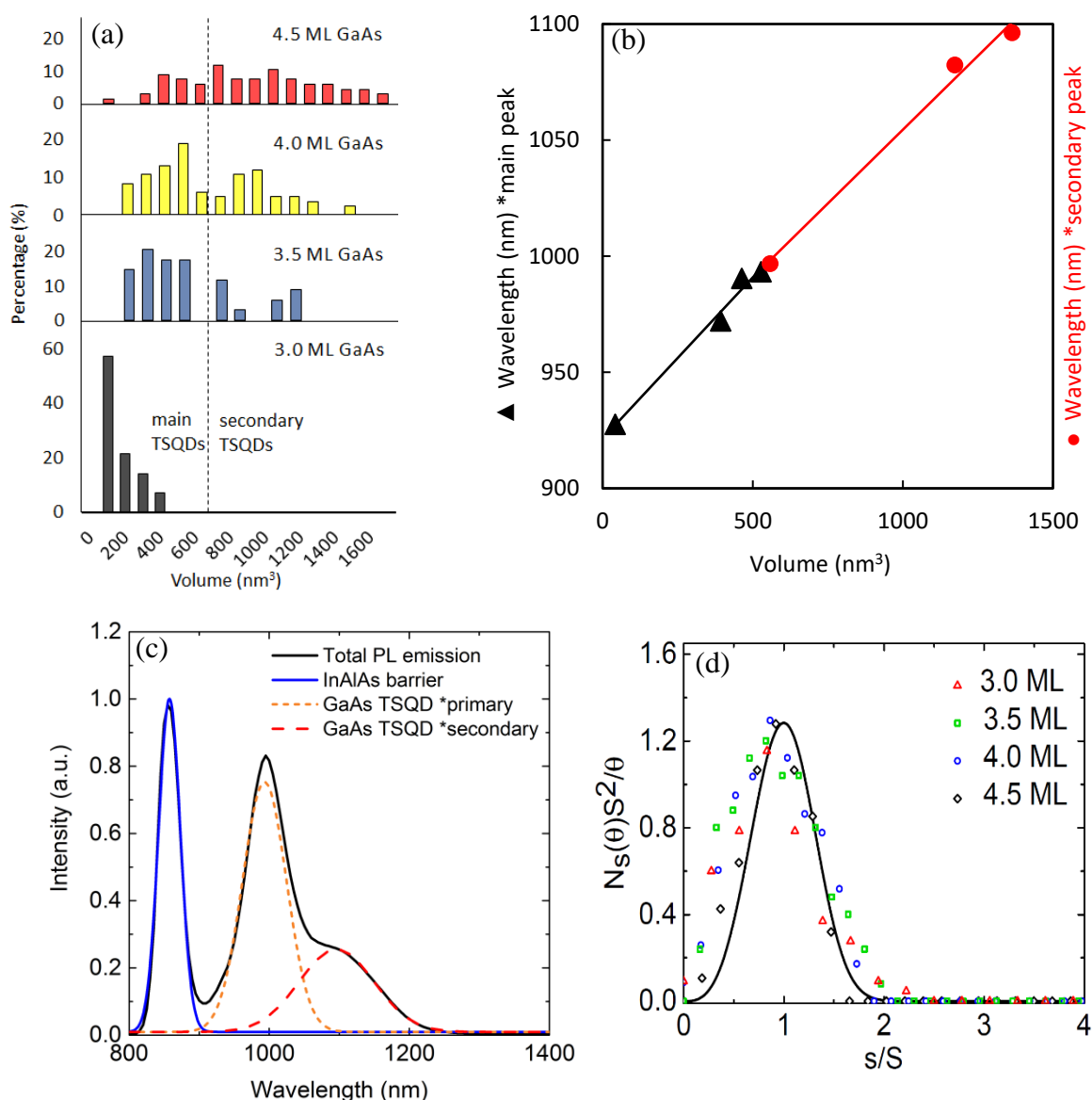


**FIG. 1: (a)** AFM of GaAs TSQDs. Height scale 4 nm. **(b)** Room temperature photoluminescence emission of GaAs TSQDs.

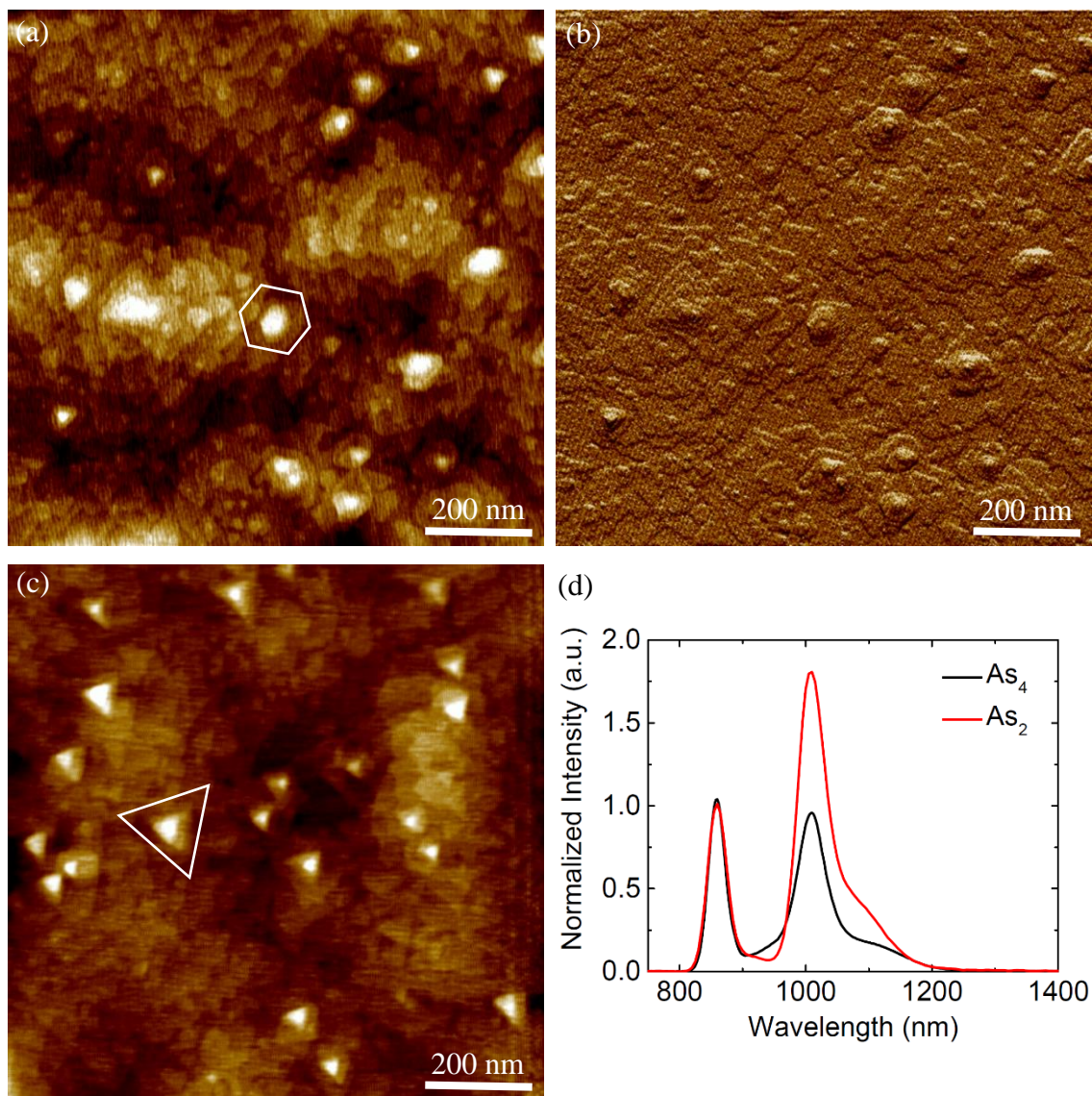
[1] Shields, Nat. Phot. 1, 215 (2007). [2] Schliwa et al., PRB 80, 161307(R) (2009). [3] Simmonds et al., APL 99, 12 (2011). [4] Yerino et al., APL 105, 251901 (2014). [5] Schuck et al., JVSTB 36(3), 031803 (2018).

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## Supplementary information:



**FIGURE 2:** (a) Histogram of TSQD volumes at 3.0 – 4.5 ML deposition. At 3.0 ML, all TSQDs skew toward a single, small population. Increasing to 3.5 ML through 4.5 ML, a secondary population  $>700$  nm<sup>3</sup> (dashed line) appears, and increases in average size and representative percentage, consistent with Ostwald ripening. (b) Plot of TSQD wavelength versus volume. The black line represents emission from the “main” population of TSQDs (left of the 700 nm<sup>3</sup> dashed line on (a)); the red line represents emission from the “secondary” TSQD population (right of the 700 nm<sup>3</sup> dashed line in (a)). The linear correlations are consistent with quantum confinement effects. (c) PL from the 4.5 ML deposition sample, with Gaussian fits for the main and secondary TSQD peaks, corresponding to the AFM results in (a) and (b). (d) Island scaling plot of 3.0 – 4.5 ML deposition, showing TSQD growth is consistent with island scaling theory. At 485 °C, GaAs(111)A TSQDs have a critical cluster size of 4 atoms (black curve).



**FIGURE 3:** (a) AFM of hexagonal GaAs TSQDs using  $As_2$  (4.5 ML deposition, 0.075 ML/s, 75 V/III ratio, and grown at 522 °C). Image  $2 \mu m^2$ , height scale 2.5 nm. (b) AFM voltage amplitude of GaAs TSQDs from (a), providing differential data to showcase hexagonal edges of TSQDs. Image  $2 \mu m^2$ , voltage scale 35 mV. (c) AFM of triangular TSQDs grown under the same growth conditions as in (a) except that we used  $As_4$  rather than  $As_2$ . Image  $2 \mu m^2$ , height scale 2.5 nm. (d) PL comparing emissions from hexagonal ( $As_2$  (a)) and triangular ( $As_4$  (c)) TSQDs. Hexagonal TSQDs have higher symmetry, and also exhibit significantly improved emission intensity. Intensities are normalized to the InAlAs barrier emission seen at 860 nm.