# Self-assembled Low Density Quantum Dot and Quantum Dot-in-nanowire Structures for Quantum Photonics

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**Abstract**— Self-assembled III-V quantum dots (QDs) are of particular attractive as solid quantum light emitters owing to their stability, narrow spectral linewidth, and short radiative lifetime. Meanwhile, semiconductor nanowires (NWs) have appeared as promising building blocks for future nanoscale electronic and photonic devices owing to their high crystalline quality and integration possibilities. To fully explore the potential of NW systems, many investigators have turned to the synthesis of articial nanostructures in NW systems, such as quantum dots (QDs)and nanoclusters,to generate fascinating multifunctional properties. Single nanostructures embedded within NWs represent one of the most promising technologies for applications in quantum photonics. With NWs, a nanostructure can be constructed by inserting a slice of lower gap semiconductor along the growth direction such as CdSe/ZnSe, In(Ga)As/GaAs, GaAsP/GaAs, InAsP/InP and GaAs/AlGaAs systems, or via self-assembled epitaxy on the faceted NWs in the radial direction, utilizing the different surface energies, partly originating from different crystal lattices of hybrid materials, as a driving force. Herein, we report our latest work on self-assembled low density quantum dot and quantum dot-in-nanowire structures for quantum photonics, which might pave the way for the fabrication of highly efficient single-photon sources (SPSs) and novel quantum optics experiments.

NWs samples were grown on GaAs (001) substrates sputtered with 20 nm silicon dioxide or Si (111) with native thin silicon dioxide by a Veeco Mod Gen-II Molecular Beam Epitaxy (MBE) system in traditional VLS growth mode [1–4]. The substrates were dipped for 2 s by 10% HF aqueous solution,and degased at 620°C for 10 min prior to growth.Growth was initiated by the condensation of a nominal 1 nm Ga in the nanocraters of the $SiO_2$ layer. The GaAs backbones were grown at 560–600°C and an $As_2$/Ga flux ratio of 12.5–20. After depositing the GaAs backbones, the samples were exposed to a high $As_2$ ambient overpressure to consume there maining gallium droplets on the top for preferential facet deposition. Self-assembled low density quantum dots were sandwiched between the GaAs core and GaAs/AlGaAs shell and coupled into the fundamental photonic mode of the hexagonal nanowire cavity.

During the epitaxial growth on the facet of NWs backbones, a strain-driven nucleation [1] of gallium-droplets (Figure 1) leads to formation of GaAs branches [2]. Particularly for the InAs QDs embedded branched NWs, the branches were found to preferentially nucleate on the very site of quantum dot as verified by STEM energy dispersive X-ray spectroscopy (EDS) line and spot scans as shown in Figure 2. Micro-PL spectra were measured at 77 K using a continuous wave He-Ne laser for above-band excitation, which is focused on a single NW with the help of white light imaging. An enhancement of $\sim 20$ times with single InAs QD signals from the branched NWs in comparison to those from the straight ones is observed (Figure 3). We attribute it to the combination of quantum confinement effect and QD-cavity interaction. For the higher band offset given by surrounding AlGaAs/GaAs thin film barriers, InAs QD exhibits a stronger confinement of excitons; while the branch helps light gathering together to the top and decreases the optical losses effectively due to its relatively large diameter, which is in consistent with numerical FDTD simulation. Sharp excitonic emission is observed at 4.2 K with aline width of 101 μeV and a vanishing two-photon emission probability of $g^2(0) = 0.031(2)$ (Figure 4). The branched GaAs nanowires turn out to be a better cavity to enhance the extraction efficiency of single InAs QDs emission.

As the most commonly used detectors in single photon characterization, silicon avalanche photodiodes present the best detection efficiency at $\sim$ 700–800 nm. To accommodate this detecting

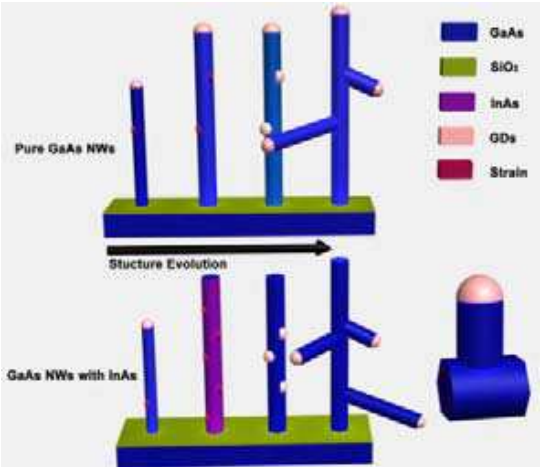


Figure 1: A schematic illustration of evolution mechanism for branched NWs.

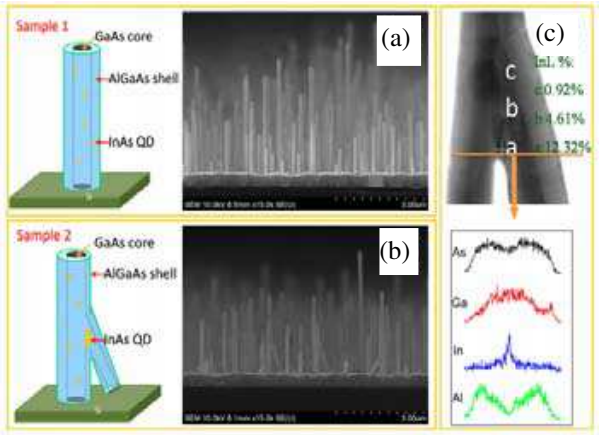


Figure 2: Typical SEM side-view images of both straight and branched NWs, (c) is the corresponding EDS measurement of branched NWs.

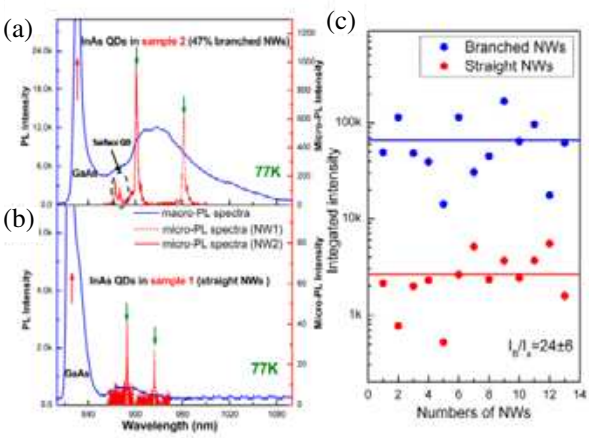


Figure 3: µPL spectra measured at 77 K for both branched and straight NWs.

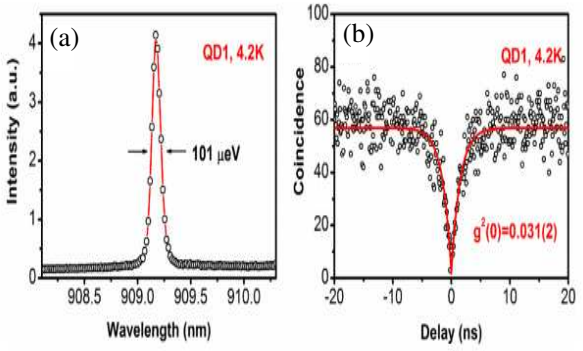


Figure 4: µPL spectra and HBT measurements of a typical InAs QD measured at 4.2 K, respectively.

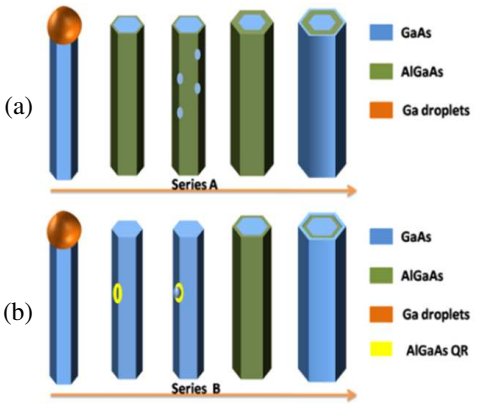


Figure 5: Schematics of the multiple-step fabrication process of GaAs QD-in-NWs.

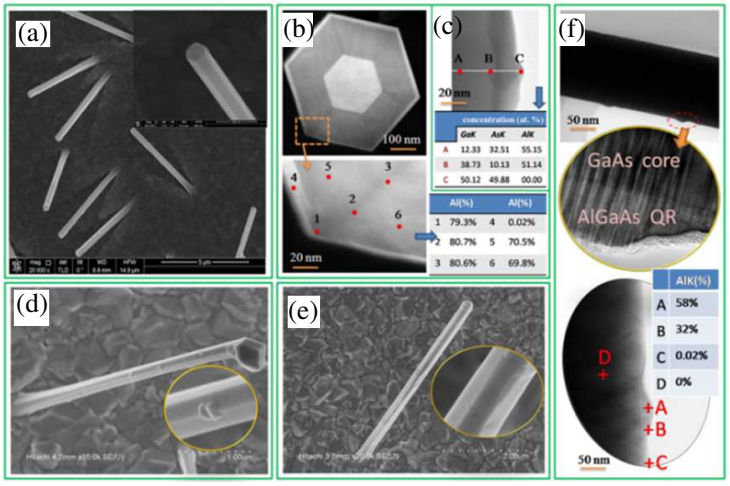


Figure 6: Typical SEM side-viewand cross-section images of uncapped NWs, EDS analysis is given for the AlGaAs QR as shown in (d)–(f).

window and get the best resolved SPSs, we first present a promising self-assembled GaAs QD-in-NW [3] system as growth by the multiple-step process schematized in Figure 5. The structure consists of a GaAs core, which functions as the primary part of the optical cavity, and low density GaAs QDs sandwiched in two $Al_{0.7}Ga_{0.3}As$ barrier layers, which serves as single photon emission source. In addition, single separated GaAs QD can be achieved by using single $Al_{0.58}Ga_{0.42}As$ quantum ring (QR) on the facet of NWs as bottom surrounding barrier and $Al_{0.7}Ga_{0.3}As$ shell as

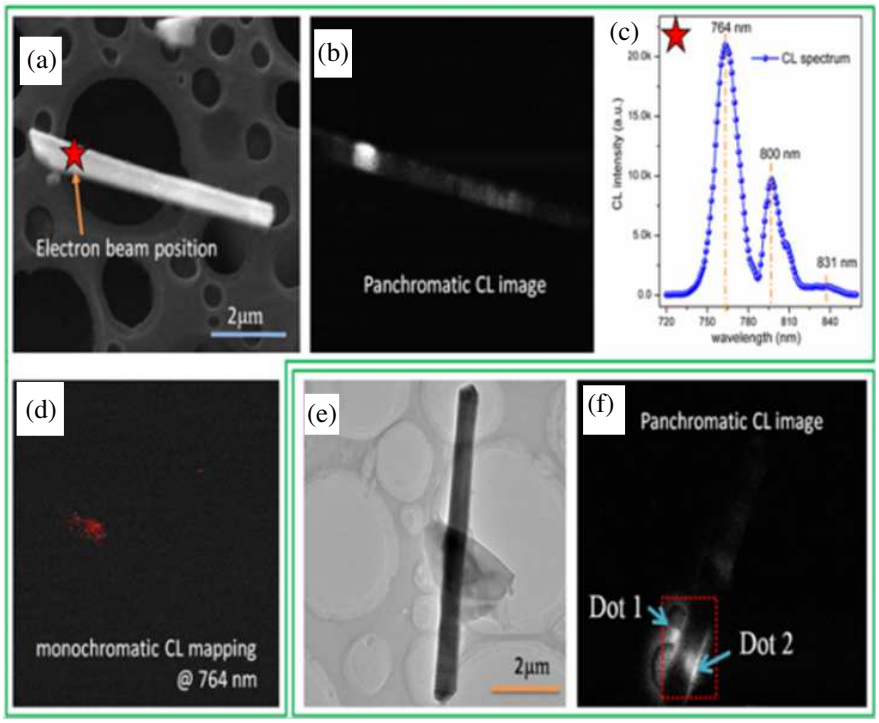


Figure 7: Cathodoluminescence of a single NW.

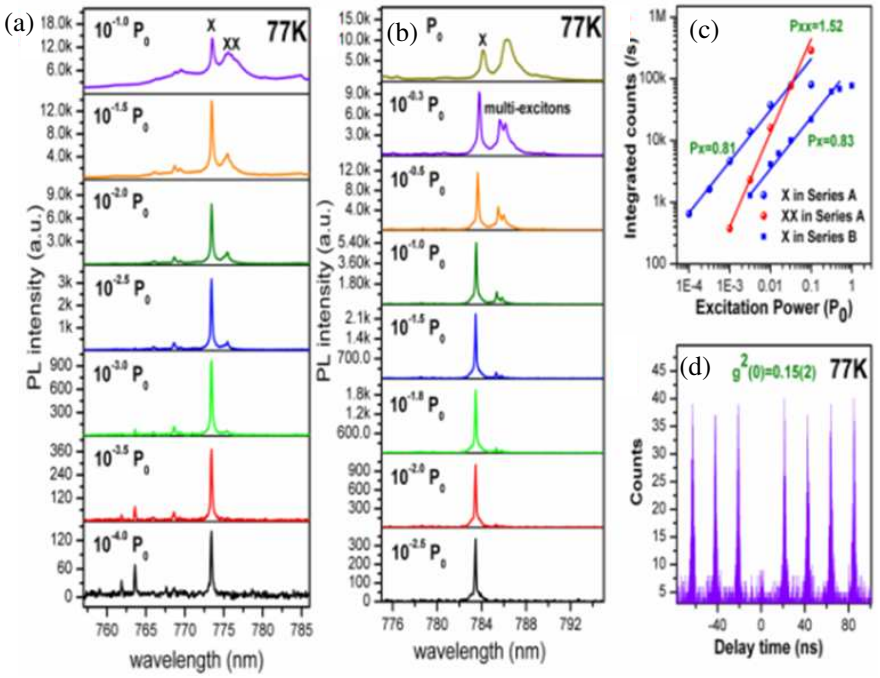


Figure 8: The excitation power dependent micro-photoluminescence spectra and HBT measurements of a typical GaAs QD.

top barrier. Cathodoluminescence measurements at 77 K (Figure 7) demonstrate spatially discrete spots ranging from 700 to 800 nm along the axial direction, indicative of the discrete distributed GaAs QDs. Sharp and enhanced excitonic emission is observed at liquid nitrogen temperature (77 K) with a emission rate of 8 MHz and a two-photon emission probability of $g2(0) = 0.15(2)$ as shown in Figure 8. The smallest linewidth observed so far is 177 μeV while most QDs showing linewidths of sub-200 μeV, and the estimated count rate demonstrates an unprecedented bright SPS even at liquid nitrogen temperature.

As mentioned previously, self-assembly bottom-up approaches cannot avoid the difficulties of its stochastic nature and suffer the random position and density. We adopted a modified droplet-epitaxy for the self-assembly of nanostructure-decorated NWs, based on strain-driven, transport-dependent nucleation of gallium droplets at high temperature (Figure 9). By tuning the deposition temperature, arsenic overpressure and amount of gallium-droplets, we were able to control the density and morphology of the structure, yielding novel single quantum dots, QR, coupled QRs, and nano-antidots (Figure 10). We achieved a single-QR-in-NW structure as shown in Figure 11. The outer and inner side lengths of the QR are about 80–120 and 35–60 nm respectively, with heights of about 3–12 nm. The optical properties were analyzed using micro-photoluminescence

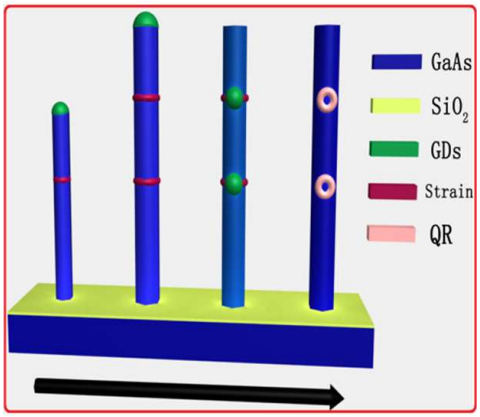


Figure 9: Schematic of the fabrication of the QR-decorated nanowires.

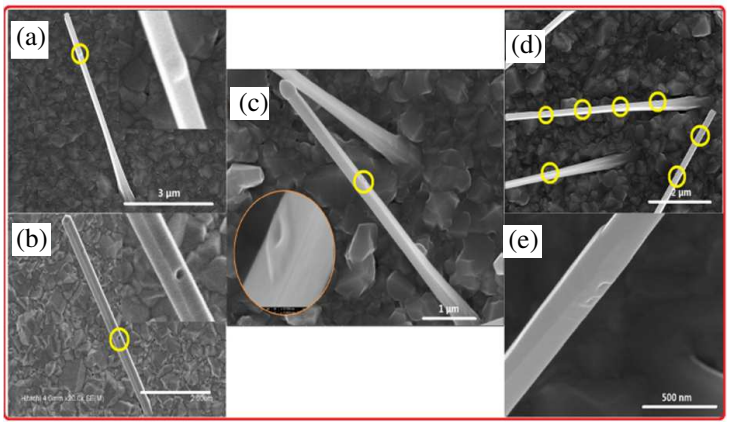


Figure 10: Representative SEM images of the obtained nanostructure-decorated nanowires.

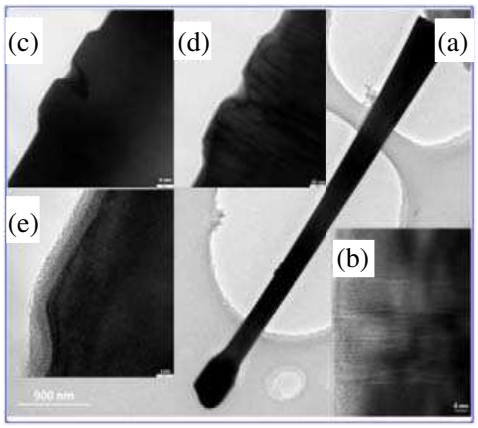


Figure 11: TEM image of a QR-decorated NW over its entire length.

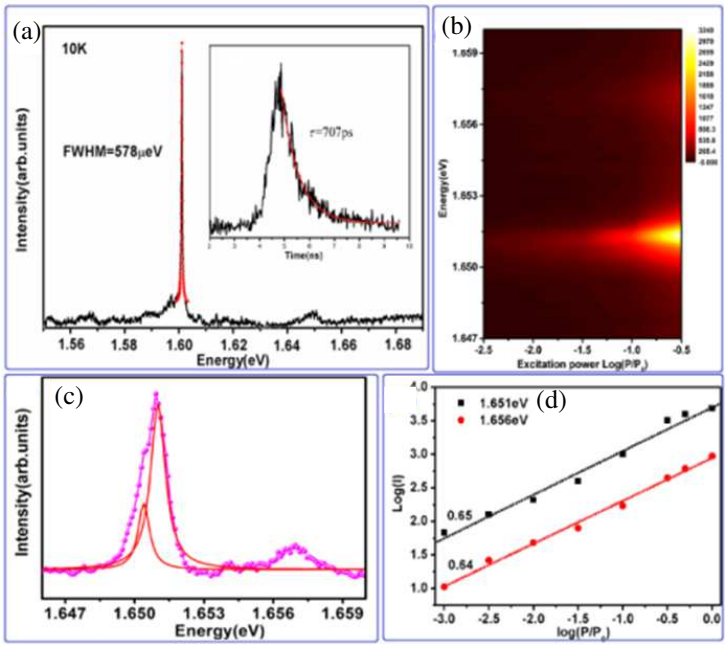


Figure 12: μPL spectroscopy for the single-QR-in-NW samples.

at 10 K (Figure 12) and the spectra show sharp discrete peaks; of these peaks, the narrowest linewidth (separation) was 578 μeV (1–3 meV), reflecting the quantized nature of the ring-type electronic states. This novel strain-driven formation mechanism can be extended to fascinatingly precisely controlling the site of nanostructures on the sidewalls of NWs, if the strain is intentionally introduced at certain sites (Figure 9), on the basis of structural phase control of NWs.

In conclusion, we have demonstrated a detailed investigation on the morphology and optical properties of self-assembled nanostructures decorated nanowires in different material systems and synthesis methods.These new nanostructures may open a new avenue to the fabrication of highly efficient single-photon sources, novel quantum optics experiments, as well as designing robust quantum optoelectronic devices operating at higher temperature required for practical applications.

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