Special Topic: Quantum Physics and Devices of Quantum Dots

Self-assembled semiconductor quantum dots decorating the facets of GaAs nanowire for single-photon emission

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ABSTRACT

In this chapter, we discuss the epitaxial growth of self-assembled quantum dots (QDs) in GaAs nanowires (NWs) and the characteristics of their single-photon emissions. We demonstrate Ga droplet-induced gold-free vapor-liquid-solid growth of hexagonal GaAs/AlGaAs core–shell NWs, branched GaAs NWs and tailored nanostructured morphologies on the NW facets. Particularly, we show two new types of QD-in-NW systems: one is a single InAs QD formed at the corner of a branched GaAs NW, and the other is a single GaAs QD formed on the NW facet. Sharp excitonic emission spectral lines are observed with vanishing two-photon emission probability. Furthermore, a single GaAs QD is achieved at the site of a single AlGaAs quantum ring (QR) on the NW facet. In addition, these NW-based single QDs are in-situ probed and integrated with single-mode optical fibers to achieve all-fiber-output single-photon sources for potential application in quantum integrated networks.

Keywords: self-assembled nanowires, quantum dots, single photon emitters, molecular beam epitaxy

INTRODUCTION

Epitaxial self-assembled III–V quantum dots (QDs), so-called ‘artificial atoms’, are of particular interest for single-photon emission, owing to their stability, narrow spectral linewidth and short radiative lifetime. To reduce their all-space emission and enhance their unit-directional emission and collection (i.e. brightness), in the last few years, several approaches have been pursued. The most successful approach is inserting the QD into a photonic structure, either a microcavity [1–3] or a photonic wire [4–6]. The coupling of a single QD to a confined optical mode of microcavity increases the light-matter interaction, leading to a fast spontaneous emission of QD (the Purcell effect) [7] and the appearance of light-matter-coupled dress states in the strong coupling regime [8]. For example, the QD-micropillar system has been reported as a promising single-photon source that combines features of high efficiency and near-perfect levels of purity and indistinguishably [9–11].

Microcavity is an efficient way to build an interface for photon-QD quantum electrodynamics, but the linewidth of the cavity mode is often too narrow to interact with both the exciton and the biexciton of QD, which challenges the collection of high-rate entangled photon pairs. Photonic wire is advantageous in offering a broadband high collection efficiency [12] and could be applied to spectrally feasible single-photon and entangled photon pairs. Moreover, it has been expected as a promising building block for future nanoscale electronic and photonic devices owing to its high crystalline quality and integration possibility. After embedding a QD in a nanowire (NW), the efficiency of light extraction can theoretically approach 100% [13].

There are two opposite approaches to the fabrication of NW-QDs: top-down and bottom-up. In the top-down approach, a wafer with epitaxial QDs is etched with the exception of nanoscale local areas that include only one or several QDs and form one-dimensional NW-like structures [14,15]. However, the light-extraction efficiency is often lost by not only the fabrication imperfections, but also the...
randomly off-axis positioning of the quantum emitters [15,16]. In the bottom-up approach, QDs are embedded into the NW structure during the growth by changing the precursors via vapor-liquid-solid (VLS) or selected area epitaxy [17–20]. This type of QD-in-NW, which is virtually defect-free, controllable in size and position, and convenient in electrical contact fabrication, in principle, meets the requirement of efficient light extraction.

The most popular configuration for the bottom-up approach to NW-QDs consists of a QD positioned on the NW axis, forming an axial heterostructure in the style of A/B/A to build quantum confinement. This configuration allows an efficient coupling of the emission of the QD to the fundamental waveguide mode of the NW if the diameter and tip of the NW are carefully designed [15]. The mismatch of the refractive indices between the semiconductor NW and the outer environment (i.e. air or vacuum) makes NW a sub-wavelength-sized waveguide, resulting in a broadband enhancement of the QD emission. High-efficiency single-photon emission has been demonstrated from the on-axial QD-in-NWs fabricated using both the bottom-up and the bottom-down methods [15,17]. In addition, electron-hole pairs can be efficiently injected into the QDs, for instance, with optically active on-axial QDs in the intrinsic region of a radial NW p-i-n junction to fabricate NW-QD light-emitting diodes [21].

On-axial precise positioning of NW-QDs is fundamental for strong coupling with the dominant mode of a cylindrical waveguide (HE_{11}). While off-axial (or on-radial) QDs, e.g. self-assembled QDs formed on the facets of core–shell heterostructure NWs according to the relevant surface/interface energy between two different materials [22], can also offer interesting opportunities for single-photon emission. Their emission could couple to other waveguide modes such as peripheral modes or air modes. On-radial QDs still represent a limited research topic compared to on-axial QDs. Therefore, in this review, we focus on the on-radial NW-QDs obtained using the bottom-up method. We will discuss self-assembled GaAs/AlGaAs core–shell hexagonal NWs, GaAs branched NWs and tailored nanostructured morphologies on the NW facet, and show two different on-radial NW-QD systems.

**Self-catalysed GaAs/AlGaAs NWs**

Due to a lower diffusivity of aluminum on the GaAs NW facet, GaAs/AlGaAs NWs are often in core–shell geometry [26]. The AlGaAs shell can passivate the surfaces of the GaAs NW core and quantum structures on it and avoid the surface recombination near QDs; also, their thickness can modify the coupling of QD emission into NW [27–29]. In this way, single-mode GaAs/AlGaAs core–shell NW lasers operating at room temperature have been proposed and demonstrated by several groups [30].

GaAs/AlGaAs core–shell NWs were often synthesized on GaAs (001) substrates in a solid-source MBE system. Firstly, the substrates were sputtered with a 10–15-nm silica layer by magnetron or ion beam sputtering. The substrates were degassed for about 10 min in order to desorb all remnant molecules adsorbed on the surface. The growth began with the condensation of nominal 1–2-nm Ga droplets (GDs) in the pinholes of the silica layer as catalyst, under ultra-low arsenic background pressure (10^{-7} torr in our MBE system). The GaAs backbones were grown at 560°C for 40 min under an As_{4}/Ga flux ratio of 12.5. Then, an interruption for 10 min was introduced under high arsenic atmosphere to crystallize the GDs on the tip before inducing the lateral growth of the NW. The NW shell
consisted of a 30-nm Al<sub>x</sub>Ga<sub>1-x</sub>As layer with Al compositions of <i>x</i> = 20∼70%. Finally, a 20-nm GaAs capping layer was deposited to protect the Al<sub>x</sub>Ga<sub>1-x</sub>As shell from oxidation.

Figure 1a exemplifies the representative side-view scanning electron microscope (SEM) image of the obtained GaAs/AlGaAs hexagonal NWs, with average diameter of 380 nm, length of 5.4 μm and sheet density of 0.03 μm<sup>-2</sup>. Some NWs have hemisphere tips originating from amorphous gallium droplets, indicative of the VLS-growth mechanism, while most NWs show nearly flat surfaces at the tips due to the complete crystallization process. We also grew NWs on GaAs (111)B substrates in the same growth parameters and characterized their cross-section by using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) along with energy-dispersive X-ray (EDX) mapping. Figure 1b shows the HAADF-STEM image. The GaAs core (lighter region) is covered with Al<sub>0.7</sub>Ga<sub>0.3</sub>As (darker region) shell and GaAs (lighter region) capping. They are all in the pure zinc-blende (ZB) phase. The formation of slightly darker stripes at some NW corners indicates an Al enrichment (∼80%), which is also observed in the EDX mapping of Al, Ga, As composition as presented in Fig. 1c. The slight aluminum enrichment is related to the difference in the chemical potential on (110) facets.

**Tailoring nanostructured morphologies on the facet of GaAs NWs**

Among various self-assembled growth techniques, droplet epitaxy is flexible and applicable for both lattice-matched and lattice-mismatched systems to fabricate many peculiar nanostructures, such as QDs, quantum-rings (QRs), nano-antidots, concentric QRs and coupled QRs [31–33]. When integrating such interesting nanostructures into NWs, morphological engineering on both the NW facets and the nanostructures on them is needed. In this section, we will discuss how to tailor the morphologies on the NW facets to form novel hierarchical nanostructures [34] that have extended properties and application potentials in a new technical field.

Figure 2 shows the influence of the arsenic overpressure during annealing on the nanostructure morphology. At a fixed growth temperature, by reducing arsenic flux from (i) 5 × 10<sup>-6</sup> torr to (ii) 3 × 10<sup>-6</sup> torr, (iii) 2 × 10<sup>-6</sup> torr and (iv) 9 × 10<sup>-7</sup> torr, we found that: (i) a QD-like island formed on the facets, with base diameter (height) of about 80–110 nm (10–25 nm) (see Fig. 2a); (ii) a small and shallow ‘circular’ hole emerges in the center of the former QD-like island (see Fig. 2b); and (iii) the hole expands and deepens to form a QR (see Fig. 2c–f). The QRs on the facets of a triangular NW are in perfect ‘square’ geometry, disconnected from the NWs ridges (see Fig. 2c), while the QRs on the...
facets of a hexagonal NW are in ‘rectangular’ geometry, extending along the axial direction and connected with the NWs ridges (see Fig. 2d). The central hole is a well-defined ‘circular’ or ‘square’ shape. The QR morphology is reminiscent of the old Chinese copper coin. Its outer and inner side-lengths are about 90–120 nm and 60–80 nm, with a height of 5–15 nm. Peculiar 3D QRs can be observed in Fig. 2e, with the central hole deep into the NW ridge, with two semicircles on the adjacent \{110\} facets, respectively. Broken QRs in the form of crescent shapes are also formed (see Fig. 2f). On further lowering the arsenic flux to $9 \times 10^{-7}$ torr, the QR seems to disappear and only a nano-hole is left (see Fig. 2g). These nano-antidots are comparable in size and geometric shape to the central hole of the QRs and extend about 10 nm deep into the NW backbone.

We think that the formation of nanostructures on GaAs NW facets consists of two major processes: nucleation of GDs and evolution of GDs into nanostructures. Ga adatoms diffuse along the radial direction of GDs and crystallize preferentially at the boundary of GDs. Meanwhile, the size of GDs shrinks, which defines the inner outline of the nanostructures. Parts of the Ga adatoms diffuse beyond the GD boundary and bond with arsenide adatoms, defining the outer outline, i.e. the diffusion boundary. The dissolution rate depends on the effective amount of gallium at certain sites and thus presents an inverted-pyramid distribution along the radial direction. The evolution of nanostructures during the MBE process relies on the arsenic flux, facet morphologies, number of GDs and deposition temperature. It is similar to plane droplet epitaxy, which is governed by the competition between the migration of group-III adatoms and the speed of crystallization due to the gradient of the surface energy [35–37].

**Strain-driven synthesis of branched GaAs NWs**

In III–V semiconductor NWs, the growth condition can be switched from axial to radial by introducing high group-V elements to crystallize group-III droplets. However, GDs will ‘survive’ if we deposit Ga atoms again at the right temperature, i.e. VLS growth will start on the NW facet by inducing some short branches. Branched NWs may increase structural complexity and enable greater function. Such structures have been explored recently for nano-devices such as nano-LED arrays, logic
circuits, solar energy, surface plasmon interference and micro-optical integration [38–44].

The main challenge of the device application of branched NWs is how to fabricate them with precise control over the dimensionality and composition, avoiding extrinsic contamination. In fact, dislocations are ‘active sites’ for the growth of straight and branched NWs, in which defects might promote additional NW growth [45]. The growth mechanism of self-catalysed branched GaAs NWs is shown schematically in Fig. 3e. During NW growth, strains, in the form of stacking faults and dislocations, accumulate on the facets [46]. This increases as the NW backbone elongates. When the backbones elongate beyond the Ga adatom diffusion length on the {110} facets, the strain sites will be more energetically favorable than the axial growth. Consequently, by intentionally introducing a gallium-rich environment, GDs will preferentially nucleate at the strain sites. Branched NW growth in traditional VLS mode is catalysed by these GDs. More branches will form under more GDs, or more ‘active sites’ related to mismatched InAs/GaAs dislocations.

Transmission electron microscopy (TEM) measurements operated at 200 keV, investigations are also shown in Fig. 3. Here, Sample (I) was grown by 60-min GaAs deposition. Sample (II) was grown by 80-min GaAs deposition and then interrupted for 10 min with the As shutter closed, followed by 20-min GaAs deposition. Sample (III) was grown by 45-min GaAs deposition, then a nominal 15-mL InAs deposition capped by 5-min GaAs, followed by a 15-min GaAs deposition. We found that Sample (I) was straight whereas the others become branched. Along the NWs, mostly ZB phase, some wurtzite (WZ) phase and twin defects were observed. Defects and stacking faults at the crochets imply large strain relaxation, especially in Sample (III).

Yu et al. [47] demonstrated the controllable fabrication of axial superlattice GaAs NWs that consist of ZB/defect-section units by controlling the Triple Phase Line nucleation. As mentioned above, we found that the number and position of branches can be significantly influenced by the number of GD seeds and strain defects on the facets. We believe that the growth of branched NWs with determined position and density can be realized based on our proposed model of controllable defect section and GD nucleation.

BRIGHT SINGLE-PHOTON SOURCES BASED ON QUANTUM DOT-IN-NANOWIRE SYSTEMS

InAs/GaAs QDs in GaAs branched NWs

The growth of InAs QDs straight on GaAs NWs with a sharp interface is an open challenge, since the difference in interface energy in InAs/GaAs NWs
often leads to kinking or branching of GaAs NWs [48–53] and InAs QD prefers to nucleate in the concave region of GaAs NW facets [53–55]. Until now, we have investigated the strain-driven synthesis of self-catalysed GaAs NWs. Decoration of facets with branches is achieved as NWs elongate or with the insertion of InAs. This type of QD-NW system has also been proved to be a better gain medium and cavity than the homogeneous systems [53,56]. In this section, we will discuss the single InAs QD locating at the junction of branched GaAs/AlGaAs core–shell NW, which is featured with higher luminescence efficiency than QD on the facet of straight NW [57].

Such InAs QD at the junction of branched NW, by design, is surrounded by a GaAs/Al_{0.3}Ga_{0.7}As thin film that not only increases the luminescence efficiency by protecting it from the surface state, but also builds a stronger 3D confinement of carriers. Figure 4a shows typical SEM images of branched GaAs NWs with InAs QDs. The Indium composition reaches a maximum at the junction and monotonously decreases away from the junction (Fig. 4b). Figure 4c and d show high-resolution TEM images of a branched NW. The pure ZB phase can be found in both the backbone and the branch of the NW. Specially, three separated diffraction patterns of backbone, branch and QD under fast Fourier transformation (FFT) are reflected at the junction part, where the blue and orange colors are assigned to the ZB crystal structures of the backbone and branch, respectively, indicating that the branch crystal plane is 25° misoriented from the [1-11] growth axis. The green diffraction pattern of the InAs QD whose {111} and {002} spots are splitting from the others reflects the mismatch between InAs and GaAs (~4%). It means that the small InAs QDs present an in-plane compressive strain of ~3.2%. Thus, a clear geometry is figured out that InAs prefers to decorate the facets of GaAs NWs, and some larger InAs QDs along with the accumulation of GDs induce the formation of branched GaAs NWs.

Micro-photoluminescence (PL) measurements are performed at $T = 4.2$ K to further characterize the detailed emission features at the branched NWs. A sharp emission peak of a typical QD (QD1) is displayed in Fig. 5a. The emission peak is observed at 909.2 nm, with an exciton linewidth of 101 μeV. This linewidth is almost comparable to those of QD-in-QWs reported by other groups [20] but larger than those of conventional self-assembled QDs (~4 μeV), which is likely owing to spectral diffusion caused by a fluctuating charge distribution in the vicinity of QDs. The presence of defects often
had a significant impact on the luminescence of nanostructures, which also been reported [58].

Figure 5b shows Hanbury Brown and Twiss-type (HBT) measurements of QD under continuous-wave above-bandgap excitation (~50% saturation level). It shows a pronounced anti-bunching with a vanishing multi-photon probability of $g^2(0) = 0.031(2)$, which confirms that it is indeed a high-purity single-photon source at this excitation power (background counts less than 2%).

We believe such unique InAs QDs embedded in the branched GaAs/AlGaAs NW as natural nanocavities are not only applicable for optoelectronic integration devices, but also useful for other quantum functional devices. For example, the branch with the InAs quantum structure on the top can be considered a quantum channel, where the Coulomb blockade can be periodically lifted as a function of gate voltage, to realize a single-electron transistor (SET); the trunk is considered a classical channel that can be designed to tune the exciton states of the InAs quantum structure [59,60]. In addition, to supply the electron/hole to a positively charged quantum structure, we can introduce an N-type/P-type $\delta$-doping layer lying 2–10 nm below the InAs quantum structure [61,62]. Or we can apply a bias voltage between the top Schottky and back Ohmic contacts (Femi Sea) or an in-plane electric field to control the charging state of the InAs quantum structure [63–65]. Spintronic devices such as spin-SET can be acquired by applying a ferromagnetic (FM) Schottky gate for electrical and optical spin-injection and detection.

**Bright GaAs quantum dot in GaAs/AlGaAs NWs**

In principle, because the interface energy of a Ga-As alloy is less than that of the Ga-As alloy, the NW will be straight when AlGaAs grows on GaAs but will be kinked to form islands or branches when GaAs grows on AlGaAs. It is induced by strain accumulation at the position of the stacking faults in the NW, or at the vertex between the two AlGaAs facets, which is known as the ‘capillarity effect’. M. Heiss et al. [66] reported that fork-shaped AlGaAs QDs can be formed at the apex of a GaAs/AlGaAs interface and perform as a stable and bright single-photon source. By introducing a high growth temperature and annealing process to enhance the mobility of the aluminum adatom, this type of QD will be excluded [67,68].

Another promising QD-in-NW structure is based on self-assembled GaAs QDs coupled to GaAs/AlGaAs NW [18]. The structure consists of a GaAs core, which functions as the primary part of the optical cavity, and low-density GaAs QDs sandwiched in between two Al$_0.7$Ga$_0.3$As barrier layers, which serves as single-photon emission source. The self-assembled QDs nucleate, as designed, on the facet or at the ridge of the hexagonal NW (i.e. on the apex between two adjacent $\{110\}$ facets), which is the favorite for stress release, as mentioned above. The QDs have proven to be very stable and reproducible, and their density can be controlled by changing growth parameters such as the growth rate, growth temperature and arsenic pressure. This QD-in-NW system can efficiently generate bright single photons (with a megahertz count rate) at liquid-nitrogen temperature (77 K) and can be easily coupled into photonic modes of hexagonal NW.

Cathodoluminescence (CL) measurement at 77 K is often used to determine the QD position. As shown in Fig. 6a and b, spatially discrete spots along the NW growth direction are found by both panchromatic and monochromatic CL, indicating that there are several bright, nanoscale emitters, while one bright spot existing in NW (Fig. 6b) represents a single QD luminescence. This is further supported by CL spectrum (Fig. 6c) and monochromatic CL mapping (Fig. 6d), where the 764-nm CL is spatially localized at only an individual spot on the backbone. The sharp emission peak of a typical QD is displayed in Fig. 6e through micro-PL measurement. The brightness of a single QD can be revealed by the integrated count rate at saturation, i.e. about 80 000 counts per second measured by charge-coupled device (CCD) detector. Taking into account the collection efficiency of our experimental setup (~1%), a single-photon flux at the first objective lens of 8 MHz is obtained. This estimated count rate indicates that, even at the liquid-nitrogen temperature, single-photon emission is very bright. Figure 6g shows the HBT-type measurement of a typical QD. It shows pronounced anti-bunching with a vanishing multi-photon probability of $g^2(0) = 0.15(2)$, which...
Figure 6. (a) Secondary electron image, (b) panchromatic CL mapping image, (c) CL spectrum where the electron beam is focused on the star position and (d) monochromatic CL mapping image detecting emission at 764 nm of a typical GaAs/AlGaAs NW. (e) Excitation power-dependent micro-PL spectra of a typical GaAs QD measured at 77 K. (f) Integrated counts of exciton (X), biexciton (XX) as a function of excitation power. (g) Intensity-correlation histogram under continuous-wave above-bandgap excitation using a HBT setup.

confirms that it is indeed a high-purity single-photon source.

In the ‘Tailoring nanostructured morphologies on the facet of GaAs NWs’ section, we mentioned that the size and density of QRs on the GaAs NW facets can be controlled effectively. The QRs here can not only serve as ideal sites for QD isolation and carrier confinement, but also show great potential for the investigation of topological circular symmetry and the Aharonov-Bohm effect.

For radial NW-QD systems, the open challenge is to circumvent the intrinsic limitation of QD random distribution. Herein, Al$_{0.58}$Ga$_{0.42}$As QRs are proposed to locate separate GaAs QDs and provide quantum confinement for carriers. Figure 7a shows a typical SEM image of Al$_{0.58}$Ga$_{0.42}$As QR at the ridges of GaAs NW, with a well-defined central hole. Two symmetrical small projections on the facets of the NW can be found after further growing GaAs QD for 10 min in a low growth rate (see Fig. 7b), indicating that our GaAs QD is well wrapped by AlGaAs QR. As a result, the observed PL spectra exhibit emission lines only from single QDs over a large energy range, as shown in Fig. 7c–f.

IN-SITU PROBING AND FIBER INTEGRATION OF SINGLE QUANTUM DOTS-IN-NANOWIRES

To build a quantum network, from a scientific as well as engineering viewpoint, two necessities have appeared essential to make these individual functionalities deterministic and applicable. The first is to fabricate determinately a spatially and spectrally matched single-photon source device. However, self-assembled QDs usually suffer from their stochastic nature and the broad inhomogeneous distributions of their spectral lines. Encouraging results were obtained in site-controlled QDs embedded in microcavity [69], while P. Senellart et al. demonstrated other pioneering work using far-field optical lithography to fabricate optically [70] and electrically driven [71] single-photons sources with spatial certainty. Herein, our NW itself can serve as an independent photoelectric device without QD-isolation techniques and it also acts as a perfect cavity [72] to enhance the emission rate and collection efficiency of QDs [73].

The second is the efficient integration of the emitted weak ‘single’ photons into commercial single-mode fibers (SMFs), which is essential for quantum networks. The difficulties lie in the ultra-small dimensions of both the semiconductor QDs and the SMFs, ultra-low working temperature and brightness of QD luminescence, which makes the alignment a technical barricade. So far, efforts [74–77] have been made to achieve fiber-coupled output of single-photon sources. In 2010, Tim Schröder et al. [78] demonstrated a fiber-integrated single-photon source by placing a pre-selected nanodiamond directly onto the fiber facet via an atomic force microscope.
Herein, we present in-situ probing and integration of a single self-assembled NW-QD to an optical SMF [79]. The SMF performs as an exciting and detecting probe to search the QD luminescence at liquid-nitrogen temperature. We transfer NWs to the facet of SMFs in situ by simply rubbing the substrate surface. By comparing the PL intensity of single QDs in a confocal microscopy setup and fiber-integrated setup, an efficient coupling of single QD exciton emission into a SMF is proved (Fig. 8a), with collecting efficiency of ~70% over a broadband wavelength. In comparison with air extraction, this increases by a factor of about 7, due to the larger refractive index of the fiber core (Fig. 8b and c). The polarization-dependent PL was studied by placing a polarizer before the entrance slit of a spectrometer. The fiber-output QD emission is strongly polarized, with a visibility of 0.27. To confirm the stability of the adhesive connection of the fiber-NW device, we put the fiber tip in and out of the liquid-nitrogen Dewar several times after a week; great consistency of QD emission was maintained unless scratching the fiber facet directly.

**ZB/WZ GaAs QDs with Short and Long Lifetimes**

As Heiss's work characterized [66], the GaAs QDs are covered by an Al(Ga)As shell and located at the AlGaAs-shell/GaAs-shell interface in a NW (see Fig. 9a). To simulate their emission properties, we solved the 3D steady-state Schrödinger equation for a single particle with effective mass [80]. We found that the emission properties of QD energy levels are sensitive on the Al(Ga)As barrier between the GaAs QD and the GaAs shell outside the AlGaAs shell (see Fig. 9a). Also, they are also affected by imperfections near the QD, such as surface states and misfit defects, resulting in various QD emission properties.

From the simulation, we found some wave-function resonances in the QD, among a continuous energy level distribution in the GaAs shell. Without phonon broadening, the QD emission reaches its intrinsic linewidth that is determined by the carrier tunneling into the QD. The 1D resonant tunneling model was applied to simulate the linewidth of QD emission (see Fig. 9b). The barrier width (\( t \)) and the height (\( V \)) influence the QD emission linewidth greatly. A thick barrier in high energy will provide a small linewidth. From rate equation analysis, QD emission lifetime and efficiency (i.e. intensity) were influenced by the parallel relaxation channel along the GaAs NW band (\( \Gamma_R \)) (see Fig. 9c) as \( t \) increases, QD emission efficiency reduces and NW emission efficiency increases. Compared to a slow \( \Gamma_R (1 \times 10^9 \text{ s}^{-1}) \), a fast \( \Gamma_R (1 \times 10^{10} \text{ s}^{-1}) \) reduces QD emission efficiency more. The QD emission lifetime decreases as the tunneling rate \( \Gamma_T \) increases (or \( t \) reduces). It reduces to 170 ps when \( t = 2 \text{ nm} \), dominated by the intrinsic QD emission.
Figure 8. (a) A schematic illustration of the experiment setup for both micro-PL and fiber-integrating; inset: the transmission efficiency for different ports of WDM. (b) Fiber-output excitation power-dependent micro-PL spectra of a typical QD at 715 nm. (c) Integrated counts of exciton (X), biexciton (XX) in (b) as a function of excitation power. (d) Azimuthal polarization analysis of the QD excitonic emission in (b).
rate $R$ (1/50 ps$^{-1}$). On the contrary, if $t > 4$ nm, $\Gamma_T$ is so tiny that most carriers decay along the $\Gamma_R$ channel, i.e. the QD emission lifetime is of $\sim\Gamma_R^{-1}$. For the GaAs NW shell in high crystal quality, a fast $\Gamma_R$ (1 $\times$ 10$^{10}$ s$^{-1}$) and QD emission are expected; for the GaAs NW shell with many imperfect states for carrier trapping, the $\Gamma_R$ channel will be very slow (e.g. 1 $\times$ 10$^8$ s$^{-1}$), leading to a large QD emission lifetime (e.g. $\sim$ 10 ns).

Semiconductors with an indirect bandgap usually have a long exciton lifetime. We attribute this QD with a long lifetime to WZ GaAs QDs [81]. As we demonstrated, the self-assembled WZ GaAs QDs in the sheath of the WZ NW segments undergo a direct-to-pseudodirect bandgap transition and are suitable for photon detection, whereas ZB GaAs QDs in the sheath of the ZB NW segments exhibit excellent single-photon features. If the ZB/WZ segments can be accurately engineered, we are able to precisely position both the direct bandgap GaAs ZB QDs and the pseudodirect bandgap GaAs WZ QDs within a single NW.

Figure 10a shows a typical PL spectrum of a single NW with many QDs embedded in it, displaying many emission lines distributed in an energy window from 1.72 to 1.96 eV. Time-resolved PL is then used to characterize the dynamic excitonic behavior of individual lines (i.e. single QDs). It is surprising to note that the excitonic dynamic behavior is rather different among various QDs even embedded in the same NW. For example, Fig. 10b shows the exciton decay of two QDs with the lifetimes of 16.63 and 0.27 ns for the emission lines at 1.74 and 1.73 eV, respectively. The short radiative lifetime of 0.27 ns is typically within the range of the direct bandgap emissions of self-assembled QDs and we could assign this emission to direct bandgap transition, whereas the 60-fold longer lifetime indicates the corresponding QD possessing a fairly different exciton recombination mechanism. The exciton lifetime of all measured 39 QDs are given in Fig. 10(c) as a function of exciton energy. It exhibits a Lorentzian distribution with its central peak located at exciton energy around 1.7 eV and QD lifetime spreading over a wide range from 0.19 to 74.46 ns, but reducing to a much smaller range (<3 ns) when the QD exciton energy is away from the central peak. This again illustrates that there are two different optical transition mechanisms in the QD-in-NW system. One type holds a long radiative lifetime and exciton energies always around 1.7 eV and the other exhibits a short lifetime with exciton energies spread over a large range from 1.6 to 1.9 eV.

**FUTURE CHALLENGES**

The recent advances in NW-QD technologies, including tailoring nanostructured morphologies, positioning single QDs, controlling NW phases, and in-situ probing and fiber-integrating of single QDs, make NW-QDs very good candidates for fabricat-
ing the next-generation single-photon sources that are applied in quantum networks. While the source brightness has reached very high values, the indistinguishability of single photons still need to improve. Such control is very difficult to obtain owing to the QD being so close to the NW surface and suffering from spectral diffusion. In this matter, controlling the electrostatic environment of the NW-QD appears critical to shrinking its linewidth. Besides, resonance fluorescence conditions \[82\] can be further introduced to generate a single-photon source near the unity indistinguishability. Additional effort should be focused on tailoring the NW-QD emission to a Gaussian far-field mode to increase the fiber-coupling efficiency. The best extraction efficiency now is reported in on-axial QDs in tailored NWs \[73,83\]. For QDs on the NW facets, whispering-gallery modes (WGMs) in NWs or micro-ring modes in AlGaAs shells must be considered. In conclusion, we highly anticipate that these bright, deterministic, fiber-integrated NW-QD systems will achieve all-fiber-output single-photon sources for quantum photonics in a low-cost but effective and flexible manner. Furthermore, if the ZB/WZ segments can be accurately engineered, we are able to precisely position both the direct bandgap GaAs ZB QDs and pseudodirect bandgap GaAs WZ QDs, i.e. both quantum photon emitters and detectors in a single NW, to achieve the generation, detection and manipulation of single photons within a single photonic wire.

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