Single-Photon Emission from GaAs Quantum Dots Embedded in Nanowires *

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A highly efficient single-photon source based on a semiconductor quantum dot (QD) is a promising candidate in quantum information processing. We report a single-photon source based on self-assembled GaAs QDs in nanowires with an extraction efficiency of 14%. The second-order correlation function $g^{(2)}(0)$ at saturate excitation power is estimated to be 0.28. The measured polarization of QD emission depends on the geometric relations between the directions of PL collection and the long axis of nanowires.

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Efficient solid-state single-photon sources are building blocks for quantum communication and quantum information processing.[1] Semiconductor quantum dots (QDs), known as artificial atoms, have discrete energy levels, high emission rate, narrow emission lines and high fidelity anti-bunching, considered as a candidate of single photon sources. However, the extraction efficiency of QDs embedded in a high refractive index bulk semiconductor is very low, typically, ∼1%,[2] which limits their applications. With decades of efforts to incorporate QDs into a cavity by using standard semiconductor growth and processing techniques, brightness of QD emission can be highly enhanced.[2−5] Recently, self-assembled QDs embedded in nanowires (NWs) pave the way to obtain a high extraction efficiency due to inherent one dimensionality, low surface roughness and, in principle, the ability to operate above and below the diffraction limit.[6] Furthermore, NWs are recognized as one of the essential building blocks for quantum photonics and provide the possibility of integration of advanced functionalities into on-chip technology because they are confined in two dimensions, allowing electrons, holes or photons to propagate freely in the third dimension.[7] Embedding QDs into an NW combines the advantages of both worlds and exhibits excellent single-photon features.[8,9]

In this Letter, we report a highly efficient single-photon source based on GaAs QDs in NWs. The single photon emission rate is up to 1.93×10^5 counts per second under pulsed excitation and the second-order correlation function at zero delay time, $g^{(2)}(\tau=0)$, is 0.28. Single-photon extraction efficiency is estimated to be 14%.

The QD-in-NW structure was synthesized on the GaAs (001) substrate by using the traditional self-catalyzed vapor-liquid-solid process via a molecular beam epitaxy (MBE) system as reported in Ref. [10]. The GaAs QDs were sandwiched in two $Al_{0.7}Ga_{0.3}As$ shell layers along with an inner GaAs core and an outmost anti-oxidation GaAs shell. The nominal diameter and average length of NWs are 380 nm and 5.4 μm, respectively. The long axis of the NWs is along the GaAs [111] direction. A confocal microscopic spectral system was used for the photoluminescence (PL) spectra, time-resolved spectra and $g^{(2)}(\tau)$ measurements. The sample was mounted inside a helium-free cryostat providing a temperature of 6 K. Excitation was performed with a cw or a pulsed semiconductor laser (repetition frequency of 80 MHz and a nominal pulse width of 63 ps) at a wavelength of 640 nm. The PL of the samples was collected through a 50× objective (NA: 0.5). The laser beam is normal incident to the sample substrate and the as-grown NW is along with the GaAs [111] direction, as shown in Fig. 1(a). For comparison of the detected QD emission intensity, the NW is removed and is put onto the GaAs substrate, as shown in Fig. 1(b). The PL was dispersed with a 0.5 m monochromator and detected with a silicon charge-coupled detector. Silicon single photon counting modules were used for time-resolved spectra and $g^{(2)}(\tau)$ measurements.

Figure 2(a) shows the evolution of QD emission lines with increasing the excitation power from 0.06 to 10.6 μW. The spectra exhibit a sharp excitonic line (X) at 1.885 eV and a bi-exciton line (XX) at 1.881 eV. The linewidth of X line is ∼304 μeV at an excitation power of 0.06 μW and increases slightly in response

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to the excitation power. As reported on other QD-in-NW systems,[11] the emission linewidth is usually larger than that of the conventional self-assembled single QDs embedded in bulk semiconductors, which is only a few μeV.[12] A broader linewidth of QD-in-NW probably results from the spectral diffusion of the carrier associated with the surface states and imperfection of the crystal structure.[13,14]

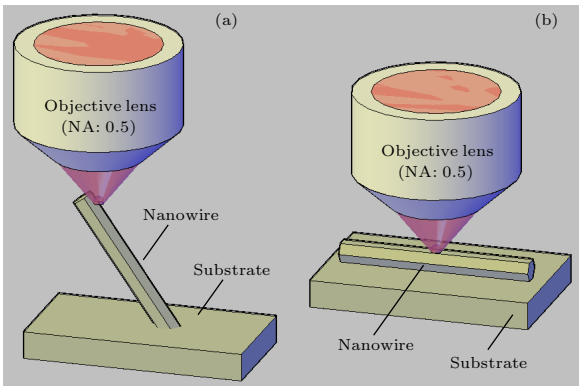


Fig. 1. (Color online) Schematic diagram of directions of the PL collection and the long axis of NWs studied. Here (a) and (b) show their configurations for the tilt NWs and the NWs placed on the substrate surface, respectively.

Figure 2(b) shows integrated intensities I_X and I_{XX} of X and XX with respect to the excitation power in double logarithmic curve, respectively. The slopes of X and XX emissions are 0.99 and 1.85, respectively, exhibiting exciton and bi-exciton emission properties. Here I_X and I_{XX} at the saturation power are approximately equal to each other, indicating a nearly perfect radiative yield of the embedded source having broadband collection efficiency.[8] The decrease of emission intensity at the higher excitation power is probably due to the fact that electrons in QDs are scattered into the wetting layer via phonon scattering or between electrons. To confirm the single-photon emission property of the X line, we measured the second-order correlation function of X emission at an exciton power of 10.6 μW (the excitation power is well above the saturate excitation power) as shown in Fig. 2(c). The anti-bunching dip at zero delay time shows a signature of single-photon emission and the fitted $g^{(2)}(0)$ is 0.28. We also note that there is a small dip at the zero delay peak as indicated by a magnification of the $g^{(2)}(\tau)$ curve shown in the inset of Fig. 2(c). This is due to the fact that the QD decay time is the same order of the excitation laser pulse width, leading to the QD capturing more than one exciton in one excitation pulse. Thus we can expect a lower $g^{(2)}(0)$ with narrower pulse width laser excitation.

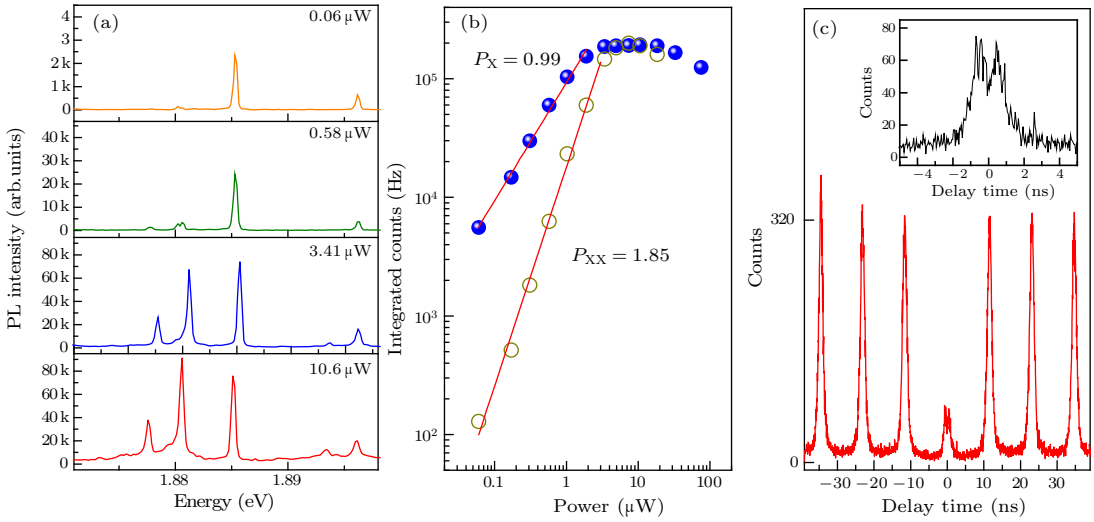


Fig. 2. (Color online) A typical single-photon emission from a QD in NW. (a) PL spectra from an emission line at different excitation powers at 6 K. (b) Integrated counts per second for the spectra shown in (a) as a function of the excitation power. (c) The second-order correlation function $g^{(2)}(\tau)$ measurement at 10.6 μW, and $g^{(2)}(0)$ is 0.28. Inset: magnified graph of $g^{(2)}(\tau)$ near zero delay time.

To estimate single photon extraction efficiency, firstly we use a cw laser at 632.8 nm to calibrate the collection efficiency of the spectral measurement setup. During the calibration, an attenuated laser beam was reflected by a silver mirror, then went through the same path as the collected PL, and finally went into the spectrometer and the detector. By measuring the intensity of laser beam, I, and the power of attenuated laser P, we can obtain the total detection efficiency of the setup $\eta = Ih\nu/P$, where h is the Plank constant, and ν is the frequency of photon. For our setup, the calibrated detection efficiency is 0.015. At the saturate excitation power, I_X is $\sim 1.93\times10^5$ counts per second. Thus the single-photon flux into the first lens is $1.93 \times 10^5(1 - g^{(2)}(0))^{1/2}/0.015 = 1.09 \times 10^7$.[5] This leads to the estimated extraction

efficiency of 14% calculated by a ratio between the single photon flux and laser pulse repetition frequency (80 MHz). It should be noted that the calculated extraction efficiency is for the as-grown NWs shown in Fig. 1(a), whereas the strongest emission direction should be along the long axis of the NWs.[1,8] For comparison, Fig. 3 shows the PL integrated intensities as a function of the excitation power for two NW configurations of PL collection as shown in Fig. 1. It is found that the QD emission intensity for NWs in the substrate surface is about one order of magnitude smaller than that for the tilt NWs at the same excitation power, showing that the collected PL intensity can be highly enhanced by adjusting the collection direction. Hence we can expect a higher extraction efficiency when the collection direction is parallel to the long axis of NWs.

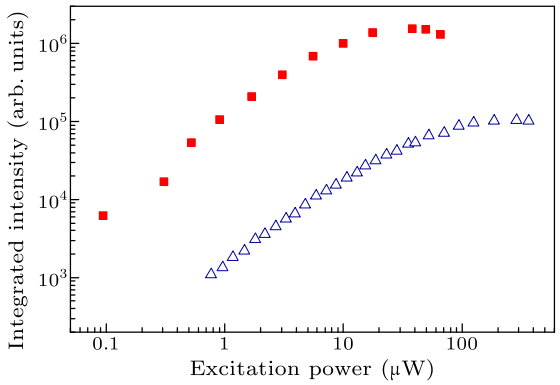


Fig. 3. (Color online) PL integrated intensity of QD emission lines for QDs in a tilt NW (red solid squares) and QDs in NWs placed on the substrate (blue open triangles) as a function of the excitation power.

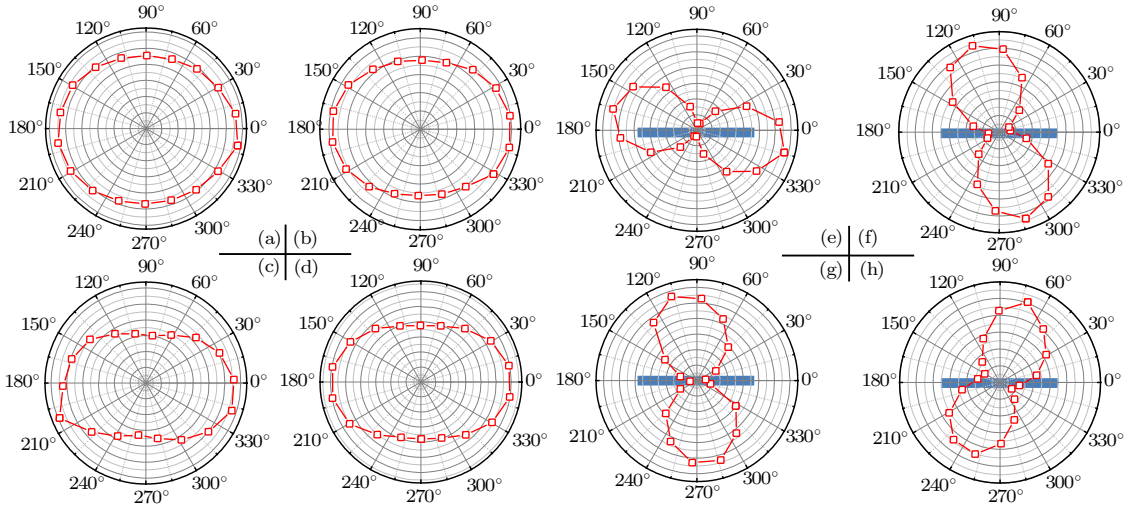


Fig. 4. (Color online) (a)–(d), (e)–(h) Polarization measurements of QD emission lines for different QDs in the tilt NWs as shown in Fig. 1(a) and for the QDs in the NWs placed on the substrate as shown in Fig. 1(b), respectively. The blue bars in (e) and (h) show the long-axis direction of NWs.

Finally, we investigate the polarization properties of the QD emission lines. Figures 4(a)–4(h) represent the polarization measurements of different QD emissions as a function of the degree of the linear polarizer for the two types of PL collection configurations shown in Figs. 1(a) and 1(b), respectively. They demonstrate that the former shows weak polarization emission, while the latter one has strongly linearly polarized emission and there is not defined polarization orientation corresponding to the long axis of NWs (here blue bars in Figs. 4(e)–4(h) represent the long axis of NWs), the same case was reported in Ref. [15]. Note that the polarization, determined by the orientation of the dipole matrix elements, is an important property of QD exciton emission for quantum light sources.[16] In self-assembled QDs, the exciton is usually a heavy hole (HH) exciton due to the quantum confinement (and strain) in conventional QDs. Thus the exciton of such QDs has two bright, orthogonally polarized states in the QD base x–y plane, separated in energy by a fine structure splitting.[17] In the case of QDs in NWs, however, there is no defined QD z-direction corresponding to the PL collection direction for the tilt NWs, which may result in the measured weak polarization PL. For the NWs placed on the substrate surface, however, each QD has different defined xy-axes, leading to the observed linear polarization without a defined angle with the NW long axis.

In conclusion, we have presented a kind of bright single-photon source based on the GaAs QDs in NWs. The integrated intensity of 1.93×10^5 counts per second and a single-photon extraction efficiency of 14% are demonstrated. This kind of single-photon source was grown by MBE through the self-assembled method, which has no requirement for post-processing from the top-down approach. Further studies are

needed to enhance the extraction efficiency and to realize polarization control.

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