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Shape-engineered epitaxial InGaAs quantum rods for laser applications

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We apply artificial shape engineering of epitaxial semiconductor nanostructures to demonstrate InGaAs quantum rods (QRs), nanocandles, and quantum dots-in-rods on a GaAs substrate. The evolution of the QRs from a zero-dimensional to one-dimensional confinement is evidenced by systematically measuring the photoluminescence and photoluminescence decay as a function of the rod length. Lasers based on a three-stack QR active region are demonstrated at room temperature, validating the applicability of the QRs in the real devices. © 2008 American Institute of Physics. [DOI: 10.1063/1.2903098]

Due to unique quantum confinement properties, both zero-dimensional (0D) quantum dots (QDs) and one-dimensional (1D) quantum wires have been extensively studied and utilized in electronic and optoelectronic devices. An interesting intermediate situation between 0D and 1D confinement is represented by elongated QDs, i.e., quantum rods (QRs). For the fundamental investigations, QRs can be used to investigate how the electronic structure evolves from a 0D to a 1D quantum system.^{1,2} From the application perspective, QRs may have potential advantages over QDs such as enabling the control of the polarization.^{1,3} However, until recently, the difficulties in fabricating QRs have limited both fundamental investigations and applications to real devices. Chemical synthesis by vapor-liquid-solid and solution-liquid-solid mechanisms has been used,^{1,4} but colloidal QRs prepared by this approach are sensitive to surface traps. Recently, the growth of columnar QDs embedded in a GaAs matrix by molecular beam epitaxy (MBE) has been explored.^{5–7} In this approach, the columnar QDs are formed by first growing a layer of seed QDs in the standard Stranski–Krastanow (SK) mode, and then depositing a short period GaAs/InAs superlattice (SL) on the GaAs substrate. The in-plane strain distribution created by the first QD layer favors the In incorporation on top of QDs,⁸ resulting in an In-rich column-shaped nanostructure. The height of these nanostructures can be independently controlled by changing the number of SL periods; indeed, the growth of columnar QDs with aspect ratio >1 was recently demonstrated,⁶ suggesting the potential to fabricate QRs. The QRs produced by this approach can be free from surface traps and are structurally compatible with the conventional GaAs/AlGaAs waveguide structure. In this paper, we (a) demonstrate the possibility of engineering the shape and composition of columnar QDs to obtain QRs with tunable aspect ratio and compositionally modulated QRs, (b) investigate the transition from 0D- to 1D-like optical properties as a function of rod length, and (c) show the applicability of such QRs to laser devices by demonstrating room-temperature lasing operation based on a QR active region.

The samples were grown by using MBE on the (001)-oriented GaAs substrate. The QRs were formed by depositing a 1.8 ML InAs QD seed layer and a short period GaAs (3 ML)/InAs (0.62 ML) SL. Growth detail and optimization of the QRs were reported elsewhere.⁷

Transmission electron microscopy (TEM) measurements on the QR samples containing SL with different number of periods (N) of 3, 16, and 35 were performed. Figure 1(a) shows the $g=(002)$ dark field cross-sectional TEM images of these samples, which demonstrate the evolution of the QR formation. Indium composition profiles across the center of the QRs are also presented. The structure of these samples consists of two parts, i.e., the vertical rods and a two-dimensional (2D) layer around them. The 2D layer consists

FIG. 1. (Color online) TEM of the QRs containing GaAs (3 ML)/InAs (0.62 ML) SL with different number of periods $N=3$, 16, and 35. (a) $g=(002)$ dark field cross-sectional TEM images of all the samples. Indium composition profiles across the center of the QRs were also presented. The curves are offset for clarity. (b) $g=(002)$ dark field cross-sectional TEM micrograph of the sample with $N=35$.

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FIG. 2. (Color online) $g=(002)$ dark field cross-sectional TEM images of (a) a candlelike nanostructure (nanocandle) and (b) a QD-in-QR structure. Indium composition profiles across the center of the nanostructures are also shown. The curves are offset for clarity.

of an InGaAs quantum well with a uniform In composition of 16%. The In composition across the center of the QRs is not perfectly uniform, a vertical composition fluctuation of about 10% with an average value about 35% is observed. The in-plane diameter of the rods is about 10 nm, which is set by the diameter of the seed QD layer and can be controlled by varying the growth parameters.⁹ The rod height can be tuned by varying the number of SL periods. The height of the rods with $N=3$, 16, and 35 is 5, 28, and 41 nm, corresponding to aspect ratios of 0.5, 2.8, and 4.1, respectively. The aspect ratio of 4.1 is extremely large and it is impossible to achieve for the conventional SK growth of QDs. No dislocation and plastic relaxation were observed even for the QR sample with $N=35$, as shown in Fig. 1(b), indicating an excellent material quality.

A unique feature of these epitaxial QRs is that the vertical profile of the In composition, and thus, of the band gap and of the strain, can be easily controlled by changing the growth conditions during the rod growth. An illustrative example of this engineering freedom is the candlelike nanostructure (nanocandle) shown in Fig. 2(a). In this case, the GaAs (3 ML)/InAs (0.62 ML) SL was replaced by a chirped GaAs (3 ML)/InAs (x ML) SL, where the InAs thickness x was gradually decreased from 0.72 to 0.56 ML at fixed steps. The In composition across the center of the rod smoothly decreases from the bottom to the top, and at the same time, the rod diameter decreases, leading to a tapered, candle like profile. As a further step, we designed and fabricated a QD-in-a-rod structure by increasing the InAs layer thickness within the SL from 0.62 to 0.72 ML at the center of the rod. The TEM image and In composition profile shown in Fig. 2(b) evidenced the formation of an In-rich region within the rod. These two examples show a clear route toward the complete control of In composition profile along the growth direction, allowing, for example, the controlled formation of coupled QDs and tunnel-injection barriers, the control of potential gradients within the rod, etc.

To confirm the 1D characteristics of the QRs, their optical properties and the carrier relaxation dynamics were investigated by microphotoluminescence (micro-PL) and time-

FIG. 3. (Color online) Normalized PL spectra (10 K) of the QR samples with $N=3$ and 35. The inset shows the PL peak energy shift as the function of the QR length.

resolved PL measurements at 10 K. Figure 3 shows the normalized low-temperature (10 K) PL spectra of the QR samples with $N=3$ and 35. A well-developed single PL peak was observed in each spectrum, indicating that the size of the QRs is very uniform. With increasing rod length, the PL peak energy redshifts (inset of Fig. 3). This indicates the delocalization of the wave functions along the QR length.^{2,4} Figure 4 presents the typical PL decay curves of the samples with $N=3$, 16, and 35. For the sample with $N=3$, the curve can be essentially characterized by a monoexponential decay with a time constant (TC) of 0.85 ns, which is very similar to that of the conventional SK growth of QDs.¹⁰ In contrast, for $N > 10$, (see e.g., $N=16$ and 35 in Fig. 4), the PL decays are characterized by a biexponential dynamics, which consists of a fast TC of 1–2 ns and a slow TC of about 10 ns. This type of biexponential decay phenomenon is often observed in QD and quantum wire structures,^{2,11,12} and attributed to the existence of nonradiative (dark) states with much longer decay time. In the present QR structure, dark states may originate from hole states strongly localized in a subsection of the rod, thus, having a small overlap with the ground electronic wave function. The fast TC increases with increasing rod length

FIG. 4. (Color online) PL decay curves of the QR samples with $N=3$, 16, and 35. The inset shows the dependence of the fast TC on the QR length.

FIG. 5. (Color online) Output power against drive current (L - I) characteristic from a $3\text{ mm} \times 10\text{ }\mu\text{m}$ QR laser stripe under pulsed operation at room temperature. Inset: lasing spectra (log scale) just above the threshold current.

(inset of Fig. 4). According to the simple picture of increasing oscillator strength with increasing exciton volume,¹³ the opposite trend would be expected. However, due to the significant difference in the electron and hole effective masses of the $\text{In}_{0.35}\text{Ga}_{0.65}\text{As}$ QR, the electron wave function is still delocalized but the hole wave function is more easily confined in a section of the rod.^{2,4,14} As the rod length increases, the overlap between these two wave functions decreases, resulting in a lower transition probability, and thus, a longer lifetime.

To validate the applicability of the QRs in the real devices, a laser device structure based on a three-stack QR active region was grown. The QR length is 36 nm, corresponding to an aspect ratio of 3.6. Figure 5 shows the output power versus drive current (L - I) characteristic for a $3\text{ mm} \times 10\text{ }\mu\text{m}$ laser at 20°C . The lasers were measured as-cleaved and unmounted, under pulsed operation. The laser presents a threshold current density of 650 A/cm^2 . A clear lasing peak is observed at 1130 nm, as shown in the inset.

In conclusion, we demonstrated the feasibility of artificial shape engineering QDs. Controlled nanostructures including InGaAs QRs, nanocandles, and QDs-in-QRs were grown by MBE. The transition characteristic of the QRs from 0D to 1D confinement type was evidenced by evaluating the evolution of the exciton energy and oscillator strength of the QRs as a function of the rod length. At room temperature, laser diodes based on a QR active region were demonstrated, validating the applicability of the QRs in the real devices.

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