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Optical properties of stacked InAs self-organized quantum dots on InP (3 1 1)B

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Abstract

We have investigated the structural and optical properties of multi-stacked self-organized InAs quantum dot (QD) structures on InP (3 1 1)B substrates grown by molecular beam epitaxy. The reciprocal space mapping measured for asymmetric (4 0 0) reflections in high resolution X-ray diffraction revealed that satellite peaks originating from a periodic structure were not only observed along the growth direction, but also along $[\bar{2}\,3\,3]$ direction within each QD plane. The mean in-plane spacing of neighboring QDs was 97 nm, which was in good agreement with direct measurement of 100 nm by using atomic force microscope. A superior three-dimensional QD superlattice with a total density of $10^{12}\,\mathrm{cm}^{-2}$ was achieved by our growth technique. We also observed a strong photoluminescence emission at 1.55 μm with a narrow linewidth of 56.07 meV at room temperature.

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1. Introduction

Recently, studies on the semiconductor quantum dots (QDs) have attracted intense research because of their potential for device applications in the quantum information technology [1,2], and optoelectronics such as QD lasers [3], semiconductor optical amplifiers [4] and next generation solar cells [5]. For this, fabrication of QDs with a sufficient density and homogeneity in the active region is required. Until present, self-assembled QDs fabricated by spontaneous islanding by Stranski–Krastanov (S–K) growth mode in lattice-mismatched heteroepitaxy were intensely investigated, and S–K QDs show a high crystalline quality and good optical characteristics with few defects and dislocations. In general, multiple stacking of QDs by S–K growth is used to increase the total QD density. However, as a consequence of gradual buildup of internal lattice strain with stacking, QD size increases with increasing number of stacked layers [6]. Recently, the concept of strain compensation was first demonstrated in PbSe/PbEuTe QDs on BaF_2 substrates [7]. By following this technique, we previously reported on the stacking of laterally ordered InAs self-organized QDs on InP (3 1 1)B substrates by strain-compensation growth technique, in which Al atoms in InGaAlAs spacer layers played an important role in preventing the segregation of In and interdiffusion of In atoms between the spacer layer and QDs [8]. We also demonstrated 30 layer-stacked InAs quantum dot solar cells for the first time [9]. In this work, we report on the structural and optical properties of stacked InAs QDs on InP (3 1 1)B substrates with 100 multiple layers of QDs.

2. Experiments

For the fabrication of 100 multiple layers of self-organized InAs QDs on InP (3 1 1)B substrates, we

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employed solid-source molecular beam epitaxy (MBE). After the oxide removal and surface cleaning, a 150-nm-thick $In_{0.52}Al_{0.48}As$ buffer layer lattice matched to InP substrate was grown at 500 °C. Then, four monolayers (MLs) of InAs QD layer and a 20-nm-thick $In_{1-x-y}Al_xGa_yAs$ spacer layer, or strain-compensation layer were consecutively grown in pair up to 100 multiple cycles as reported previously [10]. The lattice constant of $In_{1-x-y}Al_xGa_yAs$ layer was made smaller than that of InP substrate in order to satisfy the strain-compensation condition. The growth process and surface morphology were studied in situ by reflection high-energy electron diffraction (RHEED), and ex situ by atomic force microscope (AFM) and cross-sectional scanning transmission electron microscope (STEM). A high resolution X-ray diffraction (HR-XRD) was used to determine both the strained state and periodicity of three-dimensional QD superlattice structures. Photoluminescence (PL) measurements were performed at temperatures between 20 and 300 K within a closed cycle helium cryostat. The signals were detected by using a standard lock-in technique with cw 532 nm second harmonic generation (SHG) $Nd{:}YVO_4$ laser as an excitation source and a liquid nitrogen-cooled InGaAs photodetector. The excitation intensity was ~6 W/cm^2.

3. Results and discussion

Fig. 1 show the (a) topographic, and (b) two-dimensional fast Fourier transformation (2D-FFT) images, measured for the topmost QDs surface in 100 layer-stacked sample. The average diameter, height, and area density were 72.1, 8.4 nm, and $2.88 \times 10^{10}\,cm^{-2}$, respectively. To be noted was that no coalesced islands were observed, and a significantly improved size dispersion of 9.4% in diameter was obtained. Furthermore, sharp six-fold peaks and higher-order peaks in the 2D-FFT image as shown in Fig. 1(b) indicated a well defined and laterally ordered QD array structure. The ordered QD structure is known to be related to the atomic structure of (3 1 1)B surface [11], and the effect of strain field in the underlying QD layers [12]. As a result, the total density of QDs amounted to the order of $10^{12}\,cm^{-2}$, which could not be achieved by the conventional self-assembly growth technique. Fig. 2 shows the cross-sectional STEM image of stacked InAs QDs sample. No dislocations were observed, which would otherwise be generated if the build-up of lattice strain exceeded the critical limit during stacking, and a vertical alignment of QDs along growth direction was clearly maintained without an increase in the size nonuniformity.

Next, we carried out reciprocal space mapping (RSM) measurements around asymmetric (4 0 0) reflection in HR-XRD as shown in Fig. 3. In contrast to a diffuse X-ray scattering pattern that was observed for either a QW [13] or disordered QD superlattice [14], several satellite peaks in the RSM were observed not only along the growth direction q_y, but also along q_x parallel to [$\bar{2}$ 3 3] direction

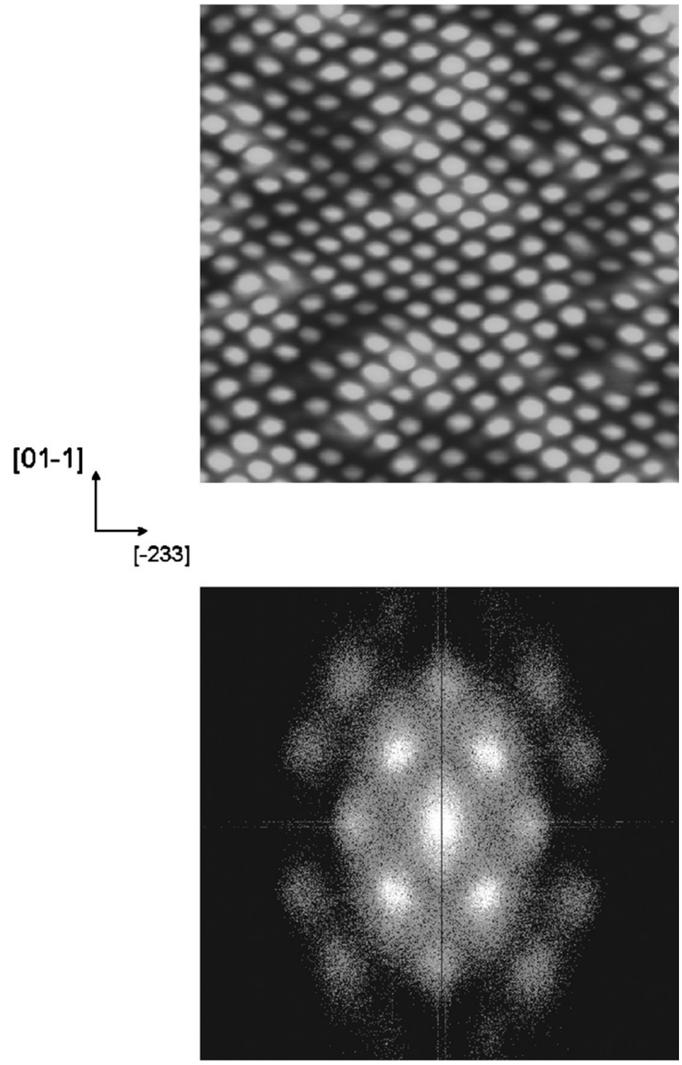

Fig. 1. (a) Topographic AFM image, and (b) 2D-FFT image of the topmost dot layer of 100 layer-stacked InAs quantum dots on InP (3 1 1)B. Scan size is 1 μm × 1 μm.

within the QD planes. The 0th-order peak F_0 in Fig. 3(a) showed a near perfect lattice-match with the peak S for InP substrate. In this case, the averaged lattice constant of quantum dot superlattice structure was nearly perfectly matched to InP, i.e. the compressive strain in each QD layer was compensated by introducing tensile strain by InGaAlAs strain-compensating spacer layer. In addition, the satellite peaks seen along q_y suggested that no lattice relaxation occurred during stacking.

The appearance of satellite peaks along q_x such as F_{+1x}, F_{+2x} and F_{-1x} in the figure indicated that the placement of QDs were highly correlated within (3 1 1)B plane, that is, QDs formed a periodic lattice structure at least along [$\bar{2}$ 3 3] direction. We calculated the mean period Λ between the neighboring QDs along [$\bar{2}$ 3 3] by using the following equation,

$$\Lambda = (n_i - n_j)\lambda/2(\sin\omega_i - \sin\omega_j), \quad (1)$$

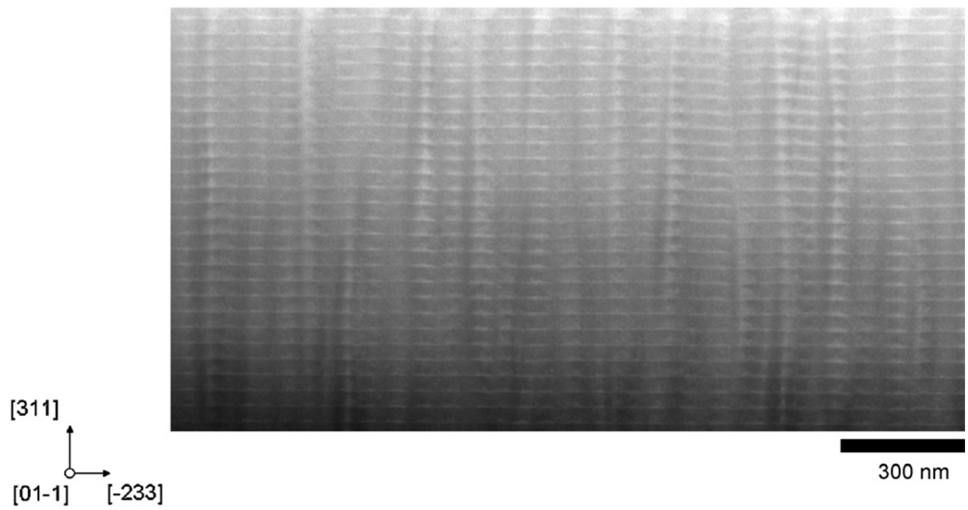

Fig. 2. Cross-sectional STEM image of stacked InAs quantum dot structure.

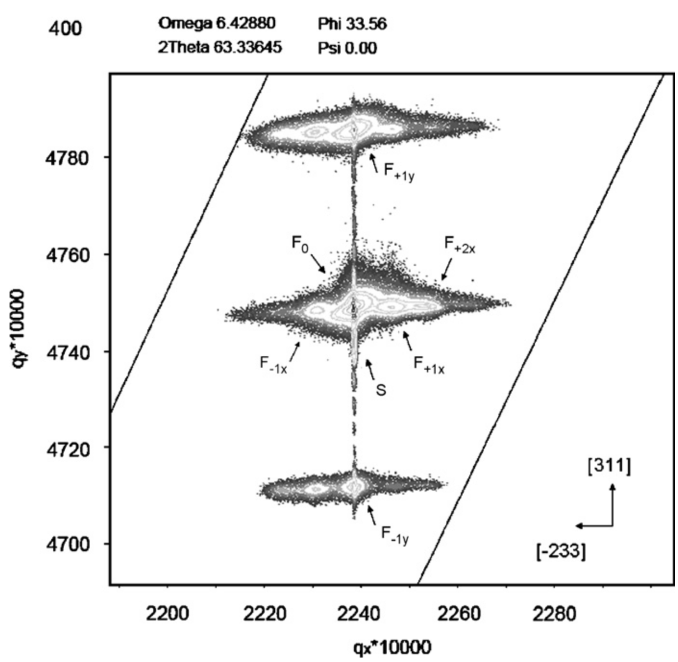

Fig. 3. RSM of diffracted X-ray intensity around (4 0 0) reflection measured for 100 layer-stacked InAs quantum dots on InP (3 1 1)B. q_x and q_y are along [$\bar{2}$ 3 3] and [3 1 1] azimuth, respectively.

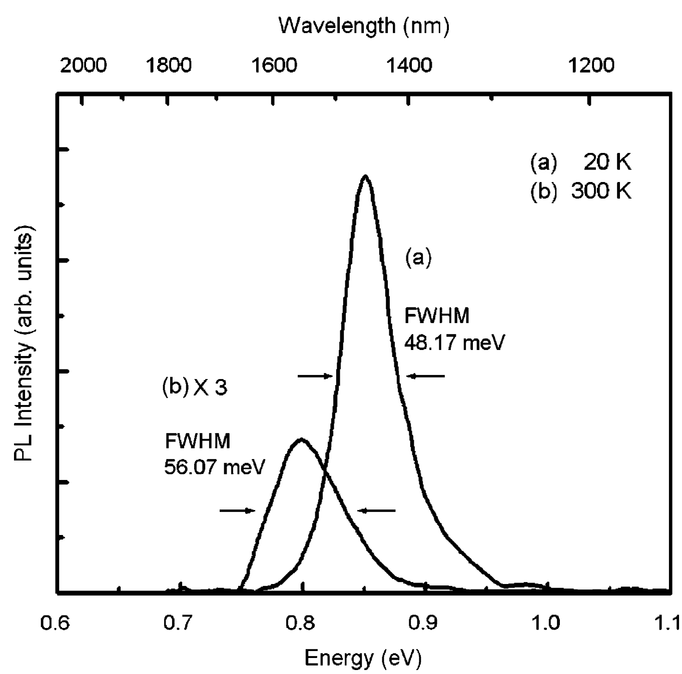

Fig. 4. PL spectra measured for 100 layer-stacked InAs QDs at temperatures between 20 and 300 K.

where n_i and n_j are the degrees and ω_i and ω_j are the Bragg angles of the ith and jth order satellite peaks, respectively. λ is the wavelength of X-ray (CuKα_1~1.5456 Å). The mean spacing between the dots was $\Lambda = 97$ nm, which was in good agreement with the direct measurement of 100 nm obtained from AFM measurements.

Fig. 4 show the PL spectra measured at (a) 20 K and (b) 300 K, respectively. The PL peak corresponding to emission from the fundamental state of QDs was clearly observed around 1.55 μm at 300 K. An asymmetric broadening of PL spectra on the high-energy side was also observed, and we believe that this tail shape was due to additional emission from the first excited state. Fig. 5 shows the temperature dependence of integral peak intensity. We observed a strong quenching of PL intensity above 160 K which was due to the onset of thermally activated nonradiative recombinations. The experimental Arrhenius plot was fitted with the following equation and the activation energy E_a of confined carriers in QDs was determined,

$$I = I_0/\{1 + C\ \exp(-E_a/k_B T)\} \quad (2)$$

where k_B is Boltzmann's constant, I_0 and C are the constants independent of temperature. We obtained a good

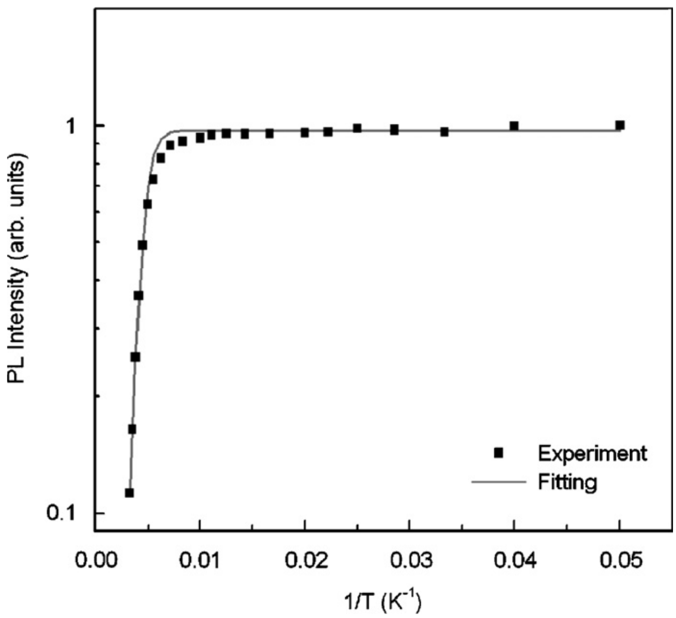

Fig. 5. PL integral intensity (closed squares) of quantum dot emission as a function of temperature. Solid line shows a fitted curve.

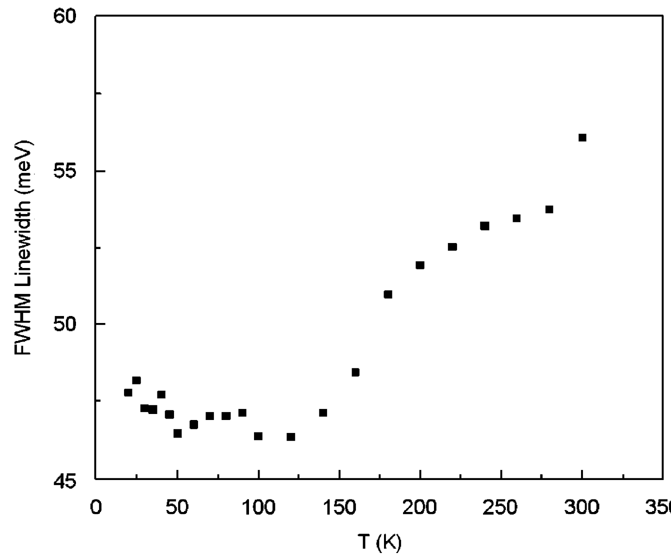

Fig. 6. PL peak linewidth of quantum dot emission as a function of temperature.

fit to the experimental data as shown in Fig. 5, and E_a was calculated to be 150 meV.

Finally, Fig. 6 shows the temperature dependence of full-width at half-maximum (FWHM) of PL peak as a function of temperature. It can be seen that a relatively narrow PL linewidth of 56.07 meV was obtained at 300 K. The penetration depth of laser beam was estimated to be about 150–200 nm and thus the number of QD layers that was probed by the laser was about 8–10 layers from the topmost surface. Although PL signal represented only the contributions from several QD layers in our measurements, the narrow linewidth was comparable to that for a single layer of InAs QDs on InP substrate reported previously [15,16]. This indicates that a superior in-plane size uniformity of QDs was obtained even after 100 layers of stacking. In addition, an anomalous decrease in PL linewidth with increasing temperature up to 100 K was observed. There have been several reports on the carrier dynamics of thermally activated carriers that migrate or tunnel from smaller sized QDs to larger dots via wetting layer. Such carrier dynamics is commonly observed in bimodal QD structures and is known to show a minimum PL linewidth at an intermediate temperature range [17], and exhibit two components following the Varshni law [18]. Here we further believe that the carriers can be transferred between QDs more easily in our stacked structure since carrier scattering would be reduced in an ideal three-dimensional superlattice structure. The observation that the integral PL intensity remained constant for intermediate temperatures as shown in Fig. 5 supported that the carrier transfer between QDs was the dominating mechanism instead of other relaxation mechanism such as non-radiative recombination.

4. Conclusion

We successfully fabricated 100 layers of multi-stacked self-organized InAs quantum dots on InP (3 1 1)B substrates, which showed a good laterally ordered structures with clear six-fold symmetry. The QDs were aligned along [$\bar{2}$ 3 3] within the plane as well as along the growth direction. Further, we observed PL emission at 1.55 μm with a narrow linewidth of 56.07 meV at room temperature that is suitable for fiber-optic communication as well as next-generation photovoltaic applications.

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