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Structural and Optical Characteristics of Laterally Self-Aligned InGaAs Quantum Dots

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The structural and the optical characteristics of laterally self-aligned InGaAs/GaAs quantum dots (QDs) have been analyzed through mutual comparisons among four samples with different parameters. An anisotropic arrangement develops with increasing number of stacks, and high-temperature capping allows isolated QDs to be spontaneously organized into a one-dimensionally-aligned chain-like shape over a few μm. Moreover, the migration time allowed by growth interruption plays an additional important role in the chain arrangement of QDs. The QDs capped at high temperature exhibit blue shifts in the emission energy, which may be attributed to a slight outdiffusion of In from the InGaAs QDs. An estimate from the energy shift reveals that the compositional variation of In is 0.05.

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I. INTRODUCTION

During the last decade, much effort has been devoted to the fabrication of nanoscale quantum dot (QD) structures on the basis of the unique features of the electron and the hole wave functions attributed to atom-like strong confinement. In particular, due to their defect-free nature, spontaneously formed QDs have attracted tremendous interest not only in the exploration of exotic properties [1] but also in applications to practical devices, such as QD-based laser diodes [2, 3], infrared photodetectors [4, 5], single-photon emitters [6], single-electron devices [7], and various other devices [8,9]. However, self-assembled QD formation technology is now confronted with some difficulties in controllability and reproducibility due to the complexity of the growth kinetics. Moreover, the site-controlled QD technique using patterned substrates or templates rests upon fundamental limits of unavoidable lithographical accessibility and considerable defects. Of current interest is, therefore, the realization of self-assembled QD systems with further improved uniformity in position and dimension, which is necessary to utilize QD-based functional devices, especially for arrayed devices.

In order to achieve regularly ordered QDs, various self-assembling techniques have been tested on a variety of processed substrates with high-index vicinal surfaces, patterned nanoscale trenches/holes or mesas, masked templates, and strain-engineered layers [10–20]. Lee *et al.* [19] and Springholz *et al.* [20] nicely demonstrated the possibility of crystal-like 2D-/3D-arrayed QD structures by using patterned stressor mesas and strain-controlled superlattices in the InAs/InGaAs and the PbSe/PbEuTe systems, respectively. In recent years, as an alternative to access regularly arrayed QDs, one-dimensional (1D) vertical and lateral self-alignment of QDs has been extensively studied by controlling only the growth parameters and the layer structures with no use of processed substrates [14–18]. Using the built-in strain anisotropy induced in stacked layers, some research groups have attempted lateral self-alignment of QDs by introducing growth interruption (GI) or *in-situ* annealing during the initial stages of the capping process [16,17]. They found that separate QDs were spontaneously aligned into the form of QD chain (QDC) during the multilayer stacking procedure and that the capping procedure played an important role in the anisotropic QD alignment. In the many studies made on the growth

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kinetics and the morphological behaviors of the self-aligned QDs and QDCs in order to identify the formation mechanisms, the reported results were rather fragmentary because of the limited range of the growth parameters. In addition, only a few have examined spectroscopic characteristics correlated with morphological behaviors.

In this paper, we report comparative analyses of the self-aligning behaviors observed in a series of InGaAs/GaAs QDs fabricated by using a multilayer stacking technique with a modified two-step capping process. The number of stacks, the growth temperature of the cap layer (CL), and the GI time were chosen as three key parameters, and atomic force microscope (AFM) images and the photoluminescence (PL) spectra were used for analyses of the structural and the optical evolutions, respectively. We present a 1D chain structure that is due to the alignment of a set of QDs over a few μm and that shows a strong PL emission.

II. EXPERIMENTAL PROCEDURES

Four self-assembled $In_{1-x}Ga_xAs(x = 0.5)$/GaAs QD heterostructures were grown on semi-insulating GaAs just-(100) substrates by using a molecular beam epitaxy (MBE) technique, and the spontaneous formation of InGaAs QDs was performed in the Stranski-Krastanow (S-K) growth mode. The growth was initiated from a GaAs buffer layer with a thickness of 0.2 μm at a temperature of 570 °C, and a 7.5-monolayer (ML) InGaAs layer was deposited for the QD formation on the buffer layer at a lower temperature of 510 °C. A simplified two-step procedure was introduced for GaAs capping. First, a 3-ML GaAs layer was deposited just after the QD formation and a GI for 30 s or 120 s was given at the QD growth temperature. Then, an additional capping of 54 MLs was completed the entire CL at the same temperature of 510 °C or at an elevated temperature of 540 °C. The layer stacking was consecutively repeated for 10 or 15 periods, and, finally, the surface QD layer with no CL was formed on top of the whole structure for the AFM measurement. The growth rates and the V/III beam-equivalent pressures (BEPs) for InGaAs/GaAs were 0.8/0.4 ML/s and 10/25, respectively, and the As beam-flux was kept constant at $(1.75 \pm 0.25) \times 10^{-5}$ Torr.

Table 1 describes the specifications for the four QD samples prepared for this study. The initial parameters were given for sample A as a reference: the stack period is 10, the CL growth temperature 510 °C, and the GI time 30 s. For mutual comparisons, variables were added one by one to form the other three samples (B, C, D), as denoted in Table 1. (Each pair of bold-faced figures indicates a variable imposed on a couple of neighboring samples for mutual comparisons.)

The 2D-3D transition of the reflection high-energy electron diffraction (RHEED) patterns showed that the spontaneous formation of QDs began to occur on a wetting layer (WL) of 6.0 MLs in the present InGaAs/GaAs system. The structural and the optical characteristics were analyzed by using AFM images and the PL spectra, respectively. PL measurements were performed by using a visible-to-near-infrared monochromator system with an Ar-ion laser (514.5 nm) and a closed-cycle He refrigerator (10 K). The nominal laser power used in this study was typically 50 mW, and the luminescence signals were detected and recovered by using a thermoelectric-cooled InGaAs photodiode (0.8 – 1.6 μm) and a lock-in amplifier. The epitaxial growth procedures and the basic properties for similar QD heterostructures have been reported elsewhere [21–23].

Table 1. Specifications for the four QD samples used in this study. Each pair of bold-faced figures indicates a variable imposed on a couple of neighboring samples for mutual comparisons.

Sample Code	A	B	C	D
Period of Stacks	**10**	**15**	15	15
Growth Temperature of Cap Layer (°C)	510	**510**	**540**	540
Growth Interruption Time (s)	30	30	**30**	**120**

III. RESULTS AND DISCUSSION

Fig. 1 presents the surface AFM profiles of QD ensembles imaged on three different scales (5 × 5, 3 × 3,

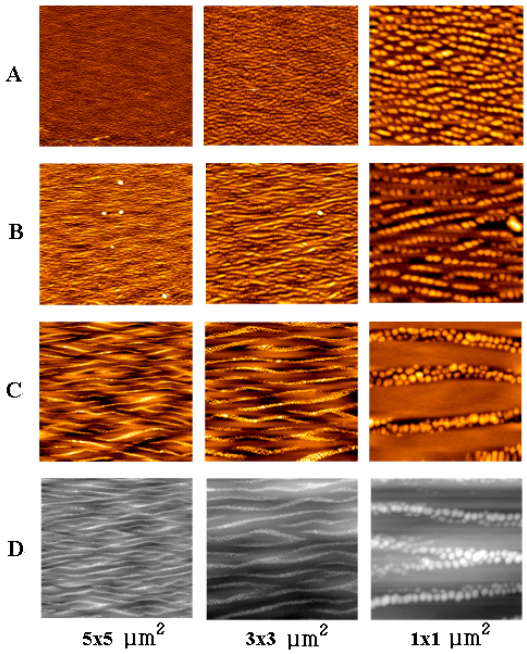

Fig. 1. Surface AFM images of QD ensembles for four samples (A, B, C, D from the top). Each structure is visualized by three kinds of images with different scales (5 × 5, 3 × 3, 1 × 1 μm^2 from the left).

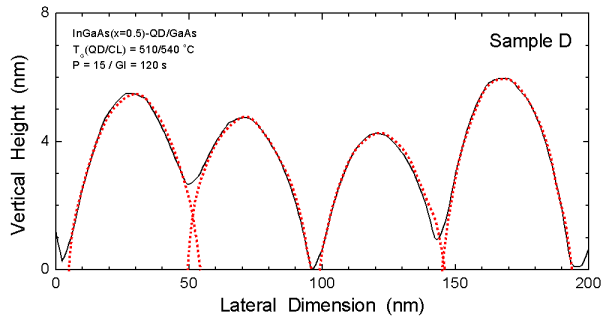

Fig. 2. Representative cross-sectional view drawn along the alignment axis for QDs of sample D. The profile shows that the aligned QDs are so tightly coupled as to exhibit chain-like behaviors.

1×1 μm^2 from the left) for the four samples (A, B, C, D from the top) listed in Table 1. As the period of stacks, the growth temperature of the CL, and the GI time are varied in order, the anisotropic arrangement becomes more and more distinguished. The AFM images of sample D clearly show 1D chains made of almost single rows of laterally aligned QDs over a few μm. This indicates that an anisotropic strain develops with increased stacks (A→B), and the capping temperature is a crucial parameter in the 1D self-alignment (B→C). In addition, the migration time allowed by GI plays an important role in the chain-like arrangement of QDs (C→D). Fig. 2 is a representative cross-sectional view of coupled QDs drawn along the alignment direction in sample D. The profile shows that the single QDs are approximately (50 $\pm$ 5) nm in diameter and (5 $\pm$ 1) nm in height and that the aligned QDs are so tightly coupled as to exhibit chain-like behaviors.

In order to support the morphological evolution discussed above, we performed spectroscopic characterizations on the same samples used for AFM imaging. The PL emission spectra taken at low temperature (17 K) are presented in Fig. 3, and a pair of curves for the peak energy (circles) and the corresponding full width at half maximum (FWHM) (squares) are plotted in terms of the sample code in Fig. 4. All the samples show strong PL emissions attributed to InGaAs QDs in the energy range of 1.24 – 1.31 eV. The spectra of samples A and B exhibit almost no variation in energy, but tend to increase in FWHM. This means that the two QD samples with different numbers of stacks basically have the same luminescent property regardless of the order of the QD arrangement, but are a little different in the FWHMs due to the increased number of QD layers. A considerable blue shift (52 meV) in the PL energy is observed between samples B and C that have shown a distinguishable change in the lateral alignment, as in AFM images. Considering that the 1D alignment effect normally brings about a shift to lower energy, we believe that the blue shift is related with thermal diffusion rather than QD alignment, as will be discussed below. Though the origin of the additional minor shift (16 meV) that appears between the PL spectra of samples C and D is not clear yet, taking into account the negligible change in FWHM (3 meV), it may come from a slight reduction in the QD size due to the prolonged GI time rather than to additional outdiffusion of In.

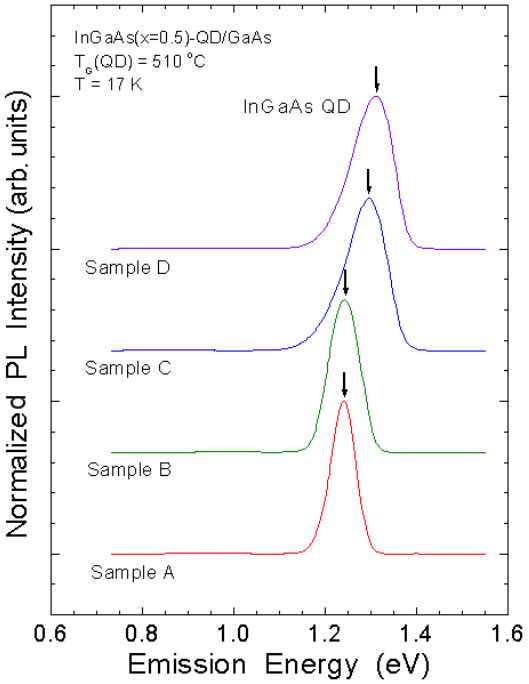

Fig. 3. PL emission spectra taken at a low temperature (17 K). All the samples show strong luminescence attributed to InGaAs QDs.

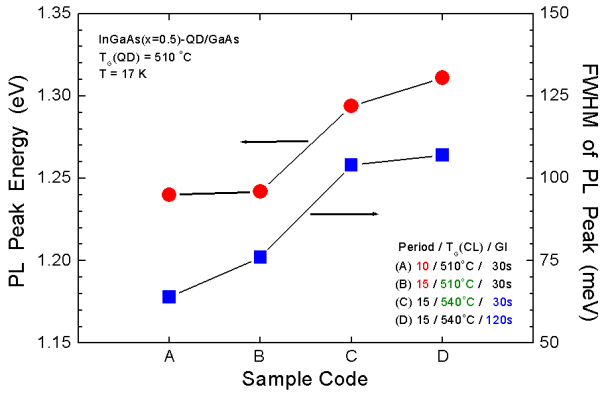

Fig. 4. Pair of curves for the peak energy (circles) and the corresponding full width at half maximum (FWHM) (squares) plotted in terms of sample code. An abrupt blue shift in the PL peak energy is observed between samples B and C.

It is generally acceptable that interdiffusion of species can take place during a capping process performed at a temperature higher than that of QD formation. In sample C capped at 540 °C, which is higher than the QD formation temperature of 510 °C, contrasted to sample

B capped at 510 °C, we can predict a slight outdiffusion of In from InGaAs QDs that results in a blue shift of PL energy. The thermal diffusion of In arising in sample C can be quantitatively analyzed by using the energy shift. In the case of a minor variation of In, an approximation can be done by using the compositional dependence of the bandgap energy (E_g) for a ternary $In_{1-x}Ga_xAs$ bulk system, $E_g(x) = 0.36 + 0.505x + 0.555x^2$ in eV [24]. If an infinitesimal change in the QD sublevel energy (ΔE) is assumed to follow the differential form of $\Delta E_g(x)$, an expression of $\Delta E = 1.065\ \Delta x$ can be derived for $x = 0.5$. With $\Delta E = 0.052$ eV, the energy shift between the PL peaks of sample B (1.242 eV) and sample C (1.294 eV), we can determine the compositional variation of In as $\Delta x = 0.05$. The considerable broadening of the FWHM of sample C shown in Fig. 4 may be indirect evidence to support this In outdiffusion. Based on this analysis, we suggest that a minor compositional variation in the InGaAs QDs may be inevitable for achieving 1D aligned QDCs because of the introduction of the high-temperature capping procedure.

IV. SUMMARY AND CONCLUSIONS

Laterally self-aligned InGaAs/GaAs QDs were fabricated by using a multilayer stacking technique with a simplified two-step capping procedure. The structural and the optical behaviors were analyzed by mutual comparisons on four kinds of samples that were different in the period of stacks, the growth temperature of the CL, and the GI time. We found that an anisotropic strain developed with increasing number of stacks and that the capping temperature played a crucial role in the formation of self-aligned QDCs. In addition, the GI time was another important variable in the 1D arrangement of QDCs. High-temperature capping gave rise to In-outdiffusion from the InGaAs QDs, which resulted in a blue shift of the PL emission, and an estimate from the energy shift gave a compositional change of $\Delta x = 0.05$. In conclusion, we suggest that a minor compositional variation in InGaAs QDs may be inevitable for the 1D self-alignment of QDCs in the case of introducing a high-temperature capping procedure.

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