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Ion beam studies of InAs/GaAs quantum dots after annealing

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Abstract

The microstructure changes of self-assembled InAs/GaAs quantum dots during RTA treatment was investigated using ion channeling and photoluminescence (PL). A small blueshift of the PL emission is observed for annealing temperatures of 650–800 °C and an obvious blueshift at 850 °C. The yield of channeled spectra decreased as annealing temperature was increased, but the yield increased while temperature above 800 °C in RTA. These results imply the strain of QD varied during RTA treatments. In addition, the As/Ga atomic ratio near the surface was determined from the surface peaks of the channeled spectrum.

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Keywords: Quantum dots; GaAs; InAs; Ion-channeling

1. Introduction

Self-assembled semiconductor quantum dots (QDs) have been used extensively in optoelectronic devices, such as semiconductor lasers and detectors [1–3]. The physical properties of the dots are strongly dependent on the strain caused by the lattice mismatch between materials (here InAs and GaAs). Thermal annealing is a way to tune the structure parameters of QDs, such as their composition, strain and size distribution [4]. The structural changes induced by thermal annealing cause changes in the QD's interband transition as well as intersublevel space energy. Generally, such a procedure leads to blueshift and narrowing in the QD's photoluminescence (PL) spectrum. However, analysis of QD's structure is difficult due to its ultra thin profile, being capped with a GaAs layer as well as its three dimension structure.

Ion channeling is a powerful technique for probing structure of thin films. It has long been used in analyzing atomic ordering in crystal structures and has been successfully applied for the study of strain in buried QDs. Previous reports have demonstrated that the strain and interdiffusion of QDs can be studied from angular scan curves in ion channeling measurements [5,6]. In this work, the strain relaxation of the QDs after thermal annealing was studied using RBS/channeling aligned energy spectra in both $\langle 100 \rangle$ and $\langle 110 \rangle$ directions.

2. Experiment

The InAs quantum dots studied in this work were grown by molecular beam epitaxy (MBE) on semi-insulating GaAs(100) substrate. Growth rates were 0.8 μm/h for GaAs and 0.056 μm/h for InAs. Arsenic pressure was 2–3×10^{-6} Torr. A buffer layer of 500 nm GaAs was grown first. Then, 0.9 nm InAs QD layers (about 2.6 monolayers) was grown at a temperature of 520 °C. All samples were capped with a 50 nm thick GaAs layer. From the atomic force microscope (AFM) image, the density of QDs was determined to be approximately $1 \times 10^{10}/\text{cm}^2$. The structure scheme is shown in Fig. 1. Post-growth annealing

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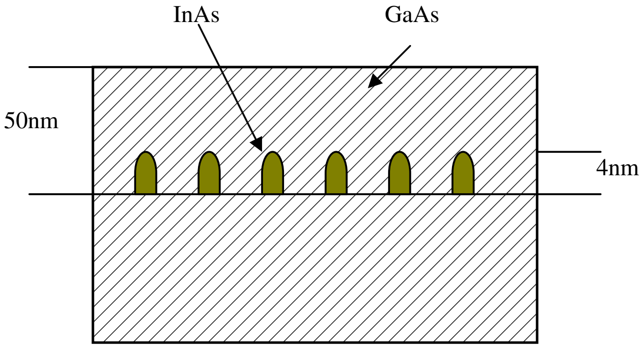

Fig. 1. Schematic diagram of the QD samples.

was performed in forming gas at temperatures of 650–850 °C for 30 s. Low temperature (25 K) photoluminescence measurement was performed with a 488 nm argon ion laser and a 75 cm spectrometer. The signal was collected using an InGaAs detector with lock-in technique.

RBS/w channeling measurement was performed with 4 MeV $^{12}C^{++}$ beam produced by the 9SDH-2 Tandem accelerator at National Tsing Hua University. The beam divergence was less than 0.02°, defined by two sets of slits 2.3 m apart. The incident particle flux was about 200 nA on the target. The sample was mounted in a three-axis goniometer with an angular resolution of <0.01°. The scattering chamber was kept at a vacuum of 2×10^{-6} torr. Backscattered particles were collected by a PIPS detector at 160° laboratory angle. The energy resolution of the system was 90 keV, determined by fitting the GaAs edge of the energy spectrum, using RBS simulation code RUMP [7]. This value is significantly lower than the energy difference between the signals from In atoms and the GaAs edge in the backscattering spectrum (526 keV). The In atom depth profile can not deduced from In peaks, as shown on the inset in Fig. 2, due to its ultra thin profile and low concentration.

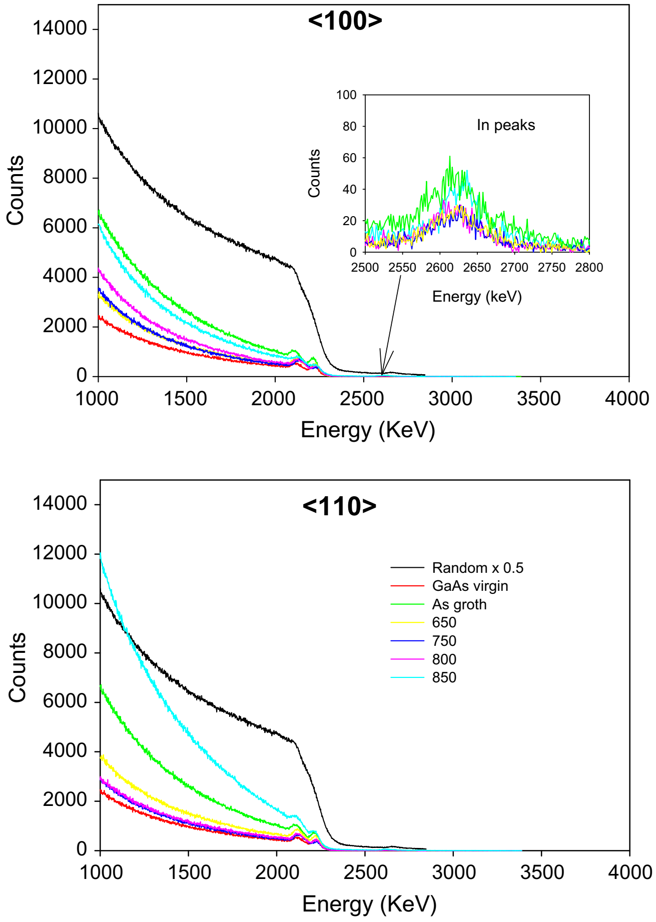

Fig. 2. RBS/w channeling spectra along the (a) 100 and (b) ⟨110⟩ axes of the samples.

3. Result and discussion

The self-assembled InAs QD growth starts with the formation of a two-dimensional InAs layer which completely covers the surface of the GaAs substrate. As the thickness of InAs film exceeds the threshold value, the island-like dots are formed on top of the 2D layer, or the wetting layer. The lateral lattice constant of the fully strained wetting layer is constrained to be the same as the underlying GaAs lattice constant and the vertical lattice constant is thus greater than the lattice constant of GaAs matrix as a result of tetragonal distortion. As a consequence of island formation, the lattice constants of the InAs dots for both the vertical and lateral directions are expected to be similar to the values for the InAs bulk crystal. Channeled ions may be dechanneled if the continuity of the channel is disrupted at the interface between dots and layers. This provides us the opportunity to look at the thin QD layer strain change after thermal treatment.

Fig. 2(a) and (b) show the RBS/w channeling spectra of the samples along the ⟨100⟩ and ⟨100⟩ directions, respectively. The aligned spectrum of GaAs wafer was also plotted as a reference. The yields of samples with InAs QDs were higher than the GaAs reference, implied some crystal distortion in the samples. Interestingly, the yields of aligned spectra in both directions did not vary monotonically with the thermal treatment temperature. Below 750 °C, the yields decreased as temperature increased, approaching to the GaAs reference. This behavior is similar to the ion implanted samples during the annealing process, namely, solid phase epitaxial re-growth. Some defects in samples are recovered by thermal annealing. Above 750 °C, the yields increased surprisingly with an abrupt increase observed at 850 °C. The yield increase indicated that the channeled ions underwent dechanneling when they passed through the InAs layer. This implied that the atoms in InAs layer displaced from their lattice sites and interdiffused with surrounding GaAs. Furthermore, the channeled yield of 850 °C annealed sample is higher than as-grown sample along ⟨110⟩ direction, but not along ⟨100⟩, indicating that the vertical displacement, along growth direction, is larger than horizontal displacement. It evidences tentatively the former cubic-like InAs dots mixed with surrounding GaAs

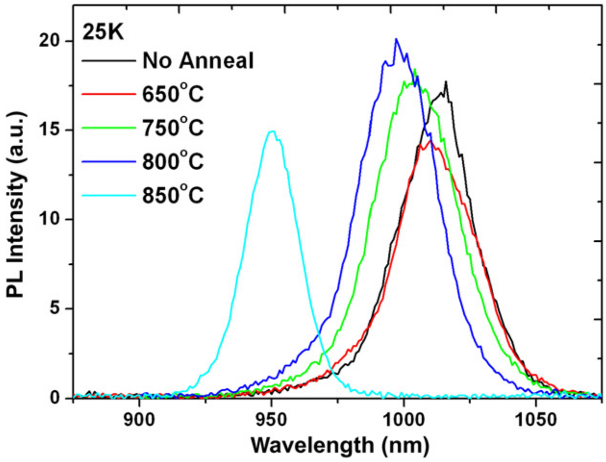

Fig. 3. Photoluminescence spectra at 25 K of QDs before and after annealing.

and gradually to become $In_xGa_{1-x}As$ with tetragonal distortion after high temperature annealing.

Fig. 3 shows 25 K PL spectra of samples before and after annealing. Below 800 °C, there is only a little blueshift. But a significant blueshift (70 nm) was observed at 850 °C with a narrower peak width compared to low temperature annealing and as-grown samples. It implied that the structure and (or) the composition of QDs was changed after annealing. From the channeling result, we know that annealing causes the capping and buffer GaAs layer to restore and strain of the QDs to relax. Strain relaxation should result in a redshift in the PL measurement due to band gap shrinkage. However, the results of PL measurements show a clear blueshift. The band gap energy is influenced by both strain and composition. However, we can deduce only the information on microstructure change of the QDs from dechanneling effect, but cannot gather any composition information because of the depth resolution limit in this work. Therefore, the explanation for this contradistinction between PL and channeling results might be attributed to the composition intermixing between the QDs and the surrounding GaAs during annealing. This is consistent with the work of Fu et al. [8], who had studied effects on interdiffusion in InGaAs/GaAs quantum dot using different capping layer. Large energy shift was also observed at high temperature (800–850 °C) annealing.

Table 1 listed the surface stoichiometry (As/Ga ratios) that calculated from Ga and As surface peaks. The As/Ga ratio decreases to 0.48 for samples under annealing temperature of 750 °C. This is due to arsenic evaporation on surface during the annealing process [9]. The ratio increases to 0.8 for 850 °C annealing, which may be due to Ga atoms desertion at high temperature.

Table 1
The As/Ga ratio calculated from the surface peaks of RBS/w channeling spectra

Sample	GaAs wafer	NA	650	750	800	850
As/Ga	1	0.7	0.67	0.48	0.52	0.81

4. Conclusion

Combining the channeling results and PL measurements, we conclude that annealing causes InAs/GaAs QDs strain to relax and atomic intermixing to take place. A significant blueshift and a large dechanneled yield were observed respectively in PL and channeling measurement, indicating that the cubiclike InAs dots mixed with surrounding GaAs and gradually to become $In_xGa_{1-x}As$ with tetragonal distortion after high temperature annealing. The atomic intermixing likely plays a bigger role than the strain effect in determining the photoluminescence emission spectrum when the QDs are annealed.

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