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X-ray diffraction and optical characterization of interdiffusion in self-assembled InAs/GaAs quantum-dot superlattices

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We report on the characterization of thermally induced interdiffusion in InAs/GaAs quantum-dot superlattices with high-resolution x-ray diffraction and photoluminescence techniques. The dynamical theory is employed to simulate the measured x-ray diffraction rocking curves of the InAs/GaAs quantum-dot superlattices annealed at different temperatures. Excellent agreement between the experimental curves and the simulations is achieved when the composition, thickness, and stress variations caused by interdiffusion are taken in account. It is found that the significant In–Ga intermixing occurs even in the as-grown InAs/GaAs quantum dots. The diffusion coefficients at different temperatures are estimated. © 2000 American Institute of Physics.

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Recently, it has been demonstrated that electronic structures of self-assembled quantum dots (QDs) could be greatly turned by thermal annealing.^{1–11} As a result of significantly modified electronic structure of the QDs, up to ~ 260 meV strong blueshift and down to ~ 12 meV pronounced narrowing of the inhomogeneously broadened luminescence peak due to spontaneous size distribution of QDs can be achieved. On the other hand, when the multifold stacked QDs are grown they have the potential to form vertically ordered QD superlattices,^{12,13,20–22} if the spacer layers are thin enough.^{14,18} Such QD superlattices have unique electronic and optical properties and hence are of great interest, in particular for fabrication of high performance QD lasers.^{15,16,19} Using ultrahigh vacuum cross-sectional scanning tunneling microscopy, Lita *et al.*²¹ found significant In–Ga intermixing in InAs/GaAs QD superlattice *in situ* annealed at 620°C for 22.5 min. Nevertheless, a detailed knowledge on atom interdiffusion mechanism in QD superlattice structures is still lacking and is obviously needed from the view of device applications.¹⁷

The information on atom interdiffusion at interfaces of heterostructures can be obtained under certain circumstances from analysis of the high-resolution x-ray diffraction patterns.²³ In this letter, we focus on investigation of thermally induced interdiffusion in InAs/GaAs QD superlattice with the high-resolution x-ray rocking curve technique. Ten periods of InAs/GaAs QD superlattice were annealed at different temperatures. The dynamical theory was employed to simulate measured x-ray diffraction data. Excellent agreement between the experiment and simulation is achieved when the strong interdiffusion effect is considered.

The InAs dots were grown on semi-insulating GaAs (001) substrate via SK growth mode by molecular beam epitaxy. Followed growth of a 300 nm GaAs buffer layer, a 3 nm $\text{In}_{0.17}\text{Ga}_{0.83}\text{As}$ quantum well (QW) layer was grown. After growth of a 100 nm GaAs spacer layer, a 10 cycled alternating 0.6 nm InAs self-assembled QD layer and 10 nm GaAs barrier layer was grown, and finally a 20 nm GaAs cap layer was grown. A more detailed growth description has been published elsewhere.²⁴ Transmission electron microscopy characterization shows that stacks of the QDs are vertically aligned.²⁴ The annealing procedures and PL system description can be found in our previous publications.⁶ High-resolution x-ray diffraction measurements were performed with a Bede Scientific D³ x-ray diffraction system.

Figure 1 shows the 10 K photoluminescence (PL) spectra measured from the samples annealed at different temperatures. The broad peak at lower energy is from the InAs QD layers, while the sharp one at higher energy is from the In–GaAs QW layer. Strong blueshift (up to 300 meV) of the luminescence peak of the InAs dot layers can be seen when the annealing temperature increases to 850°C . Furthermore, significant narrowing (down to 16.6 meV) of the emission peak of the QD layers occurs with increasing annealing temperature. The strong blueshift of the QD emission peak with annealing temperature has been attributed to the enhanced gallium–indium interdiffusion between the InAs QD layers and the GaAs barrier layers. As a result of In–Ga atom interdiffusion induced by heating, the Ga concentration in the QDs will increase, and thus the band gap of the QDs increases. Finally, we observe the blueshift of the QD luminescence peak. Another result of the In–Ga interdiffusion is the change of the inhomogeneous strain distribution inside the QD layers and at the interface. The variation of the strain also changes the electronic structure of the dots²⁵ and hence

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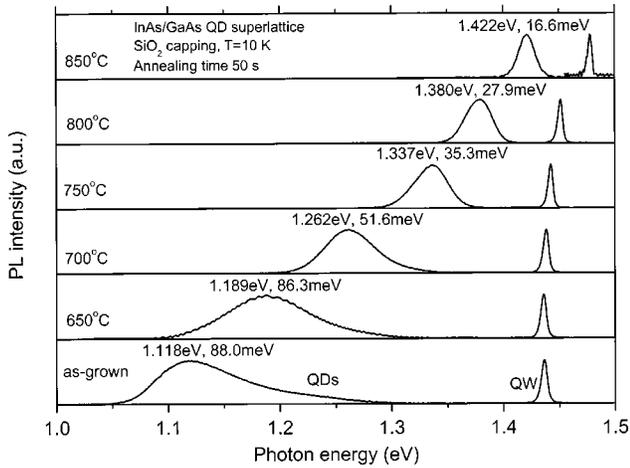


FIG. 1. PL spectra of the InAs/GaAs QD superlattice samples annealed at different temperatures. The broad emission peak at lower energy is from the QDs while the narrow one at higher energy is from the reference InGaAs quantum well.

causes change of the QD luminescence peak. In the following paragraph, we will discuss the x-ray diffraction data and the simulation results based on the Takagi–Taupin formalism.

Figure 2 shows the measured x-ray diffraction rocking curves (solid lines) of the as-grown sample and the samples annealed at different temperatures. For the as-grown sample and the samples annealed at relatively lower temperatures, clear satellite peaks appear in the rocking curves, indicating good periodicity of the samples. It should be noted that additional fine structures could be observed between the main satellite peaks. This demonstrates that the InAs/GaAs QD superlattice studied in the present work is highly periodic. Now let us look at the x-ray diffraction data of the sample annealed at 850 °C. Clearly, the higher order satellite peaks disappear. This is a typical x-ray diffraction fingerprint of the strong interdiffusion in superlattice structures.²³

According to the Takagi–Taupin theory, a differential equation for the amplitude ratio D_s/D_i of the scattered wave D_s and incident wave D_i can be obtained²³

$$-i \frac{dX}{dT} = X^2 - 2\eta X + 1, \quad (1)$$

where X , η , and T are complex quantities given by

$$X = \sqrt{\frac{F_s^-}{F_s}} \sqrt{\frac{\gamma_s}{\gamma_i}} \frac{D_s}{D_i},$$

$$\eta = \frac{-b(\theta - \theta_B) \sin 2\theta_B - \frac{1}{2}\Gamma F_0(1-b)}{\sqrt{|b|} C \Gamma \sqrt{F_s F_s^-}},$$

and

$$T = \frac{\pi C \Gamma \sqrt{F_s F_s^-} d}{\lambda \sqrt{|\gamma_i \gamma_s|}},$$

respectively. Here $\Gamma = r_e \lambda^2 / \pi V$, and $b = \gamma_i / \gamma_s$. r_e is the electron radius (here $2.8179403 \times 10^{-15}$ m is taken as its value), λ is the wavelength ($\lambda = 1.544390 \times 10^{-10}$ m for the Cu $K_{\alpha 1}$ line) of the x ray, and V is the volume of the unit cell. γ_i and γ_s are the direction cosines of the incident and

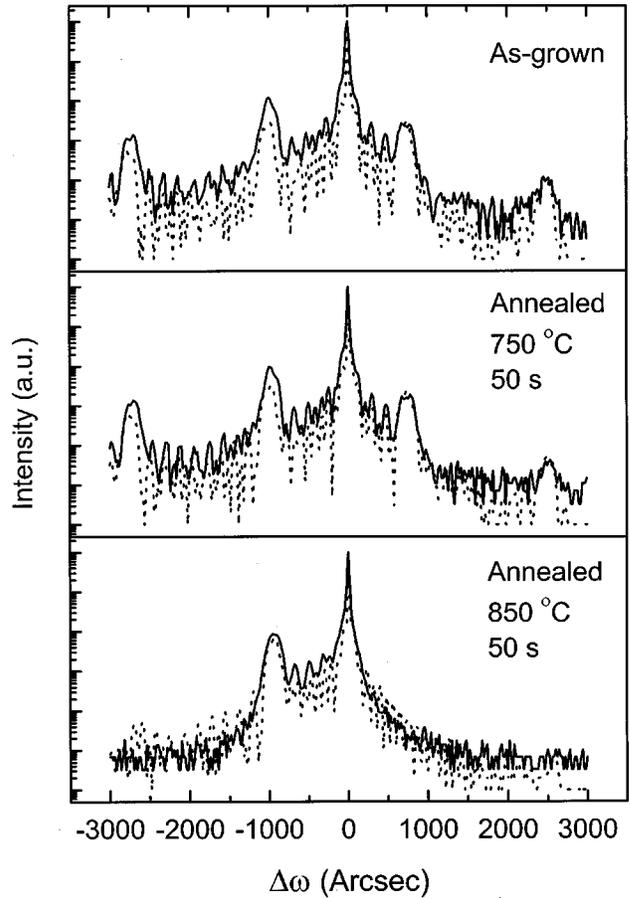


FIG. 2. Measured x-ray rocking curves (solid lines) of the samples annealed at different temperatures. Dot lines are the simulation results by using the dynamical theory.

scattered beams with respect to the internal surface normal, d is the crystal thickness, and F_0 is the structure factor for (000). F_s and F_s^- are the structure factors for (hkl) [here for (004)] and for $(\bar{h}\bar{k}\bar{l})$, respectively. θ_B is the Bragg angle, and C denotes the polarization factor ($C=1$ for σ polarization and $C=|\cos 2\theta_B|$ for π polarization). The solution of Eq. (1) is given by

$$X_d = \eta + \sqrt{\eta^2 - 1} \frac{S_1 + S_2}{S_1 - S_2}, \quad (2)$$

where $S_{1,2} = (X_0 - \eta \pm \sqrt{\eta^2 - 1}) \exp(\mp iT \sqrt{\eta^2 - 1})$. For layered structures the recursion X_d usually starts with the infinite thick substrate ($d \rightarrow \infty$)²⁶

$$X_\infty = \eta - \text{sign}(\text{Re}(\eta)) \sqrt{\eta^2 - 1}. \quad (3)$$

The reflectivity R_s is finally given by

$$R_s = \left| \frac{\gamma_i}{\gamma_s} \cdot \frac{D_s}{D_i} \right|^2 = \left| \frac{F_s}{F_s^-} \right| |X|^2. \quad (4)$$

The rocking curve of the sample is determined by the reflectivity R_s as a function of the deviation parameter η .

In order to simulate the x-ray rocking curves with the Takagi–Taupin formalism described above, the strain induced by lattice mismatch must be taken into account. However, the strain around the quantum dots and at interface is too complex to describe it in terms of a simple expression.^{25,27} As an alternative method, the average strain

is approximately viewed as a constant. We use the following equation to calculate the strain in the dot layers:

$$\varepsilon_{II}^L = \frac{a_{II}^L - a_0^S}{a_0^S}, \quad (5)$$

$$\varepsilon_{\perp}^L = \frac{a_{\perp}^L - a_0^S}{a_0^S}. \quad (6)$$

Here index L is for the heteroepitaxial layer and S for the substrate, \perp is for the growth direction, and 0 is for the unstrained case. For the thin film coherently grown on a thick foreign substrate, $a_{II}^L = a_0^S$ and hence $\varepsilon_{II}^L = 0$, a_{\perp}^L is calculated by

$$a_{\perp}^L = \frac{C_{11} + 2C_{12}}{C_{11}}(a_0^L - a_0^S) + a_0^S, \quad (7)$$

where C_{11} and C_{12} are the stiffness coefficients.

In the simulation of the x-ray rocking curves, the InAs/GaAs QD superlattice approximates to a strained $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ superlattice. During the rapid thermal annealing, the In–Ga atom interdiffusion will result in an increase of gallium concentration in QDs, and causes a decrease of the net strain in the dot layers. The effective thickness d_w (comparable to the In–Ga interdiffusion length) of the dot layers increases with increasing annealing temperature. Correspondingly, the thickness of the barrier layers d_B will decrease. However, $d_w + d_B$ remains unchanged during the interested annealing temperature range.

The dot lines shown in Fig. 2 are the simulation results to the experimental x-ray rocking curves. Excellent agreement between the experimental data and the simulations can be seen. For the case of the as-grown sample, the best simulation is achieved when we use the parameters of thickness 1.2 nm and indium concentration 50% rather than 0.6 nm and 100%. This reveals that significant In–Ga intermixing already occurs even in the as-grown InAs/GaAs QD superlattice. Our results are consistent with that reported by Lita *et al.*²¹ For the sample annealed at 650 °C for 50 s, 1.5 nm thickness and 40% In concentration were obtained through simulation. When the sample was annealed at 750 °C for 50 s, d_w extends to 1.6 nm while the In concentration decreases to 36%. When the annealing temperature increases to 850 °C, d_w is found to dramatically extend to 9 nm. The average In concentration decreases to only 5.8%. Such strong In–Ga intermixing may result in the disappearance of the QDs. In fact, for the InAs/GaAs single layer QD sample annealed at 850 °C for 50 s, our transmission electron microscopy observation⁶ directly demonstrates the destruction of the QDs by strong In–Ga interdiffusion. Assuming that d_w obtained from the simulation to the x-ray rocking curves equals the diffusion length of Ga or In, we can estimate the diffusion coefficient D by using the equation $\Delta d_w = d_w(T) - d_w(\text{as-grown}) = \sqrt{D(T)\tau}$. Here τ is the annealing time. The calculated diffusion coefficients are $1.8 \times 10^{-17} \text{ cm}^2 \text{ s}^{-1}$ at 650 °C, $3.2 \times 10^{-17} \text{ cm}^2 \text{ s}^{-1}$ at 750 °C, and $1.2 \times 10^{-14} \text{ cm}^2 \text{ s}^{-1}$ at 850 °C. It is obvious that the diffusion coefficients depend strongly on annealing temperature. In fact, the annealing temperature dependence of the diffusion coefficient is known to be exponential.²⁸

In summary, the InAs/GaAs QD superlattices were annealed at different temperatures. The In–Ga atom interdiffusion was studied by means of PL and x-ray diffraction techniques. The dynamical theory was employed to simulate the x-ray rocking curves. Excellent agreement between the experimental data and the simulations was achieved when the strong In–Ga intermixing is taken into account. Our results show that the high-resolution x-ray diffraction rocking curve technique is a sensitive tool to investigate the interdiffusion in periodic self-assembled QD superlattices.

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