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Strain study of self-assembled InAs quantum dots by ion channeling technique

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Ion channeling technique using MeV C^{++} ions was used to study strain in self-assembled InAs quantum dots (QDs) buried in GaAs matrix. Because of the use of heavy ions, we were able to observe an angular shift in the angular scan of the In signal relative to that of the Ga/As signal. This provided a direct evidence that the InAs lattice is larger than that of GaAs in the growth direction. Combining the channeling results in [100] and [110] directions and the photoluminescence emission spectrum, we conclude that the InAs QDs are under tensile strain in the growth direction and have the same lattice constant as that of GaAs in the lateral direction. Thermal annealing causes the strain to relax, first in the growth direction and then in the lateral direction as the annealing temperature increases. The photoluminescence spectra of the QDs before and after annealing indicate, however, that composition intermixing also takes place during annealing and is the dominant factor in determining the band gap energy of the QDs. © *2006 American Institute of Physics*.
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INTRODUCTION

InAs self-assembled quantum dots (QDs) have been extensively used in optoelectronic devices, such as semiconductor lasers[1–3] and detectors.[4,5] The formation of self-assembled QDs is driven by the strain originated from the lattice mismatch between InAs and GaAs.[6] The physical properties of the dots are strongly dependent on the amount of strain in and around the dots.[7] However, the strain of these buried dots is difficult to measure and so far only a very few experimental studies have yielded quantitative information on the strain.[8–10] MeV ion channeling has long been used in analyzing the atomic ordering in crystal structures and has been successfully applied for the study of strain in quantum wells. By using angle scan, one is able to calculate the amount of strain in quantum wells by measuring the shift of the scan curve from the quantum wells and that of the substrate. In this work, the strain distribution of self-assembled InAs QDs in GaAs was studied using channeling of MeV C^{++} ions. Because of the use heavy ions, we were able to measure the angular shift of the channeled signal due to the lattice displacement in the strained QDs. The strain relaxation of the QDs after thermal annealing was also studied using this technique.

Selen *et al.*[11] have studied the strain of buried InAs QDs using He^+ MeV ion channeling. However, angular shift in the channeling spectrum caused by strains was not obtained. Because of the limited height of QDs, the length of the atomic string along the ion path is not large enough to cause ion steering and a shift in the angular scan. Consequently, it was very difficult to obtain the strain information of the QDs by the angular scan. The experiment only provided evidence for the presence of strain in and around the QDs. The detailed information of strains is largely unknown.

During ion channeling study, shadow cones are formed by the surface atoms. Only a few layers of atoms close to the surface may contribute to the surface peak. Let us call the thickness of this surface layer the shadow cone length. In the present case, where a thin but strained layer is buried in a crystal matrix, the interface atoms cast a shadow cone on the strained layer underneath from the incoming particles. If we want to investigate the strain by the strain induced steering of channeling ions, the thickness of the thin strained layer has to be larger than the shadow cone length. Then the shift in the angular scan of the backscattered yield provides information of the strain in the strained layer.

Basically, the length of the shadow cone depends on shadow cone radius R_c, which is given by

$$R_c = 2\sqrt{\frac{Z_1 Z_2 e^2 d}{E}}, \tag{1}$$

where Z_1 and Z_2 are the atomic numbers of the incoming particle and the target atom, respectively, E is the energy of the incoming particle, and d is the lattice spacing along the channeling direction. Generally, the shadow cone length is reduced if R_c is large.[12] For H^+ or He^+ ions, R_c is usually pretty small which gives rise to a large shadow cone length. This, however, posts a problem for the quantum dot samples

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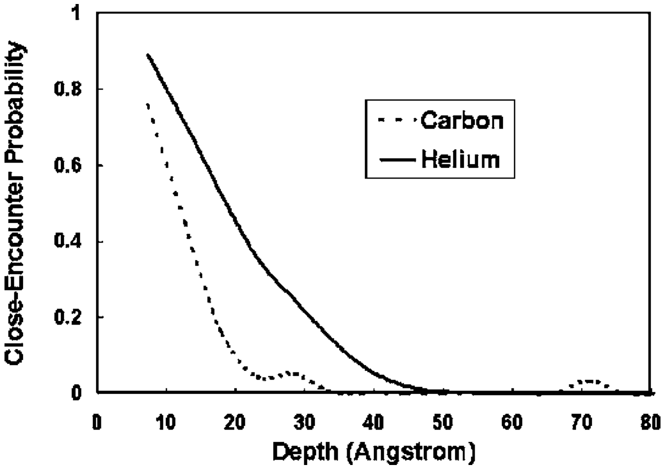

FIG. 1. Calculated result of close-encounter probabilities of C^{++} (dot line) and He^{+} (solid line) ions impinging on a [100] GaAs surface vs the depth from the surface.

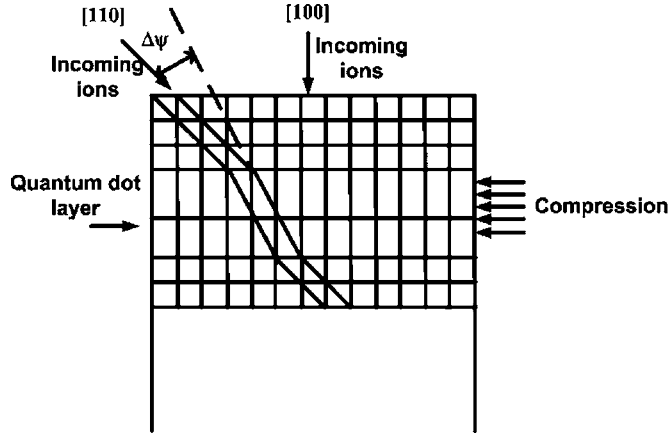

FIG. 2. Schematic diagram of how a buried strained layer is probed by ion beam.

because the height of the quantum dots, typically only a few nanometers, is smaller than the shadow cone length. Equation (1) reveals that the shadow cone radius can be increased by reducing the incoming ion's energy or using heavier ions. Former method, such as medium energy ion scattering (MEIS), has been shown to be useful in analyzing surface structure of thin films.[13,14] But it only probes the surface region that makes it unsuitable for deeply buried thin films, such as QDs. In this study, we chose to use a heavier ion, C^{++}, as incident particles. The shadow cone radius is increased and the length reduced. We were able to use the shift of the angular scan curves to study the strain of the buried QDs.

To understand the effect of atomic mass of incoming particles on the shadow cone length, we calculated the shadow cone length in GaAs along the [100] direction using 4 MeV He^{+} and C^{++} as incoming particles using the simulation package, FLUX 7, authored by Smulders and Boerma.[15] The calculated result is shown in Fig. 1. The solid and the dotted lines are the close-encounter probabilities of He^{+} and C^{++} ions, respectively. The horizontal axis is the depth from the surface. The small peaks in the carbon close-encounter probability at 28 and 72 Å were caused by the trajectory of the channeling ions which are bounced back and forth between the adjacent atomic rows. We can see that the shadow cone length is greatly reduced when C^{++} is used. At a depth of 15 Å, the close-encounter probability drops more than 100% because of the use of C^{++}. This provides us the opportunity to look at the strain distribution in the thin QD layer.

EXPERIMENTS

The samples of InAs QDs used in this study were grown by a Varian GenII molecular beam epitaxy (MBE) system. A buffer layer of 500 nm GaAs was grown first on a semi-insulating GaAs (100) substrate. Then InAs QD layers (each with 2.6 monolayers) were grown at a temperature of 520 °C. All samples were capped with a 50 nm thick GaAs layer. Arsenic pressure was $(4-5)\times 10^{-6}$ Torr. Growth rates were 1 μm/h for GaAs and 0.056 μm/h for InAs. From the atomic force microscope (AFM) image, the density of QDs was determined to be approximately 1×10^{10} cm^{-2}. Ruthterford backscattering (RBS) with channeling measurement was performed 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 ion beam current density was 200 nA/cm^{2} on the target. The sample was mounted on a three-axis goniometer with an angular resolution of less than 0.01°. Backscattered particles were collected by a passivated implanted planar silicon (PIPS) detector at 160° with respect to the direction of the incident beam. The energy resolution of the system, determined by fitting the GaAs edge of the energy spectrum, was around 60 keV. This value is low enough to separate the surface peaks of Ga and As in the aligned spectrum (104 keV). To compliment the ion channeling study, we also performed low-temperature (25 K) photoluminescence measurement using the 514.5 nm line of an argon ion laser. The signal was collected using an InGaAs detector and lock-in techniques.

RESULTS AND DISCUSSION

The channeling experiment was performed along the [100] direction, which is normal to the sample surface, and the [110] direction, 45° to the surface normal direction. An angle scan for the [100] beam was performed along the $(01\bar{1})$ surface. Another angular scan was performed for the [110] direction beam along the (001) surface. In this way we were able to probe the strain in the QDs in both the vertical and lateral directions. Figure 2 shows schematically how the channeling beam probes a buried strain layer.

We first looked at a sample with a single QD layer buried under a 50 nm GaAs cap. Figures 3(a) and 3(b) show the angular scan of the In and the Ga/As signals for the as-grown sample along the [100] and [110] directions. While the In signal is from the QDs and the wetting layer, the Ga/As signal was taken from the top 40 nm of the sample. No significant difference was observed between the curves of In and Ga/As in the [100] direction. However, an angular shift was observed in the [110] scan, which indicates a lattice distortion in the buried QD layers. It should be mentioned that the In signal from the wetting layer does not play any

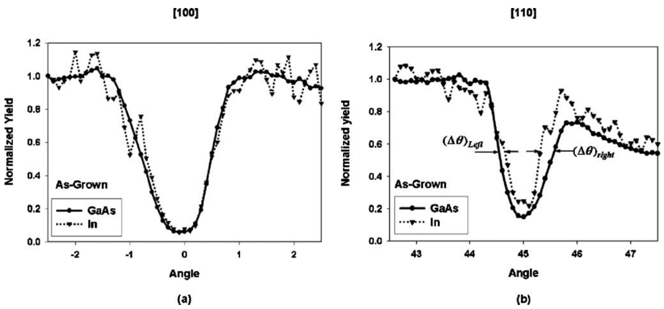

FIG. 3. Angular scan spectra along the (a) [100] and (b) [110] axes of the as-grown QD sample. The angular shift was determined by the relative change in angle position of the half maximum of the In and Ga/As curves on either side of the channeling curves. From the sign of $(\Delta\theta)_{\text{right}}-(\Delta\theta)_{\text{left}}$, we determine which direction the curves shift relative to each other. No obvious difference was observed between the curves of In and Ga/As in the [100] direction. In the [110] direction, we observe an angular shift of the In signal relative to the Ga/As signal toward the [100] direction.

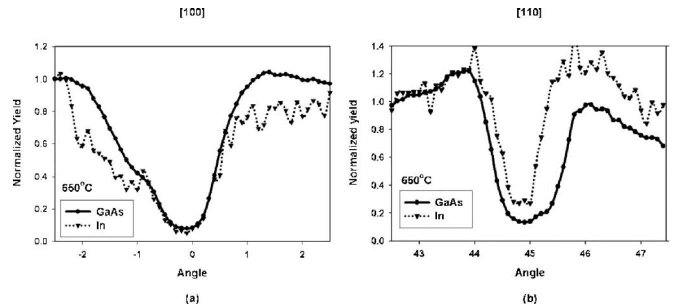

FIG. 4. Angular scan spectra of the QDs annealed at 650 °C. The In and Ga/As signals again match in the [100] direction. In the [110] direction, the angular shift of the In signal becomes larger than that of the as-grown sample.

significant role. The strain from the single monolayer of InAs is not enough to cause any change in the angular scan curve.

The size of the dots, measured from transmission electron microscope (TEM) and AFM images, was about 4 nm in height and 20 nm in width. The calculated shadow cone lengths are 4 and 2.5 nm in the [100] and [110] directions, respectively. In the [100] direction, because it is perpendicular to the strained layer, no angular shift in the angular scan can be obtained. Furthermore because the shadow cone length is about the same as the height of the QDs, according to the ion channeling theory,[12] the In atoms in the QDs behave like In impurities in a GaAs matrix. The channeling behavior of the In signal from the QD layer is determined by the position of In atoms in the flux pattern emerged from the overlying GaAs capping layer. If the In atoms deviate from their equilibrium positions, they could cause the dip of the angular scan to be narrower. The fact that the angular scans of In and Ga/As are nearly identical indicates that there is no strain relaxation in the QDs in the lateral direction. For channeling along the [110] direction, because the shadow cone length is smaller than the QD size, it becomes possible to observe the lattice distortion directly from the shift of the angular scan spectrum. From Fig. 3(b), we indeed see a slight shift of the In signal relative to the Ga/As signal toward the [100] direction. This shift provides a direct evidence that the lattice of the InAs QDs is larger than that of the GaAs matrix in the [100] or the crystal growth direction.

The effect of thermal annealing on strain relaxation in QDs was then studied. The as-grown sample was annealed at 650 and 750 °C for 30 s with a rapid thermal annealing furnace. The angular scan spectra after the sample was annealed at 650 °C are shown in Figs. 4(a) and 4(b). In the [100] direction, we again did not see any difference between the In spectrum and the Ga/As spectrum. In the [110] direction, the angular shift of the In signal, however, becomes larger than that of the as-grown sample. So the InAs lattice remains the same in the lateral direction but becomes larger, less strained, in the growth direction after annealing. In other words, we start to see some strain relaxation in the vertical direction.

The angular scan spectra of the sample annealed at 750 °C are shown in Figs. 5(a) and 5(b). The shape of the dip in the angular scans for the In signal in the [100] direction is clearly different from that of Ga/As. It has become narrower, which is clear evidence of displaced In lattice. In the [110] direction, the angular scan spectra are similar to those of the sample annealed at 650 °C. So annealing at 750 °C caused the strain to relax not only in the vertical direction but also in the lateral direction. From these observations, we reach the following conclusion. The as-grown QDs have the same lattice constant as that of GaAs in the plane perpendicular to the growth direction. In the growth direction, the InAs lattice is larger than that of GaAs. After 650 °C annealing, the InAs lattice of the QDs becomes larger or relaxed in the growth direction, but in the in-plane direction, the QDs remain strained with the lattice constant the same as that of GaAs. After 750 °C annealing, however, the InAs lattice of the QDs not only relaxes in the growth direction but also becomes relaxed in the lateral direction.

So from the channeling result, we know that the lattice constant of the InAs QDs is larger than that of GaAs in the vertical direction. But whether they are under compressive strain or tensile strain is unknown. The effect of strain on the band gap energy of the QDs can be expressed as[16]

$$E_{g,\text{QD}} = E_{g,\text{QD,unstrain}} + \Delta E_{g,\text{strain}}, \tag{2}$$

where the strain induced band gap change $\Delta E_{g,\text{strain}}$ is expressed as

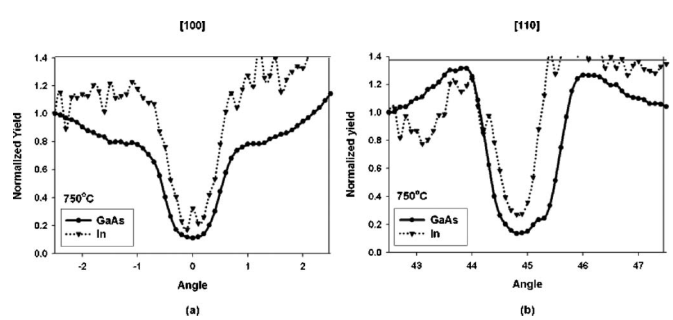

FIG. 5. Angular scan spectra of the QDs annealed at 750 °C. In the [100] direction, the In signal is narrower than the Ga/As signal. In the [110] direction, the angular scan spectra are similar to those of the sample annealed at 650 °C.

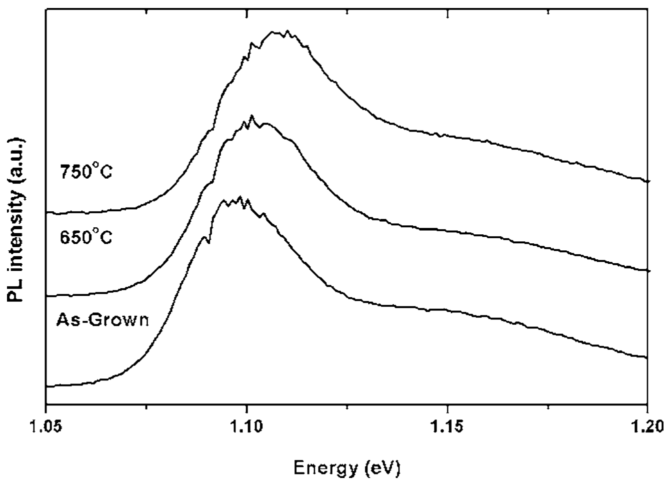

FIG. 6. Photoluminescence spectra at 25 K of QDs before and after annealing.

$$\Delta E_{g,\text{strain}} = \alpha(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) + \beta\sqrt{[(\varepsilon_{xx} - \varepsilon_{yy})^2 + (\varepsilon_{yy} - \varepsilon_{zz})^2 + (\varepsilon_{zz} - \varepsilon_{xx})^2]/2}. \quad (3)$$

The first term is the hydrostatic component and the second term the biaxial component. α and β, deformation potential coefficients of the hydrostatic and the biaxial components, are -6.08 (α) and -1.8 (β) for InAs. ε_{ij} $(i,j=x,y,z)$ are elements of the strain tensor. Since we have seen that the in-plane lattice constant of InAs is the same as that of the GaAs matrix for the as-grown sample, the lateral strain components become

$$\varepsilon_{xx} = \varepsilon_{yy} = \frac{a_{\text{InAs,strain}} - a_{\text{InAs,bulk}}}{a_{\text{InAs,bulk}}} = \frac{a_{\text{GaAs}} - a_{\text{InAs,bulk}}}{a_{\text{InAs,bulk}}}. \quad (4)$$

Substituting in the lattice constants of GaAs and InAs, we obtain $\varepsilon_{xx}=\varepsilon_{yy}=-0.067$. Equation (3) is then reduced to

$$\Delta E_{g,\text{strain}} = 0.694 - 7.88\varepsilon_{zz}, \quad (5)$$

and the QD band gap energy becomes

$$E_{g,\text{QD}} = E_{g,\text{QD,unstrain}} + 0.694 - 7.88\varepsilon_{zz}. \quad (6)$$

The band gap energy of QDs, $E_{g,\text{QD}}$, can be obtained from the photoluminescence measurement. Figure 6 shows the photoluminescence spectra of the QD sample before and after annealing. For the as-grown sample, $E_{g,\text{QD}}$ is determined to be 1.1 eV. So the strain of the QDs in the growth direction is

$$\varepsilon_{zz} = \frac{E_{g,\text{QD,unstrain}} - 0.406}{7.88}. \quad (7)$$

Since $E_{g,\text{QD}}$ is larger than the bulk InAs band gap energy (0.416 eV) because of the quantum size effect, ε_{zz} must be positive. In other words, the QDs are tensile strained in the z direction.

The ion channeling results provide us with information on lattice distortion of the QDs before and after annealing. But they cannot tell us anything on the composition changes. The photoluminescence measurement, however, provides information on the band gap energy, which is influenced by both strain and composition.

The photoluminescence spectra shown in Fig. 6 indicate that the emission peak of the QDs had a clear blueshift after the sample is annealed. This result, however, cannot be explained by the strain changes discussed above. From the channeling result, we know that annealing causes the strain of the QDs to relax, or the lattice to enlarge, first vertically then laterally. So the band gap should shrink after annealing. This should cause the photoluminescence emission to have a redshift. So the observed blueshift in the photoluminescence spectrum is obviously contrary to what is expected from the strain analysis. The reason for this discrepancy is attributed to the composition intermixing between the QDs and the surrounding GaAs during annealing as previously reported in Ref. 17. The intermixing obviously plays a bigger role than the strain effect in determining the photoluminescence emission spectrum when the QDs are annealed.

CONCLUSION

Ion channeling technique using MeV C^{++} ions was used to study strain in self-assembled InAs QDs buried in GaAs matrix. Because of the use of heavy ions, we were able to observe an angular shift in the angular scan of the In signal relative to that of the Ga/As signal. This provided a direct evidence that the InAs lattice is larger than that of GaAs in the growth direction. Combining the channeling results in the [100] and [110] directions and the photoluminescence emission spectrum, we conclude that the InAs QDs are under tensile strain in the growth direction and have the same lattice constant as that of GaAs in the lateral direction. Thermal annealing causes the strain to relax, first in the growth direction and then in the lateral direction as the annealing temperature increases. The photoluminescence spectra of the QDs before and after annealing indicate, however, that composition intermixing also takes place during annealing and is the dominant factor in determining the band gap energy of the QDs.

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[1] N. Nuntawong, Y. C. Xin, S. Birudavolu, P. S. Wong, S. Huang, C. P. Hains, and D. L. Huffaker, Appl. Phys. Lett. **86**, 193115 (2005).
[2] H. Chen, Z. Zou, O. B. Shchekin, and D. G. Deppe, Electron. Lett. **36**, 1703 (2000).
[3] N. N. Ledentsov *et al.*, Phys. Rev. B **54**, 8743 (1996).
[4] D. Pan, E. Towe, and S. Kennerly, Appl. Phys. Lett. **73**, 1937 (1998).
[5] S. Y. Wang, S. D. Lin, H. W. Wu, and C. P. Lee, Appl. Phys. Lett. **78**, 1023 (2001).
[6] A. A. Darhuber *et al.*, Appl. Phys. Lett. **70**, 955 (1997).
[7] M. Grundmann, O. Stier, and D. Bimberg, Phys. Rev. B **52**, 11969 (1995).
[8] A. Krost, F. Heinrichsdorff, D. Bimberg, A. Darhuber, and G. Bauer, Appl. Phys. Lett. **68**, 785 (1996).
[9] K. Zhang, Ch. Heyn, W. Hansen, Th. Schmidt, and J. Falta, Appl. Phys. Lett. **77**, 1295 (2000).

[10]H. S. Lee, J. Y. Lee, T. W. Kim, and M. D. Kim, Appl. Phys. Lett. **83**, 2256 (2003).
[11]L. J. M. Selen, L. J. van IJzendoorn, M. J. A. de Voigt, and P. M. Koenraad, Phys. Rev. B **61**, 8270 (1998).
[12]L. C. Feldman, J. W. Mayer, and S. T. Picraux, *Materials Analysis by Ion Channeling* (Academic, New York, 1982).
[13]J. Vrijmoeth, P. M. Zagwijn, J. W. M. Frenken, and J. F. van der Veen, Phys. Rev. Lett. **67**, 1134 (1991).
[14]T. C. Q. Noakes, P. Bailey, P. K. Hucknall, K. K. Donovan, and M. A. Howson, Phys. Rev. B **58**, 4934 (1998).
[15]P. J. M. Smulders and D. O. Boerma,Nucl. Instrum. Methods Phys. Res. B **29**, 471 (1987).
[16]S. L. Chuang, *Physics of Optoelectronic Devices* (Wiley, New York, 1995), Chap. 4.
[17]A. O. Kosogov *et al.*, Appl. Phys. Lett. **69**, 3072 (1996).