Stacked InAs Self-Assembled Quantum Dots on (001) GaAs Grown by Molecular Beam Epitaxy

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We report the stacked InAs quantum dot (QD) structures separated by GaAs barrier layers grown by molecular beam epitaxy on (001) GaAs substrate. The InAs QDs were vertically aligned up to 9th stack. The decrease of photoluminescence intensity probably due to the appearance of dislocation is observed. The optical memory effect of InAs QDs on photocurrent of Schottky barrier diode is observed for the first time.

1. Introduction

Recently, self-assembled quantum dots (QDs), which are made from InGaAs, InAs on GaAs¹⁻⁵⁾ and other strained systems, have attractive much interest in fundamental physics $^{6,7)}$ and device application.⁸⁾ Although the study on the QD density and its size control has been reported, the vertical alignment of the QDs along the growth direction⁹⁾ has received little attention. Multiple stacked QD structures have advantage on the application of tunneling mechanism to electronic and optical devices: single electron tunneling devices, quantum dot lasers and so on. Since the crystal quality of the stacked structures is not clear, much effort has to be done to investigate the strained system. In this paper, we study the stacked InAs QD structures grown on (001) GaAs substrate using molecular beam epitaxy (MBE) characterized by transmission electron microscopy (TEM) combined with energy dispersive X-ray (EDX) analysis and photoluminescence (PL) at 77 K.

We also report the optical memory effect of InAs QDs buried in Schottky barrier diode that shows the possibility of high density optical memory using QDs.¹⁰)

2. Experimental

We prepared 5 samples: (i) a single layer QDs with nominal 1.8 monolayer (ML) InAs sandwiched with GaAs layers, (ii) a double stacked QDs with nominal 1.8 ML InAs layers separated by a spacer of 10-nm-GaAs, (iii) a multiple stacked (50 periods) QDs with nominal 1.8 ML InAs layers with 15-nm-GaAs spacer layers, (iv) a multiple stacked (50 periods) QDs with nominal 2.5 ML InAs layers with 15-nm-GaAs spacer layers. We also fabricated a Schottky barrier diode with a single QD structure of nominal 1.8 ML InAs followed by an AlAs barrier for photocurrent measurement. The InAs QDs are grown by MBE at the growth temperature of 510 °C.

For the photocurrent measurement, we made the diode with transparent Au (30 nm) electrode. The QDs were excited with cw Ti:sapphire laser at $1.06 \,\mu$ m whose

photon energy is higher than the PL peak energy at room temperature. The diode was biased by a built-in voltage.

3. Results and Discussion

3-1 TEM observation of stacked InAs QDs

The TEM cross-sectional bright field image of the double stacked QDs (sample (ii)) is shown in Fig. 1. The lateral size of QDs is less than 20 nm. The upper QDs are vertically aligned to the position of the lower QDs separated by a 10-nm-GaAs barrier layer. This suggests that the upper QD is preferably assembled on the place where the strain induced by the lower QD is strongly localized.

We also observed the multiple stacked QDs (sample (iii)) with 1.8 ML InAs layers and 15-nm GaAs as shown in Fig. 2. Although the GaAs layer is thicker than that of the double structure, the vertical alignment up to the 4th layer (maximum up to 6th layer) was observed. The multiple stacked structure with 2.5 ML InAs layers, sample (iv), showed the alignment up to 9th layer. The absence of InAs QDs for further stacking can be



Fig. 1 Transmission electron microscopy (TEM) crosssectional bright field image of a double stacked InAs(1.8 ML)/GaAs quantum dots (QDs) on (001) GaAs.

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considered to be due to an In segregation into the upper GaAs layers and subsequent decrease of lattice-mismatch at InAs/Ga(In)As interface.





We observed the multiple stacked sample of (iii) with TEM combined with EDX analysis along the growth direction. Figure 3 shows the scan across one of the bottom QDs. Two relatively In-rich region appeared corresponding to top and bottom of the QD. This result is quite in contrast with the other report.⁵) The distribution of the strain field around the InAs QDs can be considered to be the same as that of In-content. This implies that the vertical alignment of the InAs QDs can be enhanced for some extent of periods which is accordance with the results of Fig. 1 and Fig. 2. The lower In-rich region is considered to be due to the wetting layer of InAs. The origin of the upper In-rich region is not known.



Fig. 3 EDX analysis on relative In, Ga, and As content from below-QD (left) to above-QD (right) in the first stack of a multiple stacked (50 periods) InAs(1.8 ML)/GaAs QDs on (001) GaAs.

The stacked QDs observed may be applicable to single electron device using multiple-tunnel junctions.¹¹)

3-2 PL of stacked InAs QDs

Figure 4 shows the PL of the samples with single, double, and multiple structures at 77 K with excitation power of 1 mW at 514 nm. The PL peaks appear between 1.14 and 1.19 eV. The integral PL intensity of the multiple stacked structure is 1/100 of those of single and double structures. The integral intensity of the multiple structure with 2.5 ML InAs layers is further small compared to the multiple 1.8 ML structure. The decrease of PL intensity is considered to be due to dislocations (not shown in Fig. 2) observed in the stacked layers by TEM which play a role as nonradiative recombination centers. A subpeak at 1.06 eV is also observed. This subpeak is regarded to come from the fairly thick (~4 nm) quantum well structures (not shown in Fig. 2.).



Fig. 4 Photoluminescence (PL) spectra of single, double, and multiple InAs/GaAs QDs at 77 K with 1 mW excitation (514 nm). All samples consist of 1.8 ML InAs. The PL of the multiple structure was magnified by 50 times.



Fig. 5 The excitation power dependence on the PL peak energy. All samples consist of 1.8 ML InAs.

Figure 5 shows the excitation power dependence on the PL peak energy. The PL peak energy of the single layer structure changes little, and the variation of the energy shift increases as the stack number increases. This reflects the layer-to-layer inhomogeneity of the multiple stacked structure. The full width at half maximum of PL spectrum is also found to be power-dependent

3-3 Optical memory effect of InAs QDs

Figure 6(a) shows the band diagram of Schottky barrier diode including single InAs QDs. Although multiple stacked QDs may be preferable for practical applications, we used simple QDs for the start. This was irradiated by sequential two optical pulses with interval of t. After the first pulse irradiation, electron-hole pairs are created in the InAs QDs and electrons escape from the QDs by tunneling or thermionic emission. The holes stay in the QD because the tunneling probability is much less than that of electrons due to their large effective mass and also due to the AlAs barrier inserted beside the left GaAs to make holes more difficult to tunnel through or recombine with electrons.

When the second optical pulse is irradiated, the residual holes decrease the photocurrent by the second pulse due to the saturation of absorption. The nonlinearity in absorption is expected to be larger than quantum well because only two electrons can be excited in QD considering spin degrees of freedom.

Figure 6(b) shows photocurrent response of the two pulse measurement we used. Figure 6(c) shows the interval-dependence of [I(0)-I(t)]/I(0). It delayed with time constant of 0.47 msec which is the retention time of the optical memory. We consider the retention time as a recombination time of residual holes with electrons of the degenerated n-type region that are thermally excited more than 0.46 eV from the Fermi level. The retention time is in a good agreement with the calculated result based on the model using the energy separation of 0.46 eV. Although the excitation photon energy is not resonant to the peak energy of the InAs QDs, optical memory effect on photocurrent of Schottky barrier diode is observed for the first time.

4. Conclusion

We observed the stacked InAs QDs structures on (001) GaAs substrate grown by MBE. The InAs QDs were vertically aligned up to 9th layer. The decrease of PL intensity probably due to the appearance of dislocation is also observed. Preliminary results of the optical memory effect of InAs QDs on photocurrent of Schottky barrier diode was observed for the first time.





Fig. 6 Optical memory effect of InAs QDs buried in Schottky barrier diode: (a) schematic energy band diagram, (b) the photocurrent response to the sequential two optical pulses (250µsec/div), (c) time-dependence of the photocurrent ratio of second to first optical pulses.

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