Energy transfer in multi-stacked InAs quantum dots

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1. Introduction

A great deal of research interest exists in the excitation energy transfer between semiconductor quantum dots (QDs), and many investigations of interlayer excitation energy transfer between QDs have been conducted based on carrier tunneling [1-4], dipole-dipole interactions [5], optical near-field interaction [6,7], and carrier hopping [8,9] in self-assembled QDs systems. It is important to evaluate the energy transfer length for applications such as signal processors and solar cells because the performance of these devices is based on these phenomena. However, it is difficult to evaluate energy transfer length systematically in a self-assembled QD system, because QDs are distributed randomly on the semiconductor surface. Even with QDs aligned vertically in a stacked structure, fabrication of a stacking structure with multiple layers is difficult because of the accumulation of strain energy.

We have developed a method of stacking InAs QDs grown on InP(311)B using a strain compensation scheme. In this scheme, spacer layers with a lattice constant slightly smaller than that of the substrate are used to embed the QD layers. Using this method, we successfully stacked 150 InAs QD layers without any degradation in QD quality. We believe that there is no limit to the number of QD layers that can be stacked using this method [10], [11]. This enables the use of the optimum number of QD layers for any given application. In addition, we can change emission wavelength by changing QDs growth thickness without degrading crystal quality, as long as the strain compensation condition is satisfied. Therefore, this material system is considered to be ideal for investigating energy transfer length among QDs. In this paper we investigate energy transfer in highly stacked QDs using the photoluminescence (PL) method.

2. Experiments

All samples were fabricated using conventional solid-source molecular beam epitaxy (MBE). We fabricated the stacked QD structures on an InP(311)B substrate. The value of the lattice constant of InP lies between the values of those of GaAs and InAs. This means that the lattice constant of InGaAlAs can be varied continuously around that of InP by controlling its composition. Thus, strain compensation can be achieved by capping the InAs QDs with an InGaAlAs layer whose lattice constant is slightly smaller than that of the InP substrate. We defined the strain-compensation conditions as:

\[ \varepsilon_{QD} = (a_{QD} - a_{sub})/a_{sub} \]  \hspace{1cm} (1)

\[ \varepsilon_{s} = (a_{s} - a_{sub})/a_{sub} \]  \hspace{1cm} (2)

\[ \varepsilon_{QD} \cdot \varepsilon_{s} = -d_{QD} \cdot \varepsilon_{s} \]  \hspace{1cm} (3)

where \( d_{QD} \) and \( d_{s} \) are the thicknesses of the deposited QDs and spacer layers, respectively, and \( a_{QD} \), \( a_{sub} \), and \( a_{s} \) are the lattice constants of the InAs, InP, and InGaAlAs spacer layers, respectively. This definition is based on the simple approximation that the total strain energy of a set, containing one layer of QDs and one spacer layer, is zero. First, we fabricated two 60-layer stacked samples with \( d_{QD}/d_{s} = 3-ML/15 \) nm and \( d_{QD}/d_{s} = 4-ML/20 \) nm to check the emission wavelength. We then fabricated modulated sample, continuously stacked N layers/one layer/ N+1 layers where N and N+1 layers correspond to a 3-ML QD layer and one layer corresponds to a 4-ML QD layer. We fabricated three samples of N=5, 10 and 20. Post-growth surface morphology was observed using an atomic force microscope (AFM) under normal atmospheric conditions. Photoluminescence (PL) measurements were carried out using the 532-nm line of a YVO laser, 250-mm monochromator, and an electrically cooled PbS detector.

3. Results and discussions

Figure 1 shows the PL spectra of 3-ML and 4-ML QDs measured at room temperature. Four-ML QDs shows longer wavelength emissions at 1607 nm, which corresponds to the ground state of 4-ML QDs depending on size of QDs. The larger size of 4-ML QDs is also confirmed by AFM measurements. There are small peaks in the shorter wavelength region of the main peak corresponding to the excited states of 4-ML QDs. This peak energy is the same as the main emission of 3-ML QDs (ground state, 1484 nm). Therefore, the energy transfer from 3ML QDs to 4-ML QDs is enhanced when 3-ML and 4-ML QDs exist in the same sample.

Figure 2 shows the PL spectrum for modulated samples N=5, 10 and 20. The ground state emission of 4-ML QDs shows a large intensity even if there is only one layer of 4-ML QDs in these samples. This clearly shows that energy...
transfer occurs from small QDs to large QDs. These amplified PL emission of 4-ML QDs were observed in all samples we fabricated (N= 5, 10 and 20).

We evaluated a ratio of PL intensity per dot layer as follows,

$$\frac{PL(\text{large QD})/layer}{PL(\text{small QD})/layer} \quad (4)$$

Where PL(small QD) and PL(large QD) means PL intensity of 3-ML QDs and 4-ML QDs. Figure 3 shows the N dependence of the ratio of PL intensity per dot layer. The ratio of PL intensity per dot layer increased with increasing N then decreased with increasing N. The maximum ratio of PL intensity per dot layer is around N=10. This result implies that long-range energy transfer should occur in these samples. The energy transfer length should depend on the condition of excitation intensity and temperature, and this should be the subject of a detailed investigation in the near future.

4. Conclusions
We fabricated a modulated stacked QD structure to investigate energy transfer among QDs. Energy transfer from small QDs to large QDs was clearly observed. Long-range energy transfer can be considered from the measurement of N dependence of PL intensity.

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References