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LETTER

Dependence of the microstructural and the optical properties on the GaAs spacer thickness in InAs/GaAs double quantum dots grown by using the Indium-flush procedure

I. Y. Jung · Y. M. Park · Y. J. Park · J. I. Lee · T. W. Kim

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Potential applications of quantum dots (QDs) have driven extensive efforts to grow high-quality QDs on semiconductor substrates by using various techniques [1]. Optoelectronic quantum devices utilizing QDs can be fabricated because QDs have discrete artificial atomic energy levels [2]. Thus, the microstructural and the optical properties of self-assembled QD systems have been particularly attractive for many years because of the potential application of QDs to optoelectronic devices [3–8]. In particular, the optical properties of QDs have been widely investigated because of interests in both fundamental physical properties and promising applications, such as QD lasers [9, 10], QD infrared photodetectors [11, 12], and QD memory devices [13] operating at lower current and at higher temperature. Among the many quantum dot structures, coupled quantum dot structures consisting of two smaller band-gap wells, which are different in band-gap energy and are separated by a thin embedded barrier, are currently receiving considerable attention for promising applications in electronic and optoelectronic devices [14–18]. Since the microstructural and the optical properties of coupled QDs are very important for optoelectronic devices based on QD structures, studies concerning the physical properties are still necessary in order to fabricate high-efficiency coupled QD devices.

Many works on the formation and the physical properties of InAs/GaAs QDs grown by using the Stranski–Krastanov (S–K) growth mode have been performed. Even though some works on the sizes and the shapes of vertically stacked self-assembled QDs grown by using an indium-flush procedure have been performed [19–21], studies concerning the dependence of the microstructural and the optical properties on the GaAs spacer thickness in InAs/GaAs closely coupled double QDs grown by using the indium-flush method have not yet been reported. While the size of the upper QD in a coupled double QD grown by using the S–K mode is larger than that of the lower QD due to a strain field [22], the sizes of the two QDs in the coupled QDs grown by using the S–K mode, together with an indium-flush procedure, are almost the same. Since the top and the bottom dots of a double QD having similar sizes and shapes is important for investigating the coupling effects of double QDs and their promising applications in electronic and optoelectronic devices, the indium-flush method, which is an effective way to control precisely the sizes and the thicknesses of the QDs, should be very useful for investigating the coupling behavior in double QDs [19]. Furthermore, very few works on the dependence of the activation energy of double QDs on the spacer thickness have been done.

This letter reports the dependences of the microstructural and the optical properties on the GaAs spacer thickness in InAs/GaAs double QDs with different GaAs spacer layers grown by using molecular beam epitaxy (MBE) with an indium-flush method. Transmission electron microscopy (TEM) measurements were performed to characterize the microstructural properties of the InAs/GaAs double QDs, and photoluminescence (PL) measurements were carried out in order to investigate the dependences of the full width at half maximum (FWHM), the peak position of the interband transitions, and the activation energy on the

I. Y. Jung · Y. M. Park · Y. J. Park · J. I. Lee
Nano-Device Research Center, Korea Institute of Science and Technology, P.O. Box 131, Cheongryang, Seoul 130-650, Korea

I. Y. Jung · T. W. Kim (✉)
Advanced semiconductor research center, Division of Electronics and Computer Engineering, Hanyang University, 17 Haengdang-dong, Seongdong-gu, Seoul 133-791, Korea
e-mail: twk@hanyang.ac.kr

GaAs spacer layer thickness in InAs/GaAs double QDs. Furthermore, the optical properties of double QDs grown by using the indium-flush method were compared with those of double QDs grown by using the S-K mode.

The two kinds of the double QD samples used in this work were grown on semi-insulating (100)-oriented GaAs substrates by using MBE, together with an indium-flush method, and consisted of the following structures: a 300-nm undoped GaAs buffer layer deposited by using MBE, 3-monolayer (ML) InAs QDs, a GaAs thin-film spacer layer (5 nm or 40 nm) deposited by using the indium-flush step, 3-ML InAs QDs, and a 20-nm undoped GaAs layer deposited by using MBE. The rationale for the indium-flush step was to eliminate any trace of In in the second half of the GaAs spacer, ensuring nominally identical growth conditions for each InAs QD layer. The indium-flush step was performed by ramping the substrate temperature over 30 s from 510 °C to 610 °C, keeping it at 610 °C for 70 s, and then ramping it back to 510 °C over 100 s. In comparison with MBE and atomic layer epitaxy, the indium-flush deposition technique can produce a more uniformly sized stacking of InAs double QDs with disk shapes. The depositions of the GaAs barriers and the InAs QDs were done at substrate temperatures of 600 °C and 490 °C, respectively. The growth rates of the GaAs layers and the InAs QDs were 0.5 ML/s and 0.09 ML/s, respectively. InAs/GaAs double QDs were also grown by using the conventional MBE method for comparison with those grown by using MBE, together with the indium flush.

The TEM observations were performed in a JEM 2000EX transmission electron microscope operating at 200 kV. The samples for the TEM measurements were prepared by cutting and polishing with diamond paper to a thickness of approximately 30 μm and then by argon-ion milling at liquid-nitrogen temperature to electron transparency. The PL measurements were carried out using a 75-cm monochromator equipped with a liquid-nitrogen-cooled InGaAs detector. The excitation source was the 514-nm line of an Ar^+ laser with a power of 30 mW, and the sample temperature was controlled between 17 K and 250 K by using a He displex system.

Figure 1 shows high-resolution TEM (HRTEM) images of InAs/GaAs double QDs with a spacer thickness of 5 nm. While the lateral size of the InAs/GaAs double QDs with a spacer thickness of 5 nm is larger than that of those with a spacer thickness of 40 nm, both of the InAs/GaAs double QDs have similar disk configurations. The typical lateral sizes of the InAs QDs with disk shapes were between 10 nm and 15 nm, and the heights of the QDs were about 3 nm. While the sizes of the upper QDs in coupled InAs QDs grown by using the conventional S–K mode were significantly larger than the sizes of the lower QDs [22], the sizes of the upper QDs in coupled QDs grown by using an indium-flush deposition were almost the same as those of the lower QDs, as shown in Fig. 1. The strain field from an InAs single dot layer immediately penetrates the grown GaAs spacer layer, after which the dot size decays rapidly with distance [23, 24]. When the thickness between double dot layers is less than the decay length, the level of the strain increases [24].

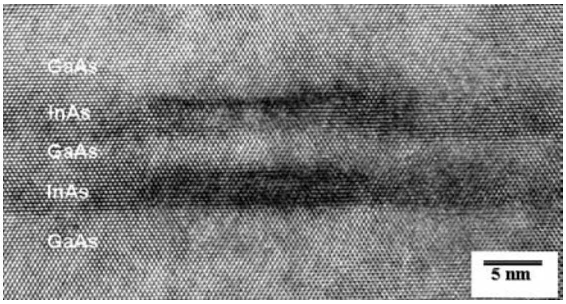

Fig. 1 High-resolution transmission electron microcopy images of InAs/GaAs double quantum dots with spacer thickness of 5 nm

Figure 2 shows PL spectra measured at 70 K for InAs/GaAs double QDs with spacer thicknesses of (a) 5 nm and (b) 40 nm. The PL spectra at 70 K for the InAs/GaAs double QDs with spacer thicknesses of 5 nm and 40 nm show broad dominant luminescence peaks at 1.221 eV and 1.227 eV, which are related to interband transitions from the ground electronic subband to the ground heavy-hole subband (E_1-HH_1) of the InAs QDs. The peak position corresponding to the (E_1-HH_1) transition shifts to higher energy with increasing GaAs spacer thickness. Although this blue shift with increasing GaAs spacer thickness may be attributed to an electronic coupling of QDs along vertical columns, our HRTEM images indicate that it is due to a one-dimensional double lattice resulting from a decrease in the size of the InAs QDs resulting from an uncoupling

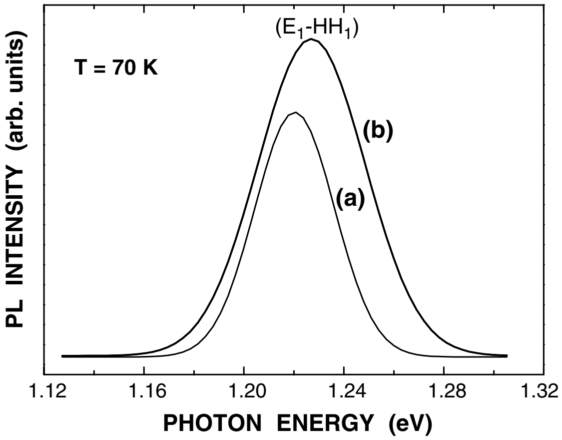

Fig. 2 Photoluminescence spectra measured at 70 K for InAs/GaAs double quantum dots with spacer thicknesses of (**a**) 5 nm and (**b**) 40 nm

effect. A similar behavior for structures containing double dot layers with different space separations was reported elsewhere [25]. The FWHMs of the peaks at 1.221 eV and 1.227 eV are 42.82 meV and 31.99 meV, respectively. The FWHM of the InAs/GaAs double QDs with a spacer thickness of 5 nm is much smaller than that of InAs/GaAs double QDs with a spacer thickness of 40 nm due to the coupling effect.

The PL spectra measured at several temperatures for the InAs/GaAs double QDs with GaAs spacer thicknesses of 5 nm and 40 nm are shown in Fig. 3. The temperature dependences of the FWHMs and the peak positions of the (E_1-HH_1) transitions of the PL spectra for the InAs/GaAs QDs with spacer thicknesses of 5 nm and 40 nm are shown in Fig. 4. As the values for the energy gaps of the InAs QDs decrease with increasing temperature, the PL peaks corresponding to the (E_1-HH_1) transitions shift to the low-energy side with increasing temperature. This variation in the peak position with changing temperature can be attributed to lattice dilation and to the electron-lattice interaction. As predicted by a previous theoretical analysis [26], the red shift clearly follows the Varshni relation.

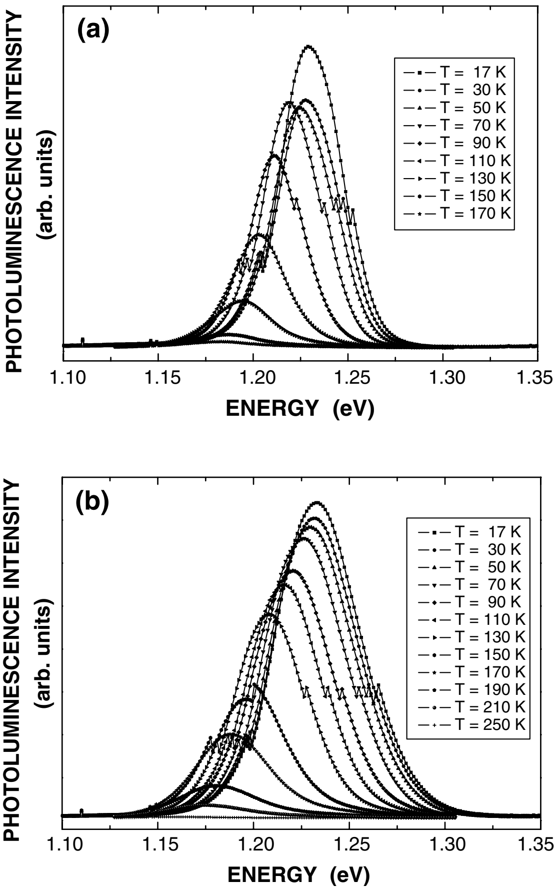

Fig. 3 Photoluminescence spectra measured at several temperatures for the InAs/GaAs double quantum dot arrays with GaAs spacer thicknessess of (**a**) 5 nm and (**b**) 40 nm

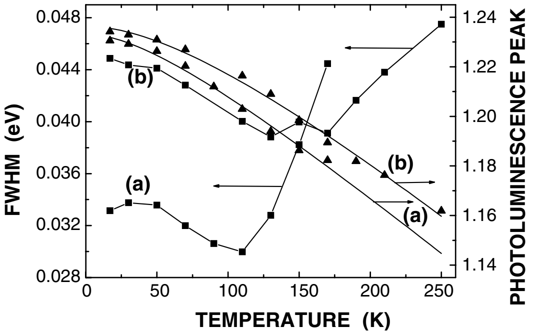

Fig. 4 Full width at half maximum and energy of the photoluminescence peak as functions of the temperature for the (E_1-HH_1) interband transitions in InAs/GaAs double quantum dots with spacer thicknesses of (**a**) 5 nm and (**b**) 40 nm. The filled rectangles and triangles represent the full width at half maximum and the energy of the PL peak for the (E_1-HH_1) interband transitions, respectively. The solid lines are guides for the eyes

As shown in Fig. 4, the FWHM of the peak related to the (E_1-HH_1) transitions also strongly depends on the temperature. The values of the FWHMs show a nonmonotonic behavior with increasing temperature. While the FWHM values of the PL spectra for InAs/GaAs double QDs with spacer thicknesses of 5 nm and 40 nm decreases with increasing temperature up to 110 K and 130 K, respectively, they increase above 130 K and 150 K, respectively. At low temperatures, the PL line shape for InAs/GaAs double QDs is predominantly affected by the inhomogeneous distribution of cluster sizes and consists of emissions from many dots of different sizes. When the temperature is increased, the carriers in the QDs are thermally excited gradually. The thermalized carriers can repopulate nearby QDs. The repopulation process will occur more frequently for dominant QD sizes because of the Gaussian distribution of QD sizes, so the PL linewidth should decrease with increasing temperature [27]. When the temperature is high enough, the effect of electron–phonon scattering becomes the dominant contribution, so the PL linewidth starts to increase with increasing temperature.

In order to investigate the quenching behavior of the PL spectrum, the temperature dependence of the integrated PL intensity was analyzed. Figure 5 depicts the temperature dependence of the integrated PL intensity for InAs/GaAs double QDs with spacer thicknesses of (a) 5 and (b) 40 nm. As the temperature is increased, the PL intensity corresponding to the confined electron peak is given by [28]

$$I = I_0/[1 + C\exp(-E_A/k_BT)], \tag{1}$$

where I is the integrated PL intensity, I_0 is the PL intensity at 0 K, C is the ratio of carriers that contribute to photoluminescence with activation to those that contribute without activation, E_A is the activation energy, and k_B is the Boltzmann constant. Equation (1) assumes that the

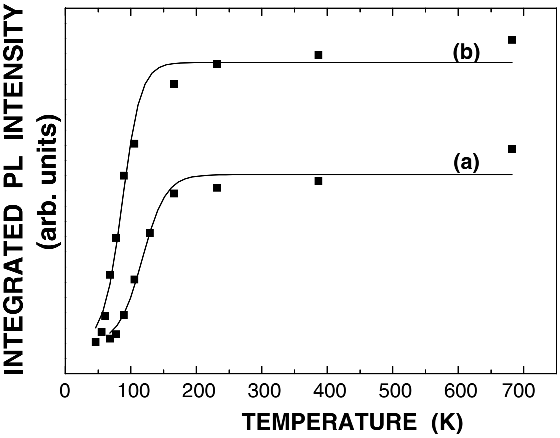

Fig. 5 Integrated photoluminescence intensities as a function of the reciprocal temperature for InAs/GaAs double quantum dots with spacer thicknesses of (**a**) 5 nm and (**b**) 40 nm. The solid lines are guides for the eyes

dominant mechanism for the PL intensity change with increasing temperature is thermal activation of carriers from the QDs to the barrier region. From Eq. (1), the activation energies of the InAs/GaAs double QDs with spacer layer thicknesses of 5 nm and 40 nm are 50 and 70 meV, respectively. The activation energy of InAs/GaAs double QDs with a GaAs spacer thickness of 40 nm is larger than that of the InAs/GaAs double QDs with a GaAs spacer thickness of 5 nm. When the InAs/GaAs QDs are grown in closely stacked double layers with a GaAs spacer thickness of 5 nm. The strain effect between the QDs with disk shapes might reduce the activation energy of the electrons confined in the InAs QDs. This behavior is totally different from the variation in the activation of electrons confined in InAs QDs with pyramid shapes [1].

In summary, the dependences of the microstructural and the optical properties on the GaAs spacer thickness in InAs/GaAs double QDs were investigated. The results of TEM measurements on the InAs/GaAs double QDs grown by using indium flush deposition showed that the InAs/GaAs double QDs had disk shapes and that the sizes of the InAs QDs were almost the same. The PL peak position corresponding to the (E_1-HH_1) transition shifted to higher energy with increasing GaAs spacer thickness due to the coupling effect, and the activation energy of the electrons confined in the InAs QDs decreased with decreasing GaAs spacer thickness due to the strain effect. The present observations can help improve our understanding of the dependences of the microstructural and the optical properties on the GaAs spacer layer thickness in InAs/GaAs double QDs.

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