Indium Segregation and its Influence to the Quantum Structures of InAs/GaAs Self-assembled Quantum Dots

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Abstract

Indium segregation during the growth of InAs/GaAs self-assembled quantum dots was investigated in a gas-source molecular beam epitaxy. With reducing the growth temperature of the GaAs cap-layer, a remarkable red-shift of the ground state energy of the 1.8 ML InAs quantum dots was observed due to suppression of the indium segregation. The number of confined states in the dots was increased from 3 to 5 and the energy separation between the neighbor states was also increased from 20 meV to 50 meV. State filling from both conventional and magneto photoluminescence revealed the multiple states in the single quantum dots. Micro-photoluminescence from a few dots showed groups of distinct emission lines from different states in the dots. Emission lines at the high-energy side possess characters of excited states with degeneracy and fine splitting.
KEYWORDS: InAs/GaAs, self-assembled quantum dots (SADs), photoluminescence, gas-source molecular beam epitaxy (GS-MBE), indium segregation, excited state.

1. Introduction

Recently, self-assembling of semiconductor quantum dots by Stranski-Krastanov mode has attracted a lot of attention. The method is simple, while the dots are coherently organized and defect-free [1-6]. A typical structure is InAs/GaAs self-assembled dots (SADs), where the 7% lattice mismatch leads to the formation of 3-dimensional InAs islands having a rather narrow size distribution. So far, their electronic states have been investigated by optical [7-9] and capacitance [10,11] spectroscopy, and device applications have been demonstrated. [12,13] However, the quantized energy states as well as the carrier relaxation processes in SADs are subjects of further studies. Since the electron and hole states in the dots are dependent on their size, composition, shape or strain distribution, great care should be taken in the control of the growth processes. Besides this, intrinsic growth phenomena such as indium segregation should be controlled. In this study, indium segregation during the GaAs cap-layer growth was investigated in a gas-source molecular beam epitaxy (GS-MBE), and its influence to the quantum structures of the InAs/GaAs dots were studied by conventional, magneto and microphoto-luminescence (PL).

2. Experimental

All the samples were grown in a GS-MBE. Gallium, indium and aluminum were used as group Ⅲ sources while arsine cracked at 950℃ was used as a group V source. The substrates were GaAs (100) vicinal surfaces inclined by 0.5° toward the [011] direction. Before the growth, the substrate was heated up to about 620℃ for 3 minutes under an arsine flow of 6 sccm. The substrate temperature was controlled by using a pyrometer with a temperature resolution of less than 1℃. A 500 nm GaAs buffer was first grown at 600℃. After the substrate was cooled to 520℃, InAs was deposited up to 1.8 ML in nominal thickness as the RHEED pattern showed change from streaky to spotty. After a growth interruption of 60 seconds down to 450℃ (sample **(a)**) or 480℃ (sample **(b)**) or of 10 seconds at 520℃ (sample **(c)**), the InAs islands were embedded in a 150 nm GaAs cap-layer. A sample (d) was prepared with the cap-layer grown at 520℃ after 80 seconds growth interruption in order to reduce the dot density for the luminescence studies from single quantum dots [14], and the temperature was ramped up to 600℃ in 5 minutes during the GaAs cap-layer growth. Mesa structures of the sample (d) were prepared by wet etching in a H_3PO_4:H_2O_2:H_2O solution over a lithographic mask pattern of 10 μm×10 μm squares

separated by 80μm from each other. The natural side-etching and faceting made the mesas shrink to 3μm×3μm squares. Conventional PL, magneto PL and micro-PL were carried out to clarify the quantum states in the SADs. Micro-PL was performed by using an optical microscope to restrict the laser spot diameter on to a mesa and the signals were detected with a charge coupled device (CCD) camera with the system resolution less than 0.3 meV.

3. Results and discussions

Figure 1 shows the 77 K PL spectra of the samples (a), (b), (c) and (d). All four spectra showed multiple peaks. The luminescence around 1.42eV or 1.47 eV is identified to be from the wetting layer and the peak at 1.51 eV comes from GaAs. With increasing growth temperature for the GaAs cap-layer, PL spectra of both the dots and the wetting layer showed blue-shift. With the increase in the excitation intensity, PL intensities at the higher energy side of the multiple peaks increase as a result of state filling. For example, PL spectra of the sample (a) under weaker excitation intensities were also given in Fig. 1. These envelopes with multiple peaks can be well fitted by Gaussians. The fitting peak energies of the Gaussians of the luminescence spectra for the samples (a), (b) and (c) are shown in Fig. 2. These Gaussian components were considered to be the ground and excited states of SADs with inhomogeneous broadening. With decreasing the growth temperature of the GaAs cap-layer, the ground state energy of the SADs was greatly decreased, while the spacing between the neighboring states was increased from 20 meV to 50 meV. We found in our previous study that compositional intermixing between InAs islands and the underlying GaAs layer is not obvious even by long growth interruption [14]. The blue-shift of the PL peaks with increasing the cap-layer growth temperature is considered to be caused by surface segregation of indium during the GaAs cap-layer growth [15,16]. Indium segregation changed the size, shape and composition of the dots through In-Ga intermixing. Ramping up the growth temperature at the start of the GaAs

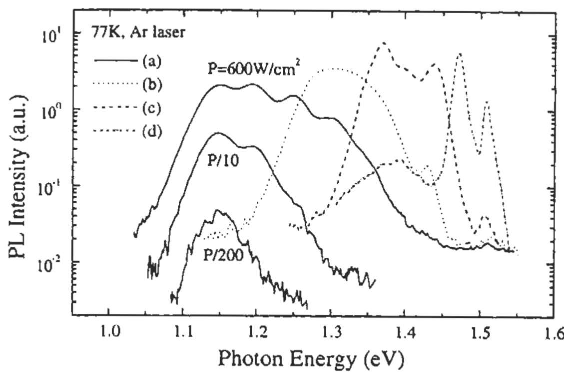

Fig.1 77K PL of 1.8 ML InAs/GaAs SADs samples. InAs islands were all formed at 520℃ but samples **(a)**, **(b)** and **(c)** were embedded in GaAs at 450℃, 480℃ and 520℃ after the growth interruption of 60, 60 and 10 seconds, respectively. The sample **(d)** was embedded at 520℃ after 80 seconds growth interruption and the temperature was ramped up to 600℃ in 5 minutes.

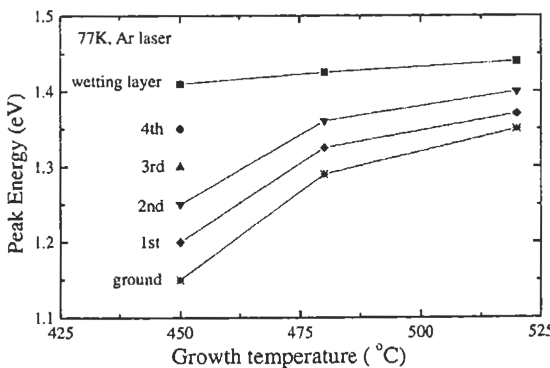

Fig.2 Gaussian fitted 77K PL peak energies from quantum states in 1.8 ML InAs/GaAs SADs. InAs islands were all formed at 520℃ but were embedded in GaAs at 450℃, 480℃ and 520℃ after a growth interruption of 60, 60 and 10 seconds, respectively.

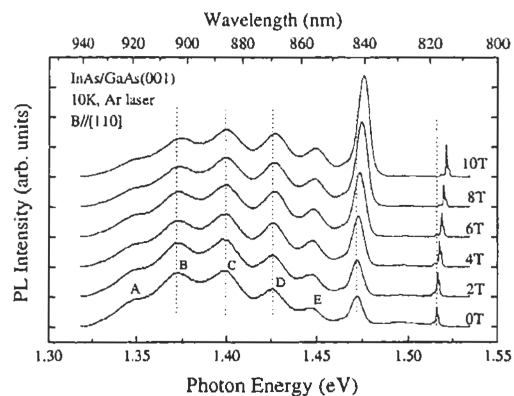

Fig.3 Magneto PL from sample **(d)**. The magnetic field was set parallel to the [011] direction that is perpendicular to the surface normal. Blue-shifts of all the peak energies and changes in their luminescence intensities were observed.

cap-layer growth accelerated the In-Ga mixing in the sample (d), giving rise to further blue-shift for both spectra of the wetting layer and the quantum dots. The increase of the gallium content in the SADs raises the energy of the ground state, while the expansion in their effective confining size results in a reduction of the energy separations among the ground and excited states. This indium segregation process is similar to that observed in the growth of InGaAs/GaAs quantum wells [17]. Under the supression of the indium segregation, InAs/GaAs SADs 39 nm, 29 nm, 22 nm and 15 nm in their average diameters were obtained by controlling the InAs growth temperature. The corresponding number of confined states observed was five, three, two and one, respectively. The details have been published elsewhere [18].

In order to understand the origin of the multiple states, magneto and micro-PL from a few quantum dots were investigated. Since the areal density of the dots in the sample (d) was greatly reduced (~10^8 cm^{-2}) during the growth interruption [14], and the spectra is within the detection range of the CCD camera, it was ideal for this study. Figure 3 shows the influence of the magnetic field to the PL spectra. The magnetic field was set along the [011] direction or perpendicular to the (100) surface normal of the sample. At 0 T, five peaks labeled A, B, C, D and E were clearly distinguished which are originated from the quantum dots. The emissions at 1.52 eV and 1.47 eV that come from the GaAs bulk and the InAs wetting layer, respectively. We have confirmed that the average size of the remaining InAs islands was increased during the growth interruption or annealing, and the PL spectra from the dots shift to the low-energy side while the position of PL from the wetting layer was almost constant [14]. This makes it possible to contain five rather than three states in the quantum dots of sample (d). The PL spectra from the dots can be well fitted by five Gaussians. The full width at half maximum of each component is about 20 meV, and the energy separation between the neighbor peaks is about 20 to 25 meV. With increasing the magnetic field up to 10 T, all the peaks show the diamagnetic blue-shift. The blue-shift of GaAs free and bound excitons is 5 meV, that of the PL from the InAs wetting layer is 3.5 meV, while the blue-shift of the dot emission peaks is about 1.5 meV. These blue-shifts show the enhancement of the carrier confinement by the magnetic field. Since excitons in the quantum dots are strongly confined, the enhanced confinement by the magnetic field has less influence on the excitons in the dots than on the excitons in the wetting layer and GaAs. Also, with increasing the magnetic field intensity, the luminescence intensitis of peak D, E, the wetting layer and GaAs bulk increased while those of peak A, B and C at the low-energy side decreased. We think that some forbidden transitions from the excited states become partially allowed due to the crossing of the excited states in a quantum dot under high magnetic field. These results support our assumption that the five peaks are originated from the ground and four excited states in the quantum dots.

Micro-PL from one of the mesas is shown in Fig. 4 under various excitation intensities ranging from 3 W/cm^2 to 100 W/cm^2. Very sharp PL emission lines from the discrete states in the quantum dots were observed. These lines form about five groups that agree with the multiple peaks in the conventional PL spectra. Line groups at the high-energy side contain more emission lines. With increasing the excitation intensity, some additional PL lines appeared as were marked by the asterisks. Saturation of the line intensities was observed gradually from the low-energy side while those in the high-energy side still grow steadily. These data reveal the presence of level filling to the excited states in single dots and the

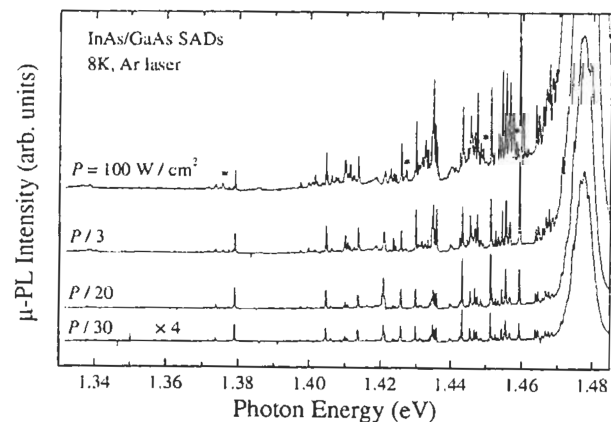

Fig.4 Excitation intensity dependence of a few InAs/GaAs SADs in the sample **(d)** by micro-PL. New emission lines appeared at high excitation intensities and the level filling was revealed.

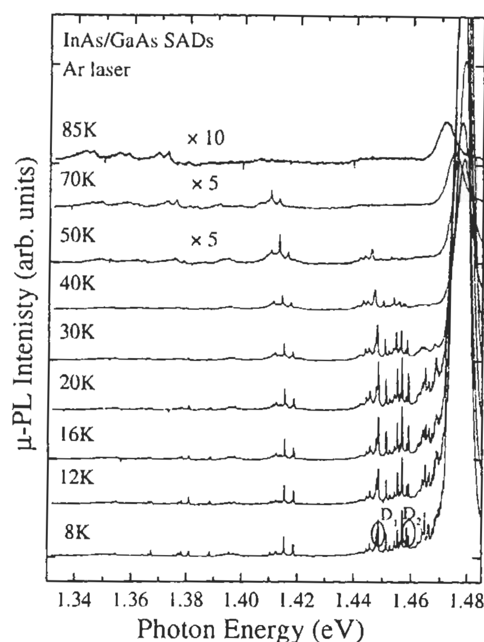

Fig.5 Temperature dependence of a few InAs/GaAs SADs in the sample **(d)** by micro-PL. With increasing the temperature, red-shift of the emission lines from the dots and reduction in their intensities start from the high-energy side were observed. Energy transfer between the two components in the doublets labeled **D1** and **D2** was suggested.

degeneracy of the excited states.

Figure 5 shows the temperature dependence of the micro-PL spectra under constant excitation intensity. Successive red-shifts of the emission lines from the quantum dots were clearly resolved with increasing the temperature. Between 8 to 20K, the energy shifts from dot emissions and that of the wetting layer were negligible, and the intensities of the emission lines were almost constant except those of two doublets labeled **D1** and **D2**. The intensity of the high-energy component of **D1** or **D2** was increased while that of the low-energy component was decreased. Since the energy separation of the two components in the doublets is about 0.6 meV, if transition between the two components is allowed, carrier may transfer to the higher energy component which may has high recombination probability through thermal activated acoustic phonons. It is considered that these fine structures appear because the degeneracy of the excited states is weakly lifted due to the anisotropy of the quantum dots. By further increasing the temperature, the intensities of the emission lines from the quantum dots are reduced successively from the high-energy side due to the depletion of the carriers. Emission from the quantum dots became invisible at above 100 K. Since the scattering probability of the exciton by phonons increases with the increase of temperature, carrier relaxation to the lower states and transitions from the lower states became favorable at high temperatures.

4. Conclusions

In summary, indium segregation during the growth of InAs SADs on GaAs (100) vicinal surfaces was studied by gas-source molecular beam epitaxy. With reducing the GaAs cap-layer growth temperature over the InAs islands, a remarkable red-shift of the ground state energy was observed due to suppression of the indium segregation. The number of confined states in the dots was increased from 3 to 5 and the energy separation between the neighboring states was also increased from 20 meV to 50 meV. State filling from both conventional and magneto photoluminescence (PL) revealed the multiple states in the single quantum dots. Micro-PL from a few dots showed groups of distinct emission lines from different states in the dots. Emission lines in the high-energy side possess characters of excited states with degeneracy and fine splitting.

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