Electrical and optical characterizations of self-assembled quantum dots formed by the atomic layer epitaxy technique

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We investigated the electrical and optical properties of InGaAs self-assembled quantum dots grown using the atomic layer epitaxy (ALE) technique. Dots–in–a–well structures were grown by alternately supplying InAs and GaAs sources on an InGaAs layer and covering with another InGaAs layer. Three samples produced with different numbers of cycles of alternate InAs/GaAs supply were characterized by capacitance-voltage and photoluminescence (PL) measurements. For the ten cycle dots–in–a–well structure, a strong zero-dimensional electron confinement was observed even at room temperature. On the other hand, for the five-cycle structure, the PL results indicate that the InGaAs quantum well structure coexists unstably with premature quantum dots. By comparing the results for samples with different numbers of cycles, we suggest that an ALE dots–in–a–well structure can be formed by the aggregation of In and Ga atoms incorporated into the InGaAs quantum well layer when the number of cycles exceeds the critical number of seven cycles.

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I. INTRODUCTION

In(Ga)As self-assembled quantum dots (QDs) have been the subject of much research due to their potential applications for optoelectronic devices.1–3 In the most popular conventional molecular beam epitaxy (MBE) technique, a strained In(Ga)As layer grown by a continuous source supply evolves into QDs in the Stranski–Krastanov (S-K) growth mode.4 However, different types of QDs grown by an atomic layer epitaxy (ALE) technique have been proposed, and a laser emission from those QDs has been demonstrated.5–7 In this ALE technique, InAs and GaAs are alternately supplied in a number of cycles. The growth rate and quantity of As can be controlled not only by cell temperatures but also by shutter operations. Moreover, migration can be enhanced by introducing an interruption time. Thus, there is a greater chance of obtaining good QD size uniformity with the ALE technique than with the conventional MBE technique via the S-K mode.

In this work, we investigated the electrical and optical properties of quantum dots–in–a–well structures grown by the ALE technique. We produced three samples with different numbers of cycles of the InAs/GaAs source supply, and performed capacitance-voltage (C-V) and photoluminescence (PL) measurements. Both C-V and PL data showed that there was a strong zero-dimensional confinement for the ten-cycle sample even at room temperature. On the other hand, only premature QDs were observed for the five-cycle sample. Based on these observations, we suggest a formation process for ALE dots–in–a–well structures.

II. EXPERIMENTAL PROCEDURES

All the samples used in this study were grown on \( n^+ \)-GaAs (100) substrate using the ALE technique. Dots–in–a–well structures were grown by alternately supplying InAs and GaAs sources on a 5-nm-thick InGaAs layer and covering with another 5-nm InGaAs layer. Growth interruptions were used between the source supply of InAs and GaAs for the enhancement of migration. The InAs and GaAs growth rates were 0.07 and 1 monolayer/s, respectively. The growth sequence of the samples is as follows: Si-doped 0.3-μm-thick GaAs buffer layer \((n \sim 1.2\times10^{18} \text{ cm}^{-3})\), Si-doped 0.1-μm-thick GaAs layer \((n \sim 2\times10^{16} \text{ cm}^{-3})\), undoped 20-nm-thick GaAs layer, ALE dots-in-a-well, undoped 20-nm-thick GaAs layer, and 0.3-μm-thick GaAs layer \((n \sim 2\times10^{16} \text{ cm}^{-3})\). Three different samples with five, seven, and ten cycles of the InAs/GaAs alternate source supply were prepared. For C-V measurements, Au-Schottky contacts were deposited on top of the samples through a shadow mask with 240-μm diameter holes and In ohmic contacts were formed on the backside of the \( n^+ \)-GaAs substrates. C-V characteristics were measured at a fixed frequency of 1 MHz at temperatures of 77 and 300 K. The optical properties of the grown samples were evaluated by PL measurements at various temperatures. An Ar-ion laser with a wavelength of 514.5 nm was used as an excitation source, and the excita-
tion power density was varied from 15 to 300 W/cm². A liquid nitrogen cooled InGaAs detector was employed to detect the luminescence.

III. RESULTS AND DISCUSSION

Figure 1 shows the PL spectra at 300 K for ALE dots-in-a-well structures with different numbers of cycles of the InAs/GaAs alternate source supply. The PL spectrum of the five-cycle sample exhibits a pronounced peak at about 1.34 eV in contrast to the other samples, which originate from the 10-nm In₀.₁Ga₀.₉As quantum well (QW) layers surrounding the QDs. As the number of cycles increases, the intensity of the peak decreases, and it is barely visible in the PL spectrum for the ten-cycle sample. However, another peak associated with QDs at around 1.2 eV becomes stronger and its position shifts to lower energy as the number of InAs/GaAs cycles increases. The PL spectra of the QDs grown by conventional MBE via the S-K mode strongly depend on the density and the size of the QDs and the surrounding materials.⁸ However, in the ALE growth of the dots-in-a-well structure, the formation mechanism of QDs is different from that produced using the S-K mode.⁶ At a low number of InAs/GaAs cycles, an intermediate structure of inhomogeneous chemical composition is expected to exist. When the PL peak denoted as premature QDs in the PL spectrum of the five-cycle sample in Fig. 1 is deconvoluted, an additional PL peak associated with a layer other than the QD or QW layers. This decomposition of the QD peak into two components is more conspicuous in the 15 K spectrum as shown in the upper inset of Fig. 2. We think that this additional peak is associated with the intermediate layer rather than with the excited state of QDs or the so-called wetting layer in S-K QDs.

The evolution of the QDs’ PL peak as a function of the number of InAs/GaAs cycles shows that QDs are not completely formed before ten InAs/GaAs cycles. If the number of InAs/GaAs cycles is insufficient, e.g., five cycles, QDs of optimum size and shape are not yet formed due to the source supply deficit, thus causing premature QDs to coexist with the intermediate layer. The nature of the intermediate layer will be explained in detail in the next paragraph. If the number of cycles increases, a dots-in-a-well structure is formed by the aggregation of In and Ga atoms incorporated into the InGaAs QW layer. Based on Fig. 1, the critical number of cycles is about seven.

Figure 2 shows the PL spectra at 15 K for the five-cycle sample at excitation power densities of 15, 75, and 150 W/cm². The upper inset in Fig. 2 shows a decomposed PL spectrum at an excitation power density of 150 W/cm², and the origin of the three peaks are assigned to the QDs, the intermediate layer, and the InGaAs QW, respectively. The conjectured structure of this sample is schematically drawn in the lower inset. Overall, an InGaAs QW layer is surrounded by GaAs barriers. In the central part of this QW layer, where alternate cycles of the InAs/GaAs source are supplied, premature QDs are formed in the middle. They are surrounded by intermediate layers of nature between QD and QW formed due to partial mixing of the central part and the QW layer. This intermediate layer is different from the wetting layer formed in the conventional MBE via the S-K mode. The wetting layer is a well-defined very thin layer acting as a QW, and the typical full width at half maximum (FWHM) of its PL peak is narrow. Since it serves as a cross-talk path for carriers between QDs, the PL peak from the wetting layer becomes pronounced at high excitation power density. On the other hand, the PL peak from the intermediate layer in the ALE dots-in-a-well structure is broad and is still observable even at low excitation power density as shown in Fig. 2.

The carriers in the premature QDs and the intermediate layers are not strongly confined due to their intermediate nature, while the InGaAs QW is well confined between the GaAs barriers. The PL peak intensity due to the QW layer...
becomes stronger as the excitation power density increases when compared with the other peaks.

The temperature dependence of the three PL peak energies for the five-cycle sample is shown in Fig. 3. The PL peak energy of QDs (filled circles) follows a sigmoidal temperature-dependence pattern, which cannot be explained simply by Varshni’s law. The abrupt decrease in PL peak energy at around 50 K is obvious. The FWHM shown in Fig. 3(b) also drops at the same temperature. These behaviors can be explained by the thermionic emission of photocarriers from the smaller QDs and their repopulation into the nearby QDs with deeper confining potentials. This repopulation process contributes to the reduction of the inhomogeneous broadening of the QDs. The FWHM increases and saturates at higher temperatures (above 150 K) as the carriers escape even from the larger QDs and occupy all the QDs with equal probability.

The PL peak energies due to both the QWs (filled triangles) and the intermediate layer (open squares) as a function of temperature are also shown in Fig. 3(a) for comparison. The temperature-dependent PL peak energies of the QW layer obey Varshni’s law using the parameters of InGaAs, whereas the PL peak energies of the intermediate layer cannot be simply fitted to this law. For S-K QDs produced by the conventional MBE technique, the temperature dependence of the PL peak energy is determined by two carrier transfer processes: the escape of carriers from QDs and carrier transfer between QDs via the wetting layer. These carrier transfer processes explain the sigmoidal temperature dependence of the PL peak energy. For the ALE dots–in–a–well structure, the temperature dependence of the PL peak energy can be explained using the intermediate layer. This layer acts as a transfer path for carriers between QWs and QDs, resulting in the sigmoidal temperature dependence of the PL peak energy from QDs. Moreover, the PL peak energy from the intermediate layer also shows the anomalous temperature-dependence pattern.

PL data show the existence of the intermediate layer in ALE growth at a low number of cycles of alternate InAs/GaAs supply. This intermediate layer has intermediate properties of QWs and QDs, and results from an evolution mechanism of QDs during the formation of InGaAs dots–in–a–well structures. Thus, ALE dots–in–a–well structures are expected to form by the aggregation of islands with compositional variations. Two-dimension-like islands (or pre-mature three-dimension-like islands) are formed from QW structures and evolve into QD structures. Owing to the migration-enhanced growth and reduction of the abrupt surface states between the QDs and the ambient material, the spontaneous formation of high quality QDs is more probable with the ALE technique than with the conventional MBE technique via the S-K mode.

Figure 4 shows the C–V characteristics of three different samples depending on the number of cycles of the InAs/GaAs alternate source supply at 300 K. For the five-cycle sample, only background capacitance was observed due to the GaAs bulk, and no steps related to the existence of QDs were observed. Normally, the accumulation of carriers in QDs leads to the appearance of a characteristic step in the C–V curve. When the reverse bias increases, the depletion region of the metal–to–semiconductor interface reaches the QDs layer and becomes fixed there. The energy states of the QDs are raised above the Fermi level, thereby allowing the QDs to release their carriers. Consequently, the capacitance remains constant until all the charge carriers from the QDs layer are removed and the depletion region starts to extend.
As the number of cycles of the InAs/GaAs supply increases, the step begins to appear and becomes very clear in the ten-cycle sample at circa -1.5 V. The width of the step of the C-V curve is related to the carrier concentration confined in the QDs layer, $Q_{QD}$, the wider the step, the higher the confined carrier concentration. The carrier concentration of the quantum dots $Q_{QD}$ is given by\(^{18}\)

$$Q_{QD} = \int g(E)f(E)dE,$$

(1)

where the density of states $g(E)$ and the Fermi-Dirac distribution function $f(E)$ is given by

$$g(E) = \frac{2N_{QD}}{\sqrt{\pi}} \frac{\exp \left( -2 \left( \frac{E-E_{QD}}{\Delta E} \right)^2 \right)}{\Delta E},$$

(2)

$$f(E) = \frac{1}{1+\exp\left( (E-E_F)/kT \right)}.$$  

(3)

Here, $N_{QD}$ is the concentration of QDs, $E_{QD}$ is the position of the electron level in the QDs with respect to the Fermi level ($E_F$) in the InGaAs QW, $\Delta E$ is the energy dispersion of the QDs density of states function, and $k$ is the Boltzmann constant. According to Eqs. (1) and (2), the ten-cycle sample may have a high density of states due to a high concentration of QDs compared with the other samples. The observation of QDs producing a zero-dimensional confinement in the ten-cycle sample is in good agreement with the PL results.

To investigate the C-V characteristics of the ten-cycle sample in detail, C-V measurements were carried out at 77 K. The results are shown in Fig. 5 together with the C-V curve at 300 K for comparison. As the temperature decreases from 300 to 77 K, the step width of the C-V curve increases, representing an increase in the carrier concentration confined in the QDs layer. This fact can be confirmed by a depth profile of the carriers, which is estimated from the depletion approximation.

$$N_{CV}(W) = \frac{C^3}{q\varepsilon_0 \frac{dC}{dV}}, \quad W = \frac{\varepsilon \varepsilon_0}{C}.$$

(4)

where $\varepsilon_0$ and $\varepsilon$ are the dielectric constants of free space and the semiconductor, respectively. $A$ is the area of the Schottky contact, and $W$ is the width of the space charge region, which is equivalent to the depth from the sample surface. In semiconductors with quantum confinement, the C-V concentration $N_{CV}$ corresponds approximately to the free carrier concentration.\(^{19}\) Figure 5(b) shows the carrier concentration as a function of the distance from the sample surface obtained from Eq. (4). The position of QDs determined from the position of $N_{CV}$ maximum is circa 330 nm, which is in good agreement with the sample’s real structure.

As the measurement temperature is lowered, the peak height increases and the FWHM narrows. The peak height and FWHM are $3.9 \times 10^{16}$ cm$^{-3}$ and about 25 nm at 300 K, while they are $8.7 \times 10^{16}$ cm$^{-3}$ and 16 nm at 77 K. The depth from the sample surface ($W$) equals the position expectation value of the carriers, and the width of the depth profile $\Delta W$ is given by the change of the position expectation values of the carriers.\(^{19,20}\) It may be concluded that the narrow FWHM at 77 K is due to a small change in the position expectation values.

**IV. CONCLUSION**

We investigated the electrical and optical properties of InGaAs dots–in–a–well structures grown by an atomic layer epitaxy technique. In the case of the sample produced with five cycles of the InAs/GaAs alternate supply, the InGaAs quantum well coexisted unstably with premature QDs. For the ten-cycle sample, a strong PL peak from the QDs appeared indicating the existence of the well-confined carriers in QDs. The existence of QDs in the ten-cycle sample was confirmed by C-V measurements showing a clear characteristic step in the C-V curve even at 300 K. The evolution mechanism for the formation of InGaAs dots–in–a–well was clarified by characterizing dots–in–a–well with different numbers of cycles of the InAs/GaAs source supply, which can contribute to the fabrication of quantum devices using the ALE technique.

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