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Effect of indium-flush method on the control of photoluminescence energy of highly uniform self-assembled InAs quantum dots by slow molecular beam epitaxy growth

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The control of the emission energy from self-assembled InAs quantum dots has been demonstrated by using indium flush. The low-temperature indium-flush method was found to control the emission energy preserving the high structural uniformity attributed to the slow dot growth. In the standard indium-flush method, where the substrate temperature was raised up from the dot-growth temperature, blueshift larger than the shift by the low-temperature indium flush was observed and was explained reasonably by the enhanced In/Ga-interdiffusion. Also, the effect of AlGaAs capping layer before the indium-flush step was studied. © 2007 American Institute of Physics. [DOI: 10.1063/1.2752598]

I. INTRODUCTION

Semiconductor self-assembled quantum dots (QDs) exhibit a variety of confinement-related optical and electronic properties useful for optoelectronic device applications such as QD lasers and optical amplifiers. In particular, broad efforts are currently underway to develop new techniques for optically controlling spin degrees of freedom in QDs. These efforts are simulated in part by some proposals to use the spin as a quantum bit in quantum computing and qubit.1–5 Some proposals rely on the energy selectivity in the optical excitation processes of spins and, therefore, require the precise control of electronic or excitonic energies. In InAs/GaAs self-assembled QDs,7,8 several techniques to control size and exciton levels have been demonstrated such as indium flush (IF),9–11 atomic force microscopy oxidation,12 rapid thermal annealing,13,14 and closely stacking.15,16

In this article, we have investigated the control of the exciton energies in self-assembled InAs/GaAs QDs with IF method, which is one of the practical solutions for reliable in-situ control of the QD height. The control was already demonstrated by Fafard and co-workers for normal growth rate of InAs dots.9–11 Here, we concentrate on the low growth rate of InAs QDs, which is known to produce uniform QD size, and also compare the low-temperature indium flushing (LT-IF), where the flushing is done at the dot-growth temperature with the standard IF by Fafard et al., where the flushing is done at the elevated temperature. We find that both conditions of the flushing temperature can control the QD height. The standard IF, however, deteriorates the uniformity of emission energy, probably due to the In/Ga-interdiffusion occurring during the flushing. The LT-IF method, on the other hand, can maintain the high structural uniformity formed by low growth rate due to the suppression of the interdiffusion.17,18

II. EXPERIMENT

The InAs QDs were grown on a GaAs (120 nm)/AlAs (10 nm)/GaAs (300 nm) buffer layer on semi-insulating (001) GaAs substrate by a molecular beam epitaxy (RIBER-MBE32P). The growth temperature was 620 °C for the buffer layer and 475 °C for the InAs QDs. The substrate temperature was lowered gradually during the growth of the buffer layers. The InAs QDs were grown at a growth rate of 1.5 × 10−3 ML/s (low rate growth conditions).19 To stabilize the InAs QDs, 90 s postannealing was performed. We used three growth sequences of capping layers as shown schematically in Fig. 1. The material of the capping layer (cap I) was GaAs (type 1 and 2) or Al0.36Ga0.64As (type 3). After cap I, 3 min IF was done in all growth sequence types. Then, two flushing temperatures were used; one is the growth temperature of the InAs QDs (we call this method the LT-IF method) for migrating indium from QD to GaAs surface (type 1) and the other was 590 °C, to which the substrate temperature was raised gradually after cap I for flushing the surface indium (type 2 and 3; this is a standard IF). For each IF condition, the thickness d of the capping layer before IF was varied. The nominal thickness d of cap I is
The IF method was developed originally to improve the shape uniformity in individual columns of the vertically stacked self-assembled QDs by Wasilewski and Fafard. At the flushing step, the substrate temperature was ramped up from the QD growth temperature in order to remove all surface indium. In this work, the substrate temperature at flushing step was kept at 590 °C during the growth of the top GaAs layer (100 nm).

The time-integrated photoluminescence (PL) of the QD ensemble was measured at 10 K. A Nd:YVO₄ laser (532 nm) was used as the excitation source and the PL spectra were detected by an InGaAs charge-coupled device (CCD). The excitation power was several milliwatts, and the laser light was focused onto the sample surface with a diameter of 200 μm.

III. RESULTS AND DISCUSSION

The IF method was developed originally to improve the shape uniformity in individual columns of the vertically stacked self-assembled QDs by Wasilewski and Fafard. At the flushing step, the substrate temperature was ramped up from the QD growth temperature in order to remove all surface indium. In this work, the substrate temperature at flushing step was kept at 590 °C during the growth of the top GaAs layer (100 nm).

![Figure 1](image-url) Schematic illustration of sample structures. The material of cap I, In-flush temperature, and thickness d of cap I are summarized also in Table I.

Table I. Summary of growth sequence.

<table>
<thead>
<tr>
<th>Sequence</th>
<th>Cap I material</th>
<th>Indium-flush temperature T (°C)</th>
<th>Cap I thickness d (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>type 1</td>
<td>GaAs</td>
<td>475</td>
<td>2.8, 4.2, 5.6</td>
</tr>
<tr>
<td>type 2</td>
<td>GaAs</td>
<td>590</td>
<td>1.4, 2.24, 3.36, 5.6</td>
</tr>
<tr>
<td>type 3</td>
<td>Al₀.₃₆Ga₀.₆₄As</td>
<td>590</td>
<td>2.3, 2.52, 3.36, 5.6</td>
</tr>
</tbody>
</table>

Figure 4 shows the time-integrated PL spectra of the type 1 and 2 samples with different cap I thickness d. The bottommost PL spectrum is for the sample without IF. The spectra for both type of samples indicate clearly that the emission energy increases as the cap I thickness decreases. Since all spectra can be decomposed well into multiple Gaussian components assuming the inhomogeneous size distribution of self-assembled QDs, the central energies of the exciton ground-state E₀ and the first excited state E₁ can be determined.

Figure 5 shows the time-integrated PL spectra of the type 1 and 2 samples with different cap I thickness d. The bottommost PL spectrum is for the sample without IF. The spectra for both type of samples indicate clearly that the emission energy increases as the cap I thickness decreases. Since all spectra can be decomposed well into multiple Gaussian components assuming the inhomogeneous size distribution of self-assembled QDs, the central energies of the exciton ground-state E₀ and the first excited state E₁ can be determined.

As seen in the upper panel of the figure, the line narrowing effect of the exciton ground state E₀ is smaller than that reported with standard IF method. The FWHM of LT-IF with 5.6 nm GaAs cap I decreases slightly from 31 meV (as-grown) to 28 meV because as-grown InAs QDs have originally a narrow PL width due to high structural uniformity by the low rate growth. On the other hand, the PL linewidth of standard-IF with same d increases to 48 meV. Therefore, we can conclude that the LT-IF can control the emission energy maintaining the highly structural uniformity achieved at the low growth rate.

Also, the figure shows the following: as the thickness d decreases, E₀ increases for both IF temperatures (475 °C, 590 °C), indicating that the QD height can be controlled by...
LT-IF and standard IF method. As compared with standard IF method (type 2), $E_0$ of type 1 exhibits the parallel redshift by $\sim 140$ meV, and $\Delta E$ of type 1 is larger but reduces similarly as $d$ decreases. For type 1, the $d$-dependence of $E_0$ and $\Delta E$ can be explained reasonably due to the QD height control by the LT-IF. The possible structural difference between type 1 and 2 is the In/Ga-interdiffusion between the InAs dot and the cap I GaAs. Therefore, the observed redshift of $E_0$ and larger $\Delta E$ of type 1 can be attributed to the suppression of the interdiffusion.

In type 3, the emission peak shift occurs at a thinner $d$ around 3 nm than type 2, where the PL peak shift starts from around 5 nm, suggesting the reduction of In transfer from InAs QDs to the surface of AlGaAs cap I because of a large
bonding energy of Al deposited on InAs QDs. Furthermore, $\Delta E$ of type 3 is larger than that of type 2 with the same $d$. It suggests the enhancement of lateral confinement by AlGaAs cap I. For some applications where the surface InAs should be avoided, type 3 can be adopted to compensate the reduction of $\Delta E$ which is observed in type 2.

Finally, in order to confirm the above picture, we performed a simple model calculation for the PL energy. The solid and dashed lines in the upper panel of Fig. 5 are the calculated results of $E_0$. We assumed a square well potential in the growth direction and two-dimensional harmonic potential in the lateral direction. In this model, effects of the interdiffusion between cap I and QD is included in the lateral confinement of QD, which is determined by the $\Delta E$ of Fig. 5. We assumed also that the height of QD was equal to the thickness of cap I. Good agreement between the experiment and the calculation indicates that the height of QD is controlled by both IF methods and the blueshift of $E_0$ in type 2 is caused by the lateral interdiffusion of In and Ga. From the calculation, the radius of type 2 is 1.3 times larger than that of type 1 and the band gap energies of QD at type 1 and 2 are estimated to be 0.895 and 1.072 eV, and the corresponding increase of Ga content $\Delta x$ in type 2 is estimated to be 0.16 without strain consideration and 0.2 with it.

IV. SUMMARY

We demonstrated that the control of photoluminescence energy from self-assembled InAs quantum dots could be achieved by using low-temperature indium-flush method. The low-temperature flush can maintain the high structural uniformity that is due primarily to the slow growth. The high-temperature flush can also control the emission energy but gives rise to reduction of the structural uniformity and lateral confinement due to the enhanced In/Ga interdiffusion. The reduction of the confinement can be compensated by using the AlGaAs cap before indium-flush step.

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