Title	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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Citation	Journal of Applied Physics, 102(1), 013515-1-013515-4 https://doi.org/10.1063/1.2752598
Issue Date	2007
Doc URL	http://hdl.handle.net/2115/49867
Type	article
File Information	GetPDFServlet copy.pdf

AIP Journal of Applied Physics

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Citation: J. Appl. Phys. **102**, 013515 (2007); doi: 10.1063/1.2752598
View online: http://dx.doi.org/10.1063/1.2752598
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JOURNAL OF APPLIED PHYSICS **102**, 013515 (2007)

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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(Received 12 April 2007; accepted 18 May 2007; published online 9 July 2007)

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[1–5] and qubit conversion.[6] 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 dots, 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 stabilitze 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 $Al_{0.36}Ga_{0.64}As$ (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

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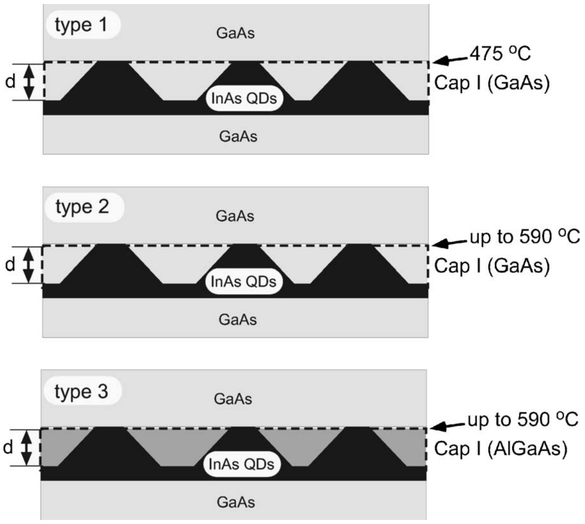

FIG. 1. Schematic illustration of sample structures. The material of cap I, In-flush temperature, and thickness *d* of cap I are summarized also in Table I.

shown in Table I. In type 1, the substrate temperature was gradually increased to 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 $\mathrm{Nd:YVO_4}$ 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.[9,10] 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 QD growth temperature in type 1 (LT-IF method). This is to minimize undesired In/Ga-interdiffusion that could induce alloy fluctuation and reduce the uniformity of QD emission. We expect excess indium to migrate from InAs QDs to the surface of GaAs cap I during this LT-IF step, that is, the growth interruption at 475 °C. Then, thin GaAs was grown for embedding InAs QDs, while the substrate temperature was increased to 590 °C in order to reduce the indium segregation to the surface. From the viewpoint of flushing the remnant indium, the LT-IF method is less effective than the standard IF method.

TABLE I. Summary of growth sequence.

Sequence	Cap I material	Indium-flush temperature T (°C)	Cap I thickness d (nm)
type 1	GaAs	475	2.8, 4.2, 5.6
type 2	GaAs	590	1.4, 2.24, 3.36, 5.6
type 3	$\mathrm{Al_{0.36}Ga_{0.64}As}$	590	2.3, 2.52, 3.36, 5.6

Structural effects of LT-IF were estimated from atomic force microscopy (AFM) images of uncapped samples. AFM imaging was done in tapping mode using a Digital Instruments Nanoscope III AFM and standard silicon nitride tips. Figure 2(a) shows highly uniform as-grown InAs QDs. Figures 2(b) and 2(c) show 2.5 nm GaAs cap I surface after LT-IF and standard-IF, respectively. Surface roughness of LT-IF and standard IF were ±0.19 and ±0.11 nm, indicating that the top of QD was removed by each IF method.

In order to estimate the indium amount of migration from InAs QDs to GaAs cap I surface and segregation to surface in LT-IF, we have also grown a doubly stacked QDs structure as shown in the inset of Fig. 3. As in the case of type 1, the substrate temperature was kept at 475 °C during the growth. We determined the critical layer thicknesses of 2D-3D transition in both QD layers by *in-situ* reflection high-energy electron diffraction (RHEED) pattern, which turned from streaky to spotty. Figure 3 shows the critical thickness Θ_2 for the upper InAs QDs (QD2) normalized by Θ_1 of the lower QDs (QD1), as a function of thickness (d_1) of GaAs barrier. The Θ_2/Θ_1 by the standard IF with d_1 of 5.6 nm (shown by the open circles) was larger than LT-IF, suggesting that more excess indium in LT-IF contributed to QD2 formation than in the standard IF. The reduction of Θ_2 compared to Θ_1 can be attributed to the indium supplied to the QD2 by the segregation. As seen in Fig. 3, Θ_2/Θ_1 of LT-IF has a clear linear dependence on d_1, which implies uniform indium incorporation in the GaAs barrier.

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 E_0 and the first excited state E_1 and the full width at half maximum (FWHM) of their PL components can be estimated. The ground-state energy E_0 for all samples is plotted as a function of the nominal cap I thickness *d* in the upper panel of Fig. 5. The error bar indicates the FWHM of ground-state emission. Furthermore, the intersublevel spacing $\Delta E(=E_1-E_0)$, which is determined basically by the in-plane confinement of QDs, is shown in the lower panel of Fig. 5.

As seen in the upper panel of the figure, the line narrowing effect of the exciton ground state E_0 is smaller than that reported with standard IF method.[9,10] 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, E_0 increases for both IF temperatures (475 °C, 590 °C), indicating that the QD height can be controlled by

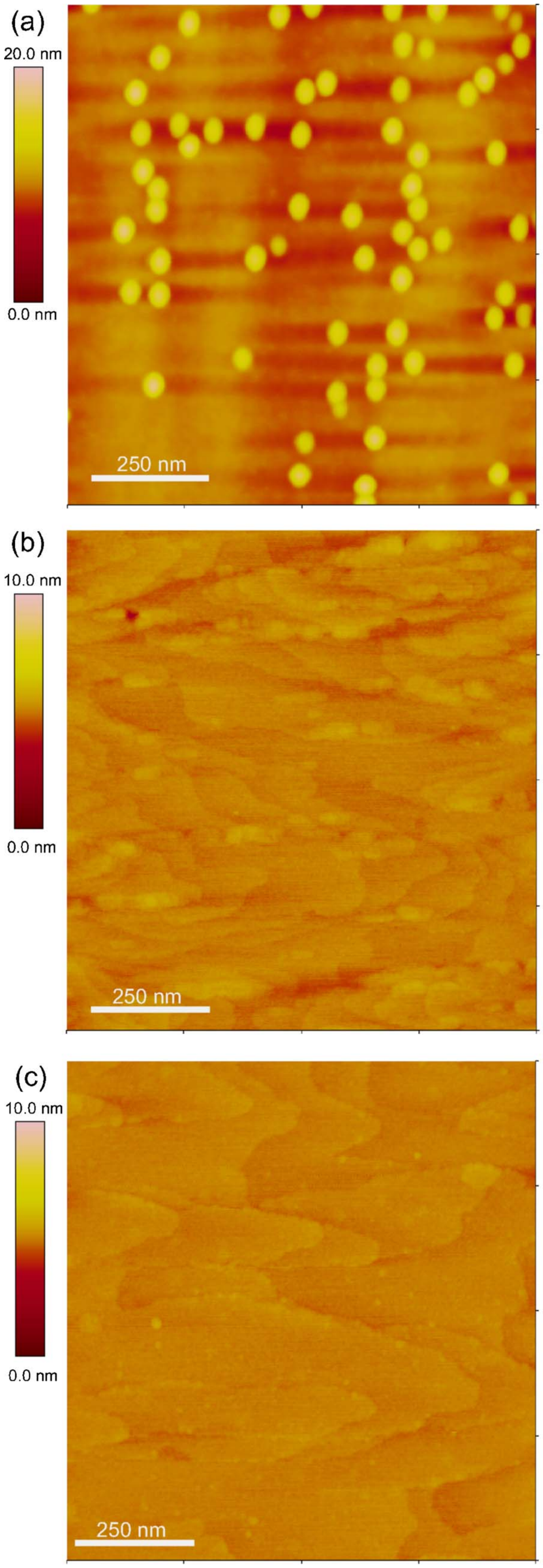

FIG. 2. (Color online) (a) 1×1 μm^2 AFM image of highly uniform InAs QDs with the density of 7×10^9 cm^{-2} without IF. (b) GaAs cap I surface (d=2.5 nm) with LT-IF, and (c) GaAs cap I surface (d=2.5 nm) with standard IF.

LT-IF and standard IF method. As compared with standard IF method (type 2), E_0 of type 1 exhibits the parallel redshift by ~140 meV, and ΔE of type 1 is larger but reduces similarly as d decreases. For type 1, the d-dependence of E_0 and ΔE 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 ΔE of type 1 can be attributed to the suppression of the interdiffusion.

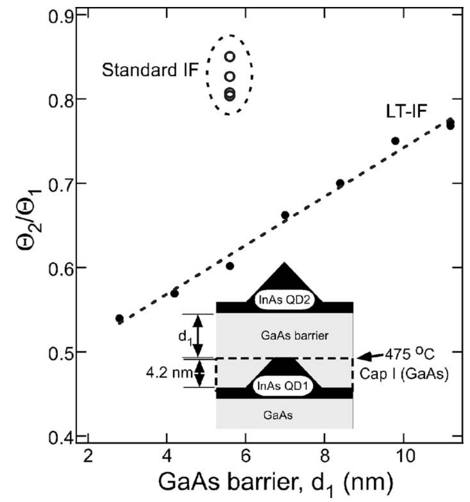

FIG. 3. Θ_2/Θ_1 of the sample with d=4.2 nm is plotted as a function of the GaAs barrier d_1 for LT-IF. The solid circles and open circles indicate the values with LT-IF and standard IF. The inset is a schematic illustration of the doubly stacked InAs QDs structure.

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

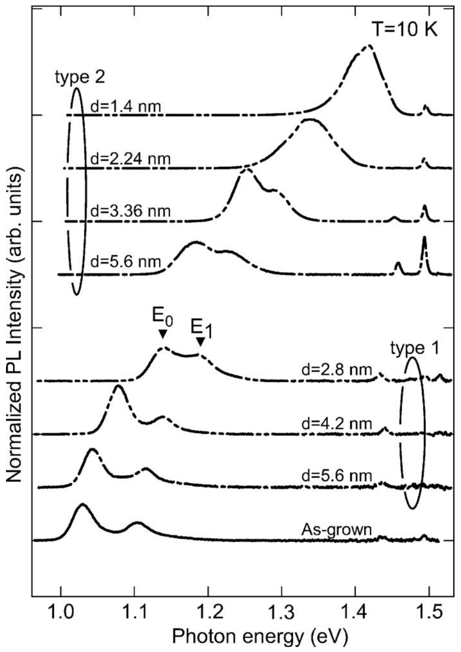

FIG. 4. Nonresonantly excited PL spectra of type 1 and 2 samples with various cap I thickness at 10 K. The bottommost spectrum is for InAs QDs without IF. All spectra are vertically displaced for clarity. E_0 and E_1 indicate the energies of the exciton ground-state and the first-excited state transitions, respectively.

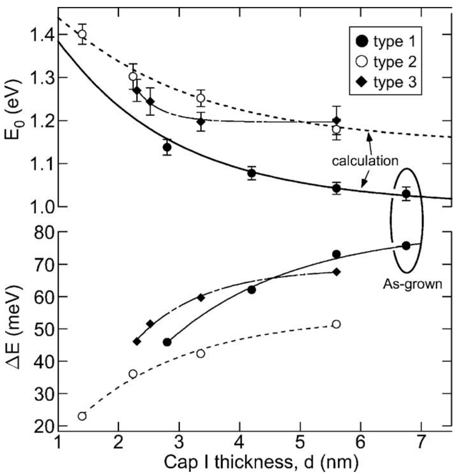

FIG. 5. The exciton ground-state energy E_0 (upper panel) and the intersublevel spacing ΔE (lower panel) as a function of the cap I thickness. The cap I thickness of as-grown sample indicates average QDs height ~6.75 nm, which is estimated by the AFM image of a reference sample in Fig. 2(a). In the upper panel, the error bar indicates the FWHM of the exciton ground-state component for each sample. Note that cap I is a capping layer before IF step. The solid circles, open circles, and solid diamonds are for type 1, type 2, and type 3 samples, respectively.

bonding energy of Al deposited on InAs QDs. Furthermore, ΔE 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 ΔE 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 ΔE 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 Δx 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.

ACKNOWLEDGMENTS

This work was financially supported by a Grant-in-Aid for Scientific Research of Japanese Ministry of Education, Culture, Sports, Science, and Technology. One of the authors (H.S.) thanks Y. Nakata of Fujitsu Laboratories Ltd. for useful discussion.

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