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Low-Temperature Scanning Tunneling Microscopy of Self-Assembled InAs Quantum Dots Grown by Droplet Epitaxy

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Abstract

Using molecular beam epitaxy, we study self-assembled InAs quantum dots (QDs) grown on GaAs (001) substrates by low-temperature scanning tunneling microscopy. We compare two different growth processes, the well-known Stranski-Krastanow mode and the more recently studied droplet epitaxy mode. We show that, for the same amount of deposited indium, the shape of the dots shows similar anisotropic tendency, but the InAs coverage dependence of the density shows that the growth mechanisms are different at the initial stage of the QD formation. By scanning tunneling spectroscopy at low temperature, we succeed in mapping the local density of states (LDOS) of a single quantum dot grown by the droplet epitaxy. Maps of LDOS are consistent with those previously reported for the Stranski-Krastanow mode; however, energy spectrum of our QDs shows different peak structures.

Keywords: Quantum Dots ; Droplet Epitaxy ; Scanning Tunneling Microscopy ; Molecular Beam Epitaxy ; Self-assembled Structure ; Local Density of States ; Strained-growth

1. Introduction

Quantum Dots (QDs) are very important for the development of new kinds of devices like low-threshold lasers, single electron transistors and single photon sources. When QDs are used as one of active components, they allow nanoscale systems new functionalities due to their zero-dimensional structure. Self-assembling of nanostructures is one of key technologies for future device fabrication. There are two well-known ways to grow self-assembled QDs by molecular beam epitaxy (MBE). One is by a simultaneous supply of indium and arsenic sources for Stranski-Krastanow (S-K) growth mode [1]. The other is by an individual supply of sources for droplet epitaxy (DE) mode, and In source supply is sequentially followed by As_4 source supply [2]. Optical spectroscopy techniques enable to investigate the electronic properties of this kind of nanostructures [3]. However, in order to map out the local density

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of states (LDOS), corresponding to $|\psi_i(E_i, x, y)|^2$, a nanoscale spectroscopic technique is required. Here, $\psi_i(E_i, x, y)$ is the wave function at the energy E_i and the location (x, y). There are a few reports on the characterization of single S-K QDs [4 - 6] by low-temperature scanning tunneling microscope (LT-STM). However, there is no report on DE QDs, although they show advantages on the control of optical properties by their shapes [7], which is hardly possible by S-K QDs. Moreover, this method enables us to grow self-assembled QDs with lattice matched materials [8]. This is contrary to S-K growth which is based on strain arising when a crystal forms an adlayer on a substrate with a different lattice constant. Thus, local characterization of individual single DE QD is required to study quantitative relationships between the electronic properties and the topographic features. In this paper, we characterize DE QDs and S-K ones by LT-STM in order to compare the two growth modes to show difference in topographic features and to highlight the fact that DE growth offers an alternative feasibility for the creation of this kind of nanostructures.

2. Experiments

A Si-doped (5×10^{17} cm^{-3}) GaAs (001) substrate is firstly heated at 580 °C to remove the oxide layer under the As_4 pressure of 1.3×10^{-6} Torr. On this surface, we grew a buffer layer of 200 nm thick Si-doped ($n = 1.3 \times 10^{18}$ or 2.4×10^{18} cm^{-3}) GaAs at 0.5 ML/s by MBE, under the same As_4 flux at the substrate temperature of 550 °C. Next, we grew 5 ML of undoped GaAs as the confinement barrier of QDs. Then, we decreased the substrate temperature to 450 °C, and grew InAs QDs. We used the same In and As_4 fluxes from the Knudsen-cells for both S-K and DE growth modes. The In flux corresponds to the conventional InAs MBE growth rate of 0.05 ML/s. The As_4 flux corresponds to the growth rate of 0.35 ML/s governed by the As_4 incorporation kinetics at the beam equivalent pressure 1.3×10^{-6} Torr. In the S-K mode, we grew InAs by simultaneous In and As supply, corresponding to a V/III ratio of 7 with the growth rate of 0.05 ML/s. On the other hand, the DE QDs were fabricated in two-steps: (i) deposition of the same amount of In in the absence of As_4 flux, followed by (ii) the exposure to As_4 flux for 60 sec. In this case the growth rate of the InAs QDs is determined by the As_4 incorporation rate, 0.35 ML/s. For the InAs coverage dependence, we grew 1.7, 1.9, and 2.1 ML InAs by S-K and DE modes. After the formation of QDs, the substrate temperature was quickly decreased to transfer the sample through ultra-high vacuum (UHV) to the directly connected LT-STM stage at 5K, without surface contamination.

The LT-STM measurements were performed at 5 K in UHV with the pressure in the order of 10^{-11} Torr. We use the topographic images to compare shape, size and density between S-K and DE QDs. For each growth mode, the density and size of QDs were measured from several images containing more than 50 of them. We also investigate the electronic properties of the DE QDs by low-temperature scanning tunneling spectroscopy (LT-STS). According to the theory of STM [9], the LDOS is proportional to the differential conductance (dI/dV) and, experimentally $dI/dV\ (V, x, y) \propto \mathrm{LDOS}\ (eV, x, y) = \sum_{\delta E} |\psi_i(E_i, x, y)|^2$, with δE being the energy resolution, determined by the amplitude of bias modulation at the low temperature. To record the $dI/dV\ (V, x, y)$ signal, we used a lock-in technique with a modulation signal of $V_{mod} = 10\ \mathrm{mV_{p\text{-}p}}$ at 700 Hz.

3. Results and discussion

3.1. Topographic features

It is found that, for all of the samples regardless of InAs coverages or growth modes, the QD density is higher along the steps than on the terraces. This is consistent with previous reports concerning S-K [10] and DE [11] modes. This is because the migration of In adatoms is obstructed by steps. This locally induces longer residence time along step edges and increases the probability for In atoms to form InAs QDs in the presence of As_4 molecules. On the other hand, some important differences are observed. We show the coverage dependence of InAs QD density in Fig. 1. In S-K mode, QDs are already formed at the InAs coverage of 1.7 ML, though in the case of DE mode, no QD is found at the same In coverage. The measured relation between the density of S-K QDs and the InAs coverage seems to be consistent with the one found by Leonard et al. [10], although the substrate temperature and the InAs growth

rate used here are different. The dependence of the QD density ρ_{QD} for S-K mode accords with a fitting by the form $\rho_{QD} = \rho_0(\theta - \theta_0)^{\alpha}$ [10] as shown in the (green) dashed line in Fig. 1. Here, θ_0 is the critical layer thickness (1.5 ML) and θ is the InAs coverage. The normalization density ρ_0 and the exponent α of 2×10^{11} cm^{-2} and 1.76 were taken From Ref. [10]. As it is shown in Fig. 1, the DE mode grown QD density shows much steeper evolution than that of the S-K growth. For 1.9 ML, it already reaches 93 % of the density at the InAs coverage 2.1 ML by the DE mode. However, it remains at 55 % of the density at the coverage 2.1 ML by the S-K mode. These results lead us to conclude that the InAs wetting layer in the case of a growth by DE mode is thicker than that of S-K mode at the initial QD formation stage, and the formation mechanism of DE QDs is different from S-K ones.

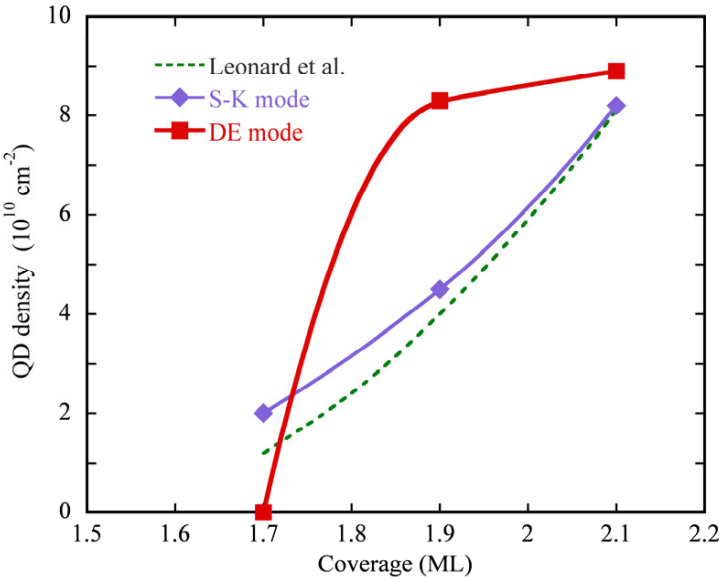


Fig. 1. Coverage dependence of QD densities by S-K and DE modes. The dashed line corresponds to the fitted curve by Leonard et al. in ref. 10. Each solid line is a guide for eyes.

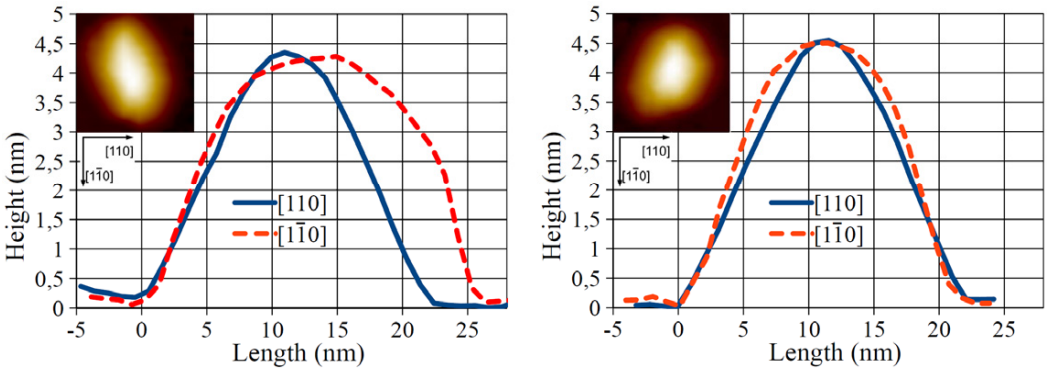
Figure 2 shows topographic profiles of InAs QDs grown with 2.1 ML deposition by (a) the S-K mode and (b) the DE mode. At this InAs coverage, QD density of S-K mode becomes comparable to that of DE mode. The measured density in the DE mode is 8.9×10^{10} cm^{-2} and that in the S-K mode is 8.2×10^{10} cm^{-2}. As summarized in Table 1, the average dimensions of the QDs for the S–K growth are higher than in the DE growth by 13% for the height, 13% in length along the [110] direction, and 19% in length along the [1$\bar{1}$0] direction. The averaged length ratio ([1$\bar{1}$0] / [110]) of QDs was 1.09 for the DE mode and 1.14 for the S-K mode. DE-grown QDs show more isotropic shape than that by S-K mode as shown in Fig. 2. This shape anisotropy difference is mainly caused by the growth mechanism difference. At the DE mode growth, In atoms first form droplets, which are liquid and have isotropic shape by the surface tension. Such droplets incorporate As_4 molecules to be grown as InAs QDs. Thus, they tend to keep initial isotropic shape. However, at the S-K mode growth, In atoms migrate under the As_4 flux to form InAs QDs on the surface. This means the migration anisotropy of In atoms determines the shape of S-K QDs. On the (2×4) reconstructed (001) surface, it is known that the migration is more frequent along [1$\bar{1}$0] direction than [110] direction by the trench shape of missing arsenic dimer rows along [1$\bar{1}$0] direction. This results in the faster growth of InAs QDs along [1$\bar{1}$0] direction. Therefore, the QD shape is more anisotropic at S-K mode growth than DE mode one. It is found that the shape anisotropy is more pronounced for smaller DE QDs. The standard deviation for each dimension is very close between two modes. It means that, if we normalize this standard deviation by the average values, DE QDs present relatively wider range of size fluctuation than S-K QDs.

Table 1: Dimensions and geometrical properties of QDs grown by S–K and DE modes at comparable densities. (InAs coverage 2.1 ML)

	average value (av)		standard deviation (sd)		sd/av (%)	
	S–K	DE	S–K	DE	S-K	DE
[001] height (nm)	3.5	3.1	0.7	0.7	19.9	21.6
[1$\bar{1}$0] length (nm)	21.7	18.3	2.4	2.4	10.9	12.9
[110] length (nm)	19.1	16.9	2.7	2.9	14.3	17.2
ratio ([1$\bar{1}$0] / [110])	1.14	1.09	0.08	0.07	7.1	6.8

3.2. Spectroscopic LDOS imaging

By using LT-STS, we succeeded in mapping LDOS of the first three energy levels of a single QD grown by DE-mode (Fig. 3). The *dI/dV* (*V, x, y*) curve spatially averaged over the QD area [Fig. 3(a)] revealed three distinct levels at sample bias voltages of V = 1.12, 1.21, and 1.57 V. Higher bias voltage corresponds to higher electron energy. The QD size in Fig. 3 is smaller than the QDs studied in refs. 5 and 6, and the spectrum shows peaks at higher voltages. The *dI/dV* (*V, x, y*) maps of the first three levels were also clearly imaged. In these maps, brighter region corresponds to higher LDOS region. They were found consistent with the previous report on S-K QDs as for the bias voltage dependence of the LDOS distribution [5]. On the energetic scale, the first state in DE QD presents no node and peak appears at the center of the dot [Fig. 3(c)]. This LDOS appearance suggests that the first state is the zero-dimensional ground state in the DE QD. The LDOS of the second one is distributed in two peaks along the long length and has one node in the center [Fig. 3(d)]. The third one presents the same characteristics but with the peaks along the short length [Fig. 3(e)]. According to Maltezopoulos et al. [5], the asymmetric shape of the QD lifts energy level degeneracy. However, spectrum for DE QD shows difference from that for S-K QD [5] in the peak structure. First lowest two states are very close in Fig. 3.(a), instead of larger splitting in ref. 5, although the causes are not yet well understood. The full width at half maximum (FWHM) of the peaks of the DE QD is about 0.1 eV, it is comparable to the FWHM of the spectrum previously reported [5]. It is revealed that the DE QDs show equivalent zero-dimensional characteristics to S-K ones. More detailed studies are necessary to make the origin of spectroscopic difference clear.



(a). S-K mode QD profile (b). DE mode QD profile

Fig. 2. Topography of InAs QDs at InAs coverage 2.1 ML grown by (a) S-K and (b) DE modes. (Each inset image area size is 22 nm × 22 nm)

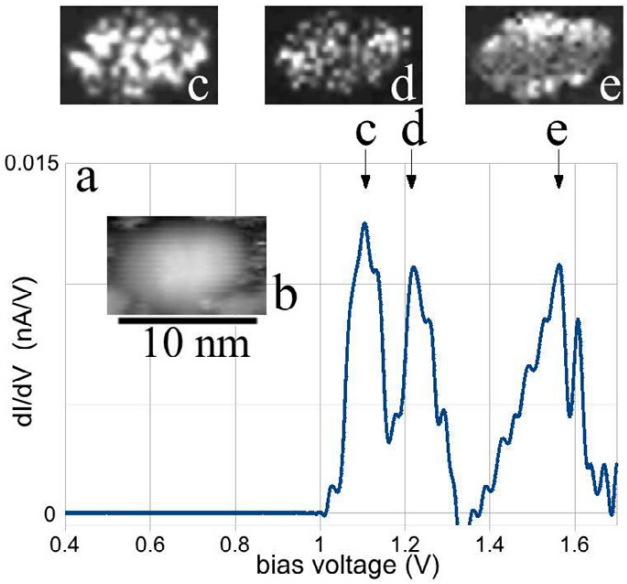


Fig. 3. (a) dI/dV signal curve averaged over the single QD grown by DE-mode. (b) topographic image of the QD. The *dI/dV* images at bias voltages (c) 1.12 V, (d) 1.21 V, and (e) 1.57 V.

4. Conclusion

In summary, we have studied the topographic and electronic properties of InAs QDs grown by DE mode on the GaAs (001) substrates to compare with those by S-K mode using scanning tunneling microscopy and spectroscopy at low temperature. Compared to S-K mode, DE mode seems to create a thicker wetting layer before QD formation and the QD density increases abruptly between 1.7 and 1.9 ML of InAs deposited, which is different from S-K mode. The QD shape analysis indicates that DE mode QDs show slightly larger size fluctuation, though their shape shows more isotropic tendency than those of S-K mode QDs. The mapping of LDOS for DE mode QDs is successfully achieved. LDOS maps are found equivalent between S-K QDs and DE ones except for the detailed spectrum structure.

Acknowledgements

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