Electronic states of self-organized InGaAs quantum dots on GaAs (3 1 1)B studied by conductive scanning probe microscope

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Abstract

We have used conductive scanning probe microscope (SPM) in high vacuum and operated at 173 K in order to investigate the electronic properties of self-organized InGaAs quantum dots (QDs) grown on GaAs (3 1 1)B and (0 0 1) substrates. Ordered InGaAs quantum dot arrays on GaAs (3 1 1)B surface were fabricated by atomic-H assisted molecular beam epitaxy (H-MBE), and Si SPM tips coated with Au which warrants electrical conductivity were used to measure simultaneously both the topographic and current images of QDs surface. From the current–voltage (I–V) curves, unique and different plateau features were observed for QDs formed on GaAs (3 1 1)B and (0 0 1) substrates. The results suggested that a high degree of symmetry of InGaAs QDs on (3 1 1)B was responsible for the observed degeneracy of electronic states and artificial atom-like states. We demonstrate that this conductive SPM technique becomes a powerful tool in studies of single electron charging of individual dots.

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1. Introduction

Recently, the studies on low-dimensional quantum dot (QD) structures have attracted intense research for applications to QD devices such as single-electron transistors [1], far-infrared detectors [2], and QD lasers [3]. Further, resonant tunneling QD diodes are regarded as excellent candidate for digital circuit applications because of their superior switching speed and low-power consumption. On the other,

unique electronic states of QDs analogous to the real atoms have been investigated by Tarucha et al. [4] who studied resonant tunneling characteristics of a GaAs-based double barrier QD diode fabricated by lithography and etching technique. And for this reason, QDs are referred to as artificial atoms. However, the electronic properties of self-organized QDs fabricated by strain-induced heteroepitaxy have not been fully understood and thus have been the target of research recently at various institutions.

While the common characterization techniques such as photoluminescence (PL) and capacitance spectroscopy are particularly useful in probing the local properties containing an ensemble of dots, scanning

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probe microscope (SPM) techniques would be advantageous, if the properties of individual dots are to be exploited with nm-scale size resolution. The use of conductive SPM tip allows us to modify the local band profile of a given QD structure with external applied bias. Furthermore, the electronic properties such as artificial atom-like energy states and shell filling of dots with single electron can be investigated using this technique [5]. We have previously reported that the electronic properties of QDs surface can be studied by conductive SPM with metal-coated Si tips [6]. In this work, we focused our attention on the transport properties such as single electron charging and related Coulomb effects in self-organized InGaAs QDs grown on GaAs substrates, which are potential candidates for the future components in optoelectronics and quantum information processing technology.

2. Experiments

Three stacked layers of self-organized $In_{0.4}Ga_{0.6}As$ quantum dots used in this study were grown on n-type GaAs (3 1 1)B and (0 0 1) substrates (doped to 1×10^{18} cm^{-3}) by atomic-H-assisted molecular beam epitaxy (H-MBE). After standard degreasing and etching steps, each GaAs substrate was subjected to cleaning in UHV by using atomic H irradiation at 580°C for 10 min in order to obtain an atomically flat surface. Then, a 300-nm-thick GaAs buffer layer (Si-doped, 2.5×10^{18} cm^{-3}) was grown at 580°C followed by a 50-nm-thick undoped GaAs spacer layer. After lowering the substrate temperature to 500°C, 8.8 ML of $In_{0.4}Ga_{0.6}As$ QDs layer was grown at a growth rate of ∼0.1 ML/s. The hydrogen backpressure was kept constant at ∼6×10^{-6} Torr during substrate cleaning and H-MBE growth.

To be particularly noted in the $In_{0.4}Ga_{0.6}As$ QDs grown on GaAs (3 1 1)B substrates that differs remarkably from those assembled on (0 0 1) surface is their unique self-organization mechanism [7]. The quantum dot growth on GaAs (3 1 1)B is known to be fundamentally different from the well-established Stranski–Krastanov (S–K) growth mode, and in fact, a complex phase separation and strain-relief mechanism are responsible for the formation of such high-density and well-ordered dot arrays on (3 1 1)B surface. Further, we can control the dot size and density by simply

controlling the deposition temperature while maintaining the structural ordering of QDs arrays [7].

After growth, samples were transferred from MBE chamber to our SPM system in dry nitrogen ambient in order to minimize natural oxidation, and pumped down by using a turbo-molecular pump to a base pressure of $< 5 \times 10^{-7}$ Torr [6]. Si tips coated with Au, which warrants electrical conductivity were used to measure both the topographic and current images of QDs surface. The current–voltage (I–V) characteristics of dots of varying size and of any other arbitrary positions on the dots surface were also studied by using the same conductive tip. Though the tip radius of an Au-coated tip was certainly larger than that of an uncoated tip, this did not affect the lateral resolution required for the topographic and current image measurements done in this work. Further, removal of Au from the tip during scanning was not observed under our operating conditions.

Each QDs sample was thermally treated in SPM chamber at 70°C for 1 h before proceeding to SPM characterization in order to remove residual surface water film [6]. The surface water film is known to result in a strong adhesive force as well as defocusing of applied electric field between the tip and sample [8]. In order to prevent the tip from crashing into the sample during heating due to thermal expansion, the tip was placed close to, but not in a direct contact to the heated sample. During the SPM image and I–V curve acquisition, a constant contact force was applied via an electronics feedback control, and the measurements were conducted in contact mode in vacuum and using a liquid nitrogen cooled stage at 173 K.

In this work, we focused our attention to clarifying the transport properties such as single electron charging and related Coulomb effects in self-organized InGaAs QDs grown on GaAs substrates by using simple and reliable method of conductive SPM technique.

3. Results and discussion

Fig. 1 shows (a) schematic sample structure, and (b) calculated ideal conduction band diagrams under no bias and a finite forward bias V, respectively. The samples were configured to form a double tunnel barrier junction structure. The bottom-most dot layer was intended to improve the quality of the top two QDs
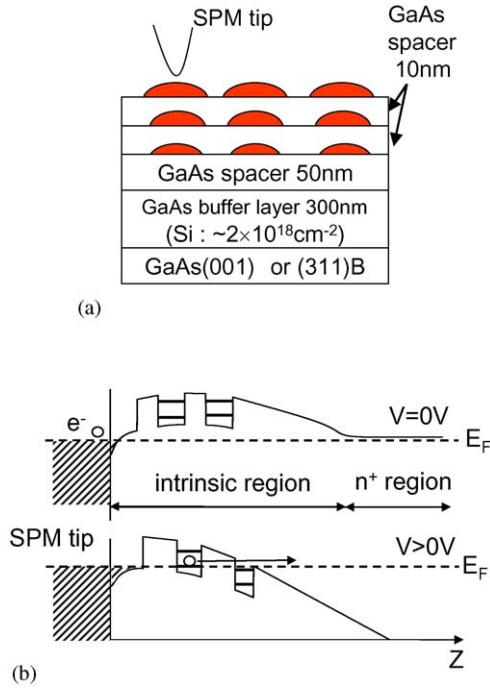
(a)

(b)

Fig. 1. (a) Schematic sample structure, and (b) calculated ideal conduction band diagrams under no bias ($V = 0$) and a finite forward bias ($V > 0$), respectively.

Fig. 2. AFM images of topmost $In_{0.4}Ga_{0.6}As$ quantum dots surface of 3-stack sample on (a) GaAs (3 1 1)B and (b) (0 0 1) substrate, respectively. Scan size is 1 μm × 1 μm.

Fig. 3. $I–V$ curves measured at 173 K on a single $In_{0.4}Ga_{0.6}As$ QD on (a) GaAs (3 1 1)B, (b) (0 0 1) and (c) wetting layer, respectively.

layers, and to align them vertically during stacked growth. No dislocations were observed in transmission electron microscope (TEM) measurements. As seen in Fig. 1(b), the ground state of electrons in the middle QD would lie above the Fermi level of tip under no bias condition, which is one of the necessary conditions in order to observe Coulomb related effects in transport studies. Moreover, it is known that an electron accumulation occurs on the surface of InAs due to the positively charged surface states in InAs, and this effect consequently leads to a lowering of Schottky barrier height between InAs and tip [5]. Therefore, the topmost dots were used in effect as zero-dimensional nano-emitters to inject electrons to single QDs that were buried directly below the topmost dots [9].

Fig. 2 show the atomic force microscope (AFM) images of topmost $In_{0.4}Ga_{0.6}As$ QDs surface of 3-stack QDs sample on (a) GaAs (3 1 1)B and (b) (0 0 1) substrate, respectively. The average QD diameter, height, and arial density were (a) 49.45 nm, 8.82 nm and 1.3×10^{10} cm^{-2} on (3 1 1)B and (b) 54.82 nm, 9.23 nm

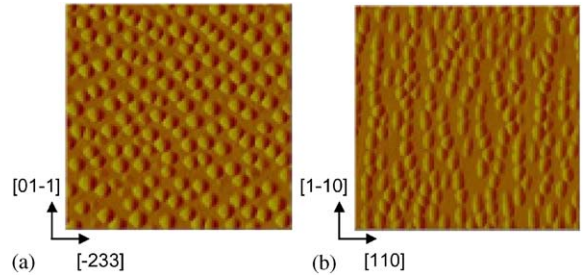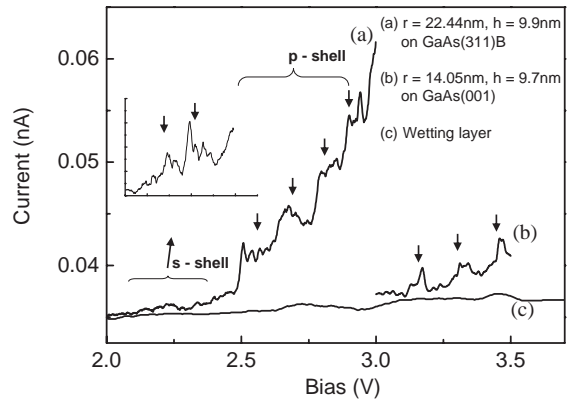and 1.69×10^{10} cm^{-2} on (0 0 1), respectively, and thus were nearly identical. However, it can be seen that an ordered QDs array structure was maintained on (3 1 1)B substrate after strain accumulation during stacked growth. Further, each QD was circular-disk shaped with a higher degree of structural in-plane symmetry. On the other hand, wire-like islands elongated along [1–10] were observed for QDs grown on GaAs (0 0 1) substrate after stacking of 3 dot layers.

Next, $I–V$ curves were taken by placing the tip on top of various isolated QDs. Fig. 3(a) shows typical $I–V$ curve measured on a single QD formed on GaAs (3 1 1)B. The QD size was 22.44 nm in radius and 9.9 nm in height, respectively. Similarly, Fig. 3(b) shows a curve obtained for a single QD formed on (0 0 1) with 14.05 nm in radius and 9.7 nm

in height, respectively. As the QDs on (0 0 1) were ellipse-shaped as shown in Fig. 2(b), we approximated them to circular disks in discussing the QD size in the later section. Staircase-like characteristics were clearly observed in Figs. 3(a) and (b), while the $I–V$ curve (c) that was measured on a wetting layer region showed no apparent feature.

Staircase characteristics generally correspond to Coulomb blockade and related tunneling phenomena, i.e. charging of single electrons into a QD. As shown in Fig. 3(a), two closely spaced plateaus were first observed immediately after onset of current, followed by a larger gap and four closely spaced and equidistant plateaus. This characteristic feature was in good agreement with the recent reports of single electron charging of a QD as an artificial atom [4,10]. Thus, we attribute the first doublet of plateaus to tunneling through the ground state (s-shell), and the quartet to tunneling through the first excited state (p-shell) of QD. The gap between the two should then correspond to energy spacing between the ground and first excited state in a dot due to quantum confinement. The mean width of plateau ΔV was estimated to be 138.6 mV from Fig. 3(a). Thus, single electron charging energy ΔE_{ex} can be estimated as 8.64 meV by taking the bias-to-energy conversion factor of η, which was given as fractional amount of bias that changes the energy of QD to be $\sim$6.23 meV/V into account. Meanwhile, calculated charging energy ΔE_{cal} was 7.34 meV by approximating the QD as a circular metal disk of capacitance given by $C = 8\varepsilon R$, where R is the QD radius. Therefore the approximation showed a good agreement with our measured result.

On the other hand, $I–V$ curve shown in Fig. 3(b) exhibited only three equidistant plateaus and was remarkably different from Fig. 3(a). We think that these plateaus originated from single electron charging of QD, however, degeneracy of energy states or shell structure of artificial atom was not formed in this sample. Following a similar approximation as above, the mean width of plateau ΔV was estimated to be 164.51 mV from Fig. 3(b). This would give $\Delta E_{ex} \sim$9.81 meV, which was again in fair agreement with calculation of $\Delta E_{cal} \sim$11.72 meV, taking the bias-to-energy conversion factor of $\eta \sim$4.50 meV/V into account in this case. We attribute the difference between Fig. 3(a) and (b) to clear difference of dot shape. It is known that a high degree of symmetry
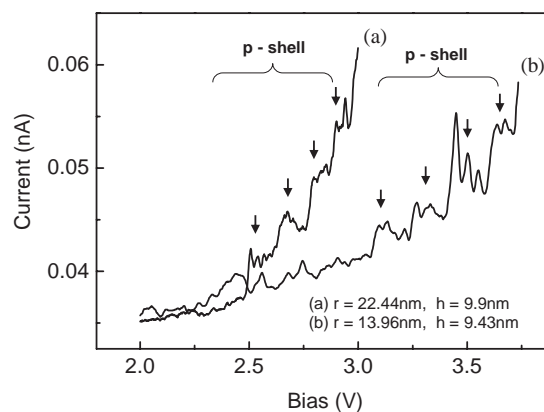

Fig. 4. $I–V$ curves measured at 173 K with the tip placed on single QDs self-organized on GaAs (3 1 1)B substrate for two different QD sizes (a) 22.44 nm in radius and 9.9 nm in height and (b) 13.96 nm in radius and 9.43 nm in height, respectively.

of dot is in fact responsible for the degeneracy of quantized energy level. This is because the symmetry of two-dimensional harmonic potential requires a cylindrical symmetry with a soft boundary profile [4].

We have then investigated the dependence of $I–V$ characteristics on dot size. Fig. 4 represent the $I–V$ curves measured on QDs self-organized on GaAs (3 1 1)B substrate for two different dot sizes (a) 22.44 nm in radius and 9.9 nm in height and (b) 13.96 nm in radius and 9.43 nm in height, respectively. First, it can be observed that a larger dot is more conductive than a smaller dot. This is due to the difference in local surface potentials, which would be larger for small dots [5,11]. Second, four plateaus (p-shell) were clearly seen in both $I–V$ curves but the separation was larger for a smaller dot than a larger dot. This is due to the size effect since smaller dot would have a larger charging energy.

Finally, Fig. 5 plots the measured single electron charging energies for several $In_{0.4}Ga_{0.6}As$ QDs formed on GaAs (3 1 1)B and (0 0 1). The solid line represents the calculated charging energies by approximating QDs as circular metal disks of capacitance given by $C = 8\varepsilon R$. It can be seen that the experimental results obtained in this work were in fair agreement with this simple calculation. Major origins for deviation included: (1) approximating the shape of more of an elliptical dot formed on (0 0 1) to a circular disk, and (2) approximating the size of middle QD which was
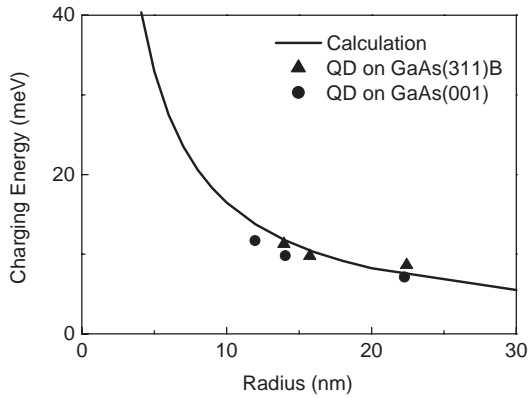
Fig. 5. Measured single electron charging energies for several In$_{0.4}$Ga$_{0.6}$As QDs formed on GaAs (3 1 1)B and (0 0 1). The solid line shows the calculated charging energies by approximating QDs as circular metal disks.

buried below a topmost QD the same as the topmost QD size. From TEM studies, upper QDs were larger in radius than dots in lower layers due to strain accumulation with stacking of dot layers. Nonetheless, we have successfully demonstrated that conductive SPM technique becomes a powerful tool in studies of single electron charging of individual quantum dots.

4. Summary

We have used conductive SPM in high vacuum and operated at 173 K in order to investigate the electronic properties of self-organized In$_{0.4}$Ga$_{0.6}$As QDs grown on GaAs (3 1 1)B and (0 0 1) substrates. The 3-stacked layers of self-organized InGaAs QDs were grown by H-MBE. The bottom-most QDs layer was intended to improve the quality of the top two dot layers, and to align them vertically during stacked growth. On the other, the topmost QD was used as a zero-dimensional nano-emitter to inject electrons to one QD that was buried directly below this topmost dot. This configuration is advantageous in studies of single electron charging of individual QDs.

From the I–V curves, unique plateau features were observed for QDs formed on GaAs (3 1 1)B and (0 0 1) substrates. The results suggested that a high degree of symmetry of InGaAs QDs formed on GaAs (3 1 1)B is responsible for the observed degeneracy of electronic states and artificial atom-like shell structure. The QDs formed on (0 0 1) did not show such degeneracy. These considerations were in good agreement between measured and simple calculation model of charging energies of single electrons in to a quantum dot.

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