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Self-organized InGaAs quantum dots grown on GaAs (3 1 1)B substrate studied by conductive atomic force microscope technique

Yoshitaka Okada[a,*], Masashi Miyagi[a], Kouichi Akahane[a], Mitsuo Kawabe[a], Hidemi Shigekawa[b]

[a] *Institute of Applied Physics, University of Tsukuba, 1–1–1 Tennodai, Tsukuba 305-8573, Japan*
[b] *CREST, Japan Science and Technology Corporation (JST), Institute of Applied Physics, University of Tsukuba, 1–1–1 Tennodai, Tsukuba 305-8573, Japan*

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Abstract

We have used conductive atomic force microscope (AFM) in a high vacuum in order to investigate the electronic properties of self-organized $In_xGa_{1-x}As$ quantum dots (QDs) on GaAs (3 1 1)B substrates. The QDs were fabricated by atomic H-assisted molecular beam epitaxy, and Si AFM tips coated with Au, which warrants electrical conductivity were used to measure both the topographic and current images of QDs surface simultaneously. The conductive AFM measurements were performed in vacuum at room temperature and at lowered temperatures. With this technique, the current–voltage (I–V) characteristics of QDs of varying sizes, and of any other arbitrary positions on the QDs surface can also be studied by using the same conductive AFM tip. It was found that the center of a QD is more conductive than its periphery, and the surface in between the QDs is highly resistive. The differences in the conductance are thought to be due to the local modification of surface bending associated with the surface states. Further, we have shown that the conductance becomes spatially uniform at all points over the packed and ordered QDs at low temperatures, which could be explained by lateral coupling of these strained QDs.

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1. Introduction

Recently, the studies of the electronic and optical properties of *self-assembled* semiconductor quantum dots (QDs) have gained increasing attention [1–5]. While the common characterization techniques such as photoluminescence (PL)

*Corresponding author. Tel.: +81-298-53-5293; fax: +81-298-53-5205.
E-mail address: okada@ims.tsukuba.ac.jp (Y. Okada).

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and capacitance spectroscopy are particularly useful in probing the local properties containing an ensemble of QDs, the scanning probe microscope (SPM) technique becomes more advantageous, if the properties of individual QDs are to be exploited with nm-scale size resolution [6–8].

Meanwhile, we have previously reported that $In_xGa_{1-x}As$ QDs *self-organized* on high-index GaAs (3 1 1)B substrates exhibit a unique structural and optical characteristics compared to the more commonly studied InAs QDs grown on (0 0 1) substrates [9,10]. The QDs growth mode on GaAs (3 1 1)B surface has been shown 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 QDs arrays on (3 1 1)B surface. However, the electronic properties of individual QDs as well as of QDs surface itself have not been fully understood at present. Our aim is to investigate the surface properties of such a unique ordered $In_xGa_{1-x}As$ QDs array fabricated on GaAs (3 1 1)B substrate by using conductive atomic force microscope (AFM) technique.

A conductive AFM tip touches the substrate surface with an nm-scale contact area, typically 5–10 nm in diameter. Though the tip/surface contact area has not been determined exactly, it can be estimated with a reasonable accuracy by assuming a deformable metallic sphere representing the tip in contact with a deformable substrate surface [11]. The use of conductive AFM tip allows us to modify the local band profile of a given QD structure with external applied bias, whose size varies typically in the range of 20–50 nm in diameter. Consequently, the current–voltage (I–V) characteristics of QDs of varying sizes, and of any other arbitrary positions on the surface can be studied with conductive AFM tips. Furthermore, the electronic properties such as artificial atom-like energy states and shell filling of QDs with single electrons can be investigated with this type of technique [5,12]. Our previous study reported in Ref. [13] was conducted in an uncontrolled ambient in air. Thus, the obtained results were sensitive to the effect of surface oxidation due to the presence of an adsorbed thin layer of water and hence lacked in reproducibility. As a continuing effort, we have employed an UHV–AFM and all the samples were pre-heated in vacuum to remove the water layers prior to the measurement.

2. Experiments

Self-organized QDs were fabricated by depositing $\sim$12.7 monolayers of undoped $In_{0.4}Ga_{0.6}As$ on n-type GaAs (3 1 1)B substrate (doped to $1\times10^{18}\,cm^{-3}$) by atomic H-assisted molecular beam epitaxy (H-MBE), as reported in detail elsewhere [9,10]. To be particularly noted is the unique self-organization characteristics of $In_{0.4}Ga_{0.6}As$ QDs grown on GaAs (3 1 1)B substrate, and that the QDs size and density can be modulated simply by controlling the deposition temperature, while retaining the structural *ordering* of QDs array. Self-organized QDs studied in this work were fabricated at 520°C, and the QDs density and average size were $\sim3.3\times10^{10}\,cm^{-2}$ and 50 nm, respectively. The growth temperature was higher than that used in Ref. [13], for which the QDs were fabricated at 500°C resulting in a shorter inter-dot spacing. The hydrogen backpressure was kept constant at $\sim6\times10^{-6}$ Torr during substrate cleaning and H-MBE growth.

Si tips coated with Au, which warrants electrical conductivity of the tip, were used to measure both the topographic and current images of QDs surface. The I–V characteristics of QDs of varying sizes and of any other arbitrary positions on the QDs surface were also studied by using the same conductive tip. Though the tip radius of an Au-coated tip was 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. 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 either at room temperature, or cooled temperature at 173 K. The samples were transferred from the MBE chamber to our SPM system in dry nitrogen

ambient, which was then pumped down with a turbo-molecular pump to a base pressure of $< \sim 5 \times 10^{-7}$ Torr.

In this work, the following fundamental issues were investigated; (1) effect of thermal treatment of the sample on the subsequent SPM measurements, and (2) effect of structural ordering of $In_xGa_{1-x}As$ QDs on GaAs (3 1 1)B surface on local conductance measurements. These studies are thought to be essential, if the electronic properties such as artificial atom-like energy states and shell filling of QDs with single electrons are to be reliably investigated.

3. Results and discussion

First, it is known that applying a positive bias above threshold to a semiconductor surface with respect to SPM tip would lead to oxidation of the surface in the vicinity of tip apex due to the presence of a thin surface water layer. In fact, SPM tip-induced oxidation has been studied in detail in both Si [14,15], and GaAs [16–19]. If the conductance measurements are to be performed *in air*, one therefore needs to take into consideration of the upper limit of bias permissible for reliable measurements, of around <4 V in our case [13], thereby avoiding undesirable oxidation of the QDs surface. Furthermore, the surface water film is known to result in a strong adhesive force and defocusing of applied electric field between the tip and sample [15]. For this reason, each QDs sample was thermally treated in vacuum at 70°C for 1 h before proceeding to SPM characterization *in vacuum*. 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. Fig. 1 shows the traces of force curves measured in a high vacuum at room temperature (a) before, and (b) after the thermal treatment. The horizontal axis d represents the distance between the sample and tip. A minus d means that the tip is pushed against the sample. The vertical axis represents the atomic force generated between the sample and tip. Hence a positive or negative force means a repulsive or adhesive force. It was observed that before thermal treatment, the maximum adhesive force between the tip and sample under our scanning condition was ~ 16 nN, which was then reduced by a factor of 5 after thermal treatment at 70°C for 1 h. Thus, this moderate thermal treatment is useful in removing the surface water layer efficiently from the QDs surface in vacuum and this condition was used throughout this work. A more rigorous heat treatment of 200°C for 1 h was recently reported by Ono et al. [20].

Next, the topographic and current images, and the respective line scan images of $In_{0.4}Ga_{0.6}As$ QDs on GaAs (3 1 1)B substrate measured at room temperature are shown in Fig. 2(a) and (b). Both

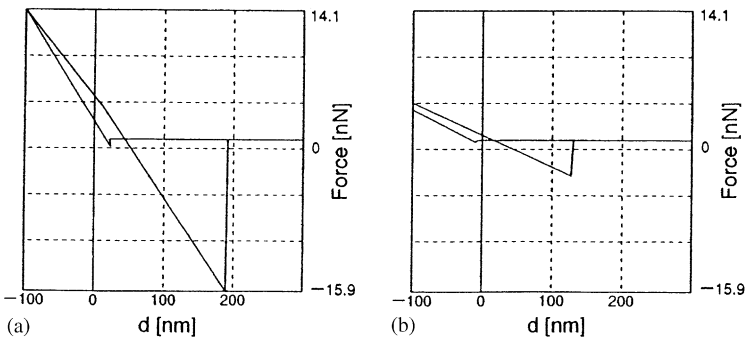

Fig. 1. Traces of force curves measured in high vacuum at room temperature (a) before, and (b) after the thermal treatment at 70°C for 1 h. The horizontal axis d represents the distance between the sample and tip. A minus d means that the tip is pushed against the sample. The vertical axis represents the atomic force generated between the sample and tip. Hence a positive or negative force means a repulsive or adhesive force. The maximum adhesive force before the thermal treatment between Au-coated Si AFM tip and QD was reduced by a factor of 5 after the treatment.

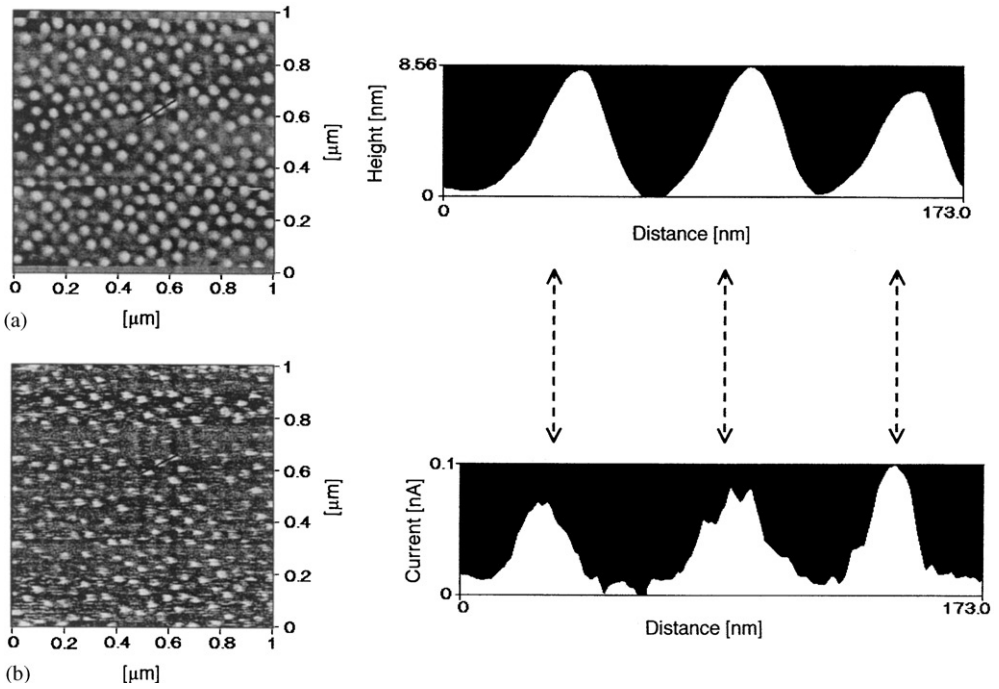

Fig. 2. (a) Topographic, and (b) current image of $In_{0.4}Ga_{0.6}As$ QDs grown on GaAs (3 1 1)B substrate measured in vacuum at room temperature after thermal treatment. Respective cross-sectional images along solid lines are also shown. Both images were obtained simultaneously at a sample bias of $V = 1$ V and the scan size was $1.0\,\mu m \times 1.0\,\mu m$.

images were obtained simultaneously at a sample bias of $V = 1$ V and the scan size was $1.0\,\mu m \times 1.0\,\mu m$. It can be observed that the topographic QDs size and height are relatively uniform and average size is ~ 50 nm. The QDs size was also measured in a separate experiment by operating the AFM in tapping mode which would provide an improved lateral resolution, and we have confirmed the observed QDs sizes in contact mode and tapping mode were identical. A partial structural ordering is also evident from Fig. 2(a). Also shown in Fig. 2(b), the local surface potential is dependent on the position along the top of QDs as well as on the position on the surface. Near the center of a QD was more conductive than its periphery, and if the tip was placed in between the QDs, the surface was found to be highly resistive.

These results are much similar to, and in fact have reproduced the observations reported for self-assembled InAs QDs grown on GaAs (0 0 1) by Tanaka et al. [7], in which the differences in the conductance were attributed to the local modification of surface bending associated with the surface states in InAs QDs and wetting layer. Though the growth mechanism of InAs QD on GaAs (0 0 1) by S–K mode differs from that of $In_xGa_{1-x}As$ QD on GaAs (3 1 1)B as discussed in the previous section, the fundamental features that are common to and responsible for the obtained results in both cases could be summarized as follows; (1) The area with no QDs is expected to be similar to that of bulk GaAs surface. On a bare GaAs surface, Fermi level would be pinned strongly at the mid bandgap energy by the negatively charged surface states, resulting in the formation of a surface depletion layer and hence suppression of current flow or conductance. (2) An electron accumulation occurs on the surface of InAs due to the positively charged surface states [21], which consequently leads to the lowering of Schottky barrier height. (3) The effect of surface potential lowering would be larger at the center of QD than its periphery.

This would in turn suggest that the effect of surface potential lowering would be larger for a large QD than a small QD [7]. Our present results as shown in Fig. 2 should be carefully compared with those previously reported in Ref. [13]. The QDs sample studied in Ref. [13] was fabricated at a lower growth temperature than that in Fig. 2, for which the QDs were smaller in average size of ~ 40 nm with a small size fluctuation, and almost twice in density $\sim 6 \times 10^{10}\,\mathrm{cm}^{-2}$. In such a closely packed QDs array surface on GaAs (3 1 1)B, the Schottky barriers were small and the local surface potential was thus practically uniform at all measured points over the whole (3 1 1)B surface at *room temperature* as of Fig. 5 in Ref. [13].

Next, the topographic and current images, and the respective line scan images of $In_{0.4}Ga_{0.6}As$ QDs on GaAs (3 1 1)B substrate measured at a lower temperature of 173 K are shown in Fig. 3(a) and (b). Here the QDs substrate was the same as that of Fig. 2, but an area with closely spaced QDs was deliberately chosen. By lowering the temperature to 173 K, the thermal current or Schottky conduction would be reduced at least by a factor of ~ 2 compared to room temperature. Both images were again obtained simultaneously at a sample bias of $V = 1$ V as before, but the scan size was reduced from Fig. 2 and it was $0.5\,\mu\mathrm{m} \times 0.5\,\mu\mathrm{m}$ in this case. It can be observed from the figure that, (1) the general characteristics observed at room temperature and at 173 K are essentially identical in the case of isolated QDs and at positions in between neighboring QDs as seen in Fig. 2. (2) However, for packed and ordered QDs, which are, for example, indicated by a solid line in the figure, the conductance and hence local surface potential were observed to be spatially uniform at all measured points over these QDs. Thus, for closely packed and ordered QDs surface structure, neighboring QDs are thought to be laterally coupled at low temperatures, which would then lead to a higher electrical conductance than that of

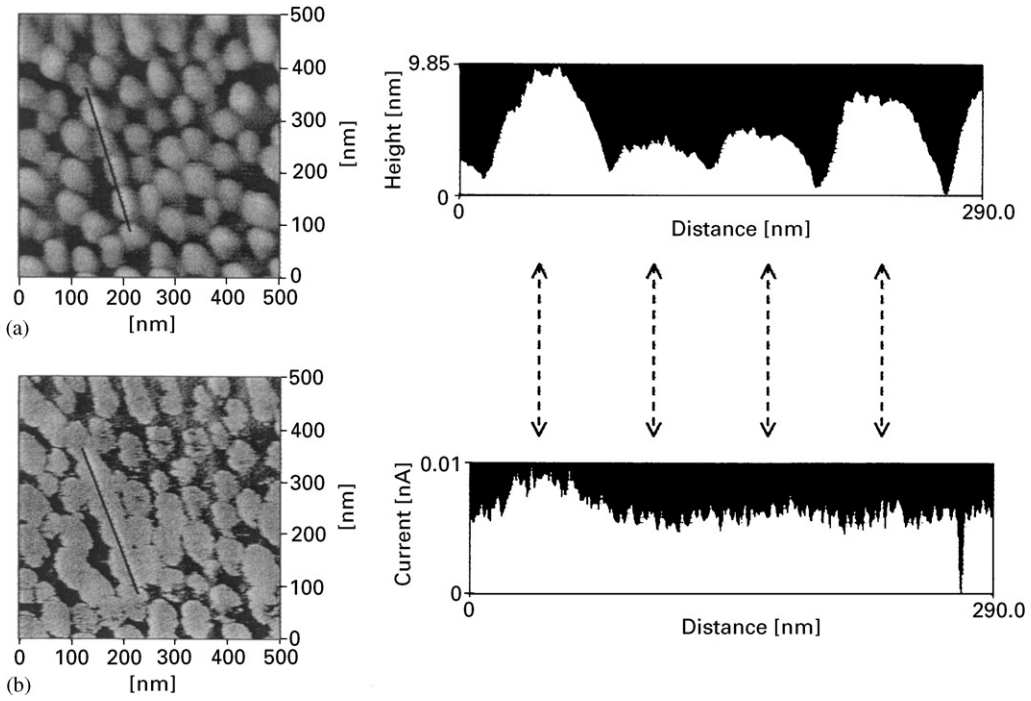

Fig. 3. (a) Topographic, and (b) current image of closely spaced $In_{0.4}Ga_{0.6}As$ QDs grown on GaAs (3 1 1)B substrate measured in vacuum at 173 K. Respective cross-sectional images along solid lines are also shown. Both were obtained simultaneously at a sample bias of $V = 1$ V and scan size was $0.5\,\mu\mathrm{m} \times 0.5\,\mu\mathrm{m}$.

isolated QDs [22]. This is further evidenced by the local I–V measurements conducted at the respective points on QDs surface. The results are plotted in Fig. 4: on top of isolated $In_{0.4}Ga_{0.6}As$ QD (marked 1), on top of packed QD (marked 2), and in between packed QDs (marked 3), respectively. It can be seen that a closely packed pair of QDs has a higher conductance and also a non-linear step-like characteristics than an isolated QD. A physical modeling of the observed non-linear I–V characteristics in view of coupling of QDs requires a thorough analysis as reported previously [22], and the modeling together with clarification of inter-dot coupling strength on dot separation is currently under study.

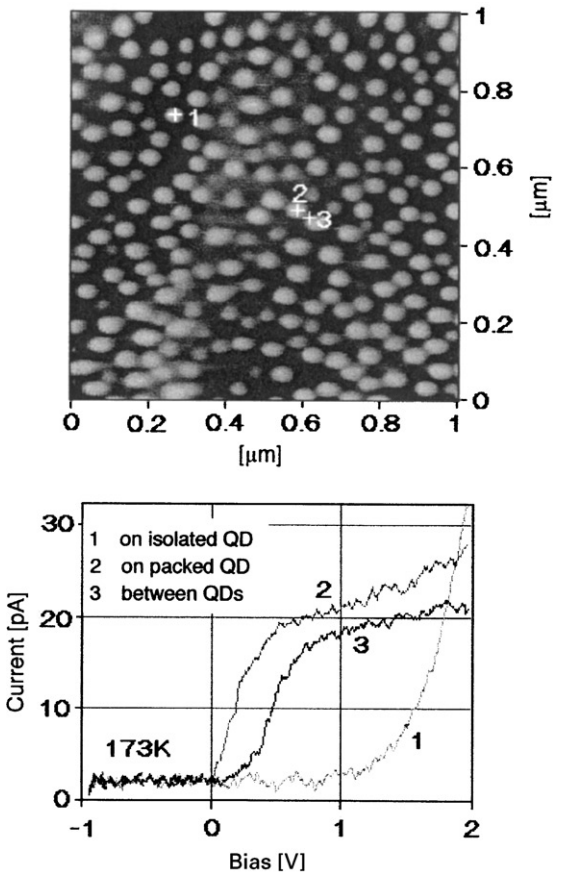

Fig. 4. I–V characteristics measured at respective points on QDs surface: on top of isolated $In_{0.4}Ga_{0.6}As$ QD (marked 1), on top of packed QD (marked 2), and in between packed QDs (marked 3).

The coupled QDs are expected to become the important elements in future quantum computing and QD memory applications [23–26]. In order for a strong coupling of neighboring QDs to occur, clean and identical electronic barriers must exist in between each QDs. Not only the size uniformity, but also the structural ordering of QDs becomes an important factor and to this regard, we have convincingly shown that self-organized $In_{0.4}Ga_{0.6}As$ QDs on GaAs (3 1 1)B substrates are promising candidate.

4. Summary

We have used conductive AFM technique to investigate the electronic state of $In_xGa_{1-x}As$ QDs surface grown on GaAs (3 1 1)B substrate. In order to ideally minimize the effect of adsorbed water layer on the surface, we first demonstrated, in terms of adhesive force acting between the tip and sample surface, that a moderate thermal treatment of QDs sample at 70°C for 1 h is very useful in suppressing the effect of surface water layer. The maximum adhesive force before thermal treatment between the tip and sample under our scanning condition was reduced by a factor of 5 after the given treatment. Second, the center of QD was found to be more conductive than its periphery, and the surface in between the QDs was measured to be highly resistive. The differences in the conductance were attributed to the local modification of surface bending associated with the surface states. Third, we showed that closely spaced and ordered QDs, which are characteristic of those fabricated on GaAs (3 1 1)B could be electronically coupled at low temperatures. A physical modeling of such a non-linear I–V characteristics in view of coupling state of QDs is currently under study, however, we have shown that the properties of QDs surface can be studied by this local probe technique.

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