Size, density, and shape of InAs quantum dots in closely stacked multilayers grown by the Stranski–Krastanow mode

Ken-ichi Shiramine[a] and Shunichi Muto
Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan

Tamaki Shibayama and Heishichiro Takahashi
Center for Advanced Research of Energy Technology, Hokkaido University, Sapporo 060-8628, Japan

Tamotsu Kozaki and Seichi Sato
Division of Quantum Energy Engineering, Graduate School of Engineering, Hokkaido University, Sapporo 060-8628, Japan

Yoshiaki Nakata and Naoki Yokoyama
Fujitsu Laboratories, Ltd., Atsugi 243-0197, Japan

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Closely stacked multilayer structures of InAs islands with intermediate-layer thicknesses d of 3, 6, 10, and 20 nm were grown by the Stranski–Krastanow mode of molecular beam epitaxy and were observed using transmission electron microscopy (TEM) and atomic force microscopy (AFM). The multilayers consisted of five InAs layers each of a thickness of 1.8 monolayers and four GaAs layers each of a thickness d. Columns of coherent islands were observed by cross-sectional TEM. Changes in the size and density of the islands with d, determined by AFM, could be explained in terms of (i) change in the vertical pairing probability of islands, (ii) detachment of In from the top of the island, and (iii) surface segregation of In. The observed AFM images of the islands were elliptical. Their major axis was in the $[\bar{1}10]$ direction, and the length of the minor axis was 80% of that of the major axis. [DOI: 10.1116/1.1605429]

I. INTRODUCTION

Semiconductor islands formed by the Stranski–Krastanow (S–K) mode of heteroepitaxial growth are known as self-assembled quantum dots.[1] The S–K mode is a growth mode in which three-dimensional growth (island growth) succeeds initial two-dimensional (2-D) growth (layer growth). Island formation by the S–K mode is a self-organization process. Growth of InAs on GaAs is a typical example of growth by the S–K mode; the present study concerns InAs/GaAs growth. The islands grown by the S–K mode are coherent and are sufficiently small to show a quantum-size effect.

By alternate growth of S–K InAs island layers and thin intermediate GaAs layers, islands are aligned in the growth direction to form a column.[2,3] For two island layers, islands of an upper layer are formed above existing islands in a lower layer; this is because an in-plane interatomic distance of GaAs above the islands becomes close to that of InAs, and the potential energy of In adatoms is smaller there.[2]

The state of electrons in S–K islands is changed by close stacking of the island layers.[3] The effective size of islands in a closely stacked multilayer structure is larger than that in a single-layer structure because electronic states in respective islands couple through thin intermediate layers by the tunnel effect and extend to the entire column. As a result, the peak wavelength of the photoluminescence (PL) spectrum of islands shifts to a longer wavelength by close stacking, and the width of the spectrum decreases simultaneously.

Let us discuss the details of the above change in the PL spectrum of S–K islands caused by close stacking of the island layers.[3,4] We now consider a column with intermediate layers of a thickness d and n layers of islands (Fig. 1). Because the height h is smaller than the diameter of a S–K island, here we assume that the energy level of a confined electron in an island depends only on h; i.e., the island is assumed to be equivalent (approximately) to a quantum well. As a result, the transition energy E, corresponding to the peak photon energy of the PL spectrum, between discrete electron energy levels in the island is expressed as $E \propto h^{-2}$. Assuming an extreme case of strong coupling, the peak wavelength of the PL spectrum shifts to a longer wavelength by close stacking with the increase in the height from h to $(n-1)d+h$ (Fig. 1). The distribution of the transition energy $\Delta E \propto \Delta h/h^3$ in the single layer, where Δh is the height distribution, decreases to $\Delta E' \propto \Delta h/[(n-1)d+h]^3$ by closely stacking, and the change is equivalent to size equalization. If the island size is equalized by closely stacking, in reality, $\Delta E'$ decreases further.

A quantum-dot laser, in which an island layer is used as an active layer of laser operation, is considered as a possible application of S–K islands to devices. The distribution, corresponding to the PL linewidth, of the transition energy in islands due to the size distribution deteriorates the quantum efficiency of the quantum-dot laser. In addition, a laser diode with an emission wavelength of 1.3 μm is in demand for optical communications, although the peak wavelength of the PL spectrum of islands is 1 μm. These two problems can be solved simultaneously by closely stacking because the PL spectrum changes as described above.

[a]Author to whom correspondence should be addressed; electronic mail: shira@eng.hokudai.ac.jp

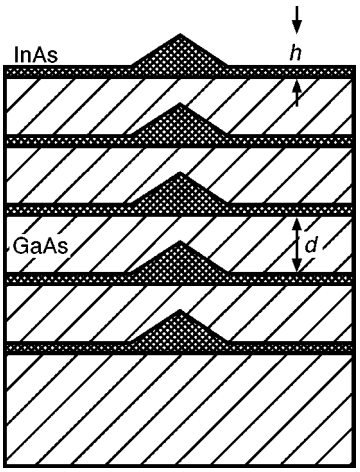

FIG. 1. Schematic illustration of a closely stacked multilayer structure of S–K InAs islands. The height of the column is expressed by $(n-1)d+h$ in disregard to the thickness of the wetting layer; where n is the number of island layers, d is the thickness of the intermediate GaAs layer, and h is the height of the island.

Many reports have been published about closely stacked S–K islands. Xie *et al.* pointed out that islands form a pair in the growth direction in two closely stacked island layers.[2] Solomon *et al.* reported electronic tunnel coupling in columns:[3] this is the first proposal of close stacking of S–K islands. It was reported that the island size in the top layer increases by close stacking.[5–7] Solomon *et al.* reported in-plane ordering of islands in the top layer of multilayers.[8]

In the present study, closely stacked multilayer structures of S–K InAs islands with various intermediate-layer thicknesses d were grown using molecular beam epitaxy (MBE) and were observed using transmission electron microscopy (TEM) and atomic force microscopy (AFM). The changes in the island size and density with d were determined systematically. The change will be explained comprehensively in terms of (i) change in the vertical pairing probability of islands, (ii) detachment of In from the top of the island, and (iii) surface segregation of In. The observed AFM image of the islands was elliptical; it reflected an anisotropy in the island shape. Its major axis was in the $[\bar{1}10]$ direction, and the length of the minor axis was 80% of that of the major axis.

II. EXPERIMENT

Closely stacked multilayer samples, which consist of S–K InAs island layers and intermediate GaAs layers, and a sample of a single island layer were grown using solid-source MBE (Varian, Gen II). Five growth runs were carried out for multilayers with intermediate-layer thicknesses d of 3, 6, 10, and 20 nm, and a single layer. A Si-doped (001) GaAs substrate, in which the dopant concentration is $1-2\times10^{18}\,\mathrm{cm}^{-3}$, was fixed on a Mo block by In solder and was installed in the MBE; then native oxide was eliminated from the substrate surface by thermal etching at 680 °C for 1 min under As_4 irradiation. A 400 nm GaAs layer, a 200 nm $Al_{0.3}Ga_{0.7}As$ layer, and a 100 nm GaAs layer were

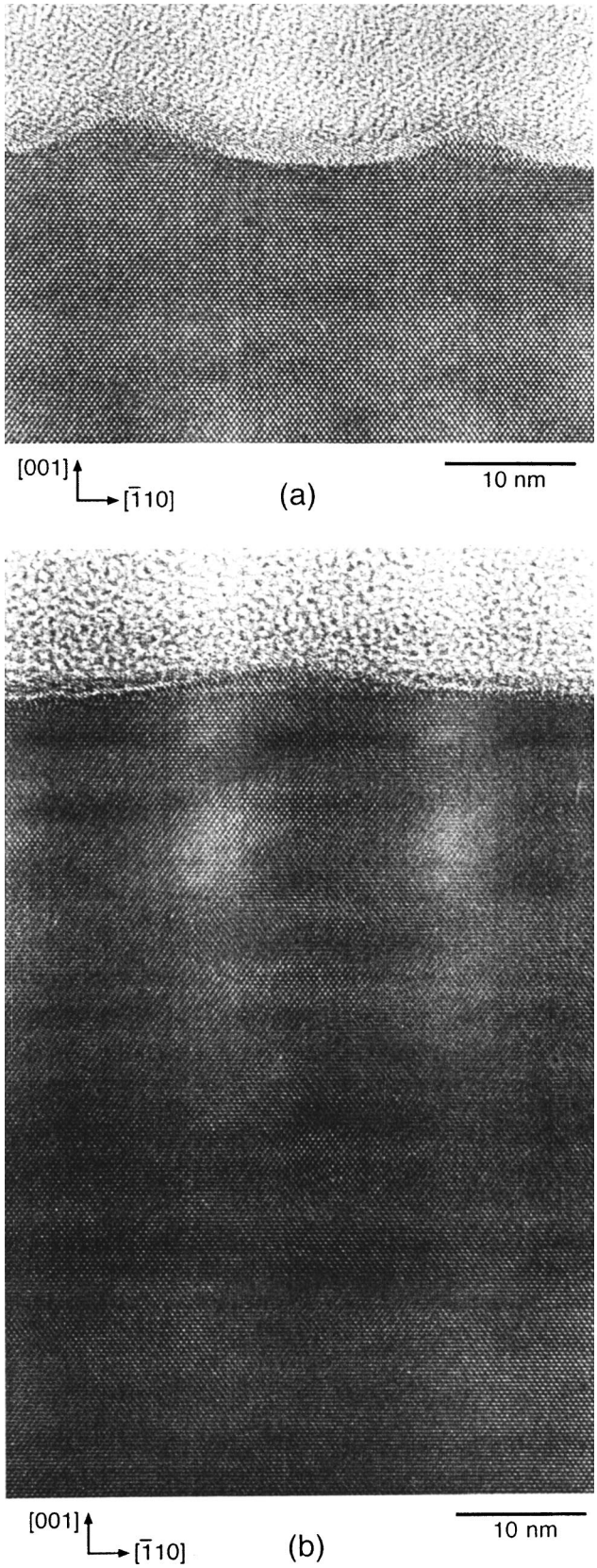

FIG. 2. Lattice images of S–K islands obtained by (110) cross-sectional TEM observations of (a) the single layer, and (b) the closely stacked multilayers of which an intermediate-layer thickness d is 10 nm. A column is observed in (b). Wetting layers are also observed beside the islands in (b).

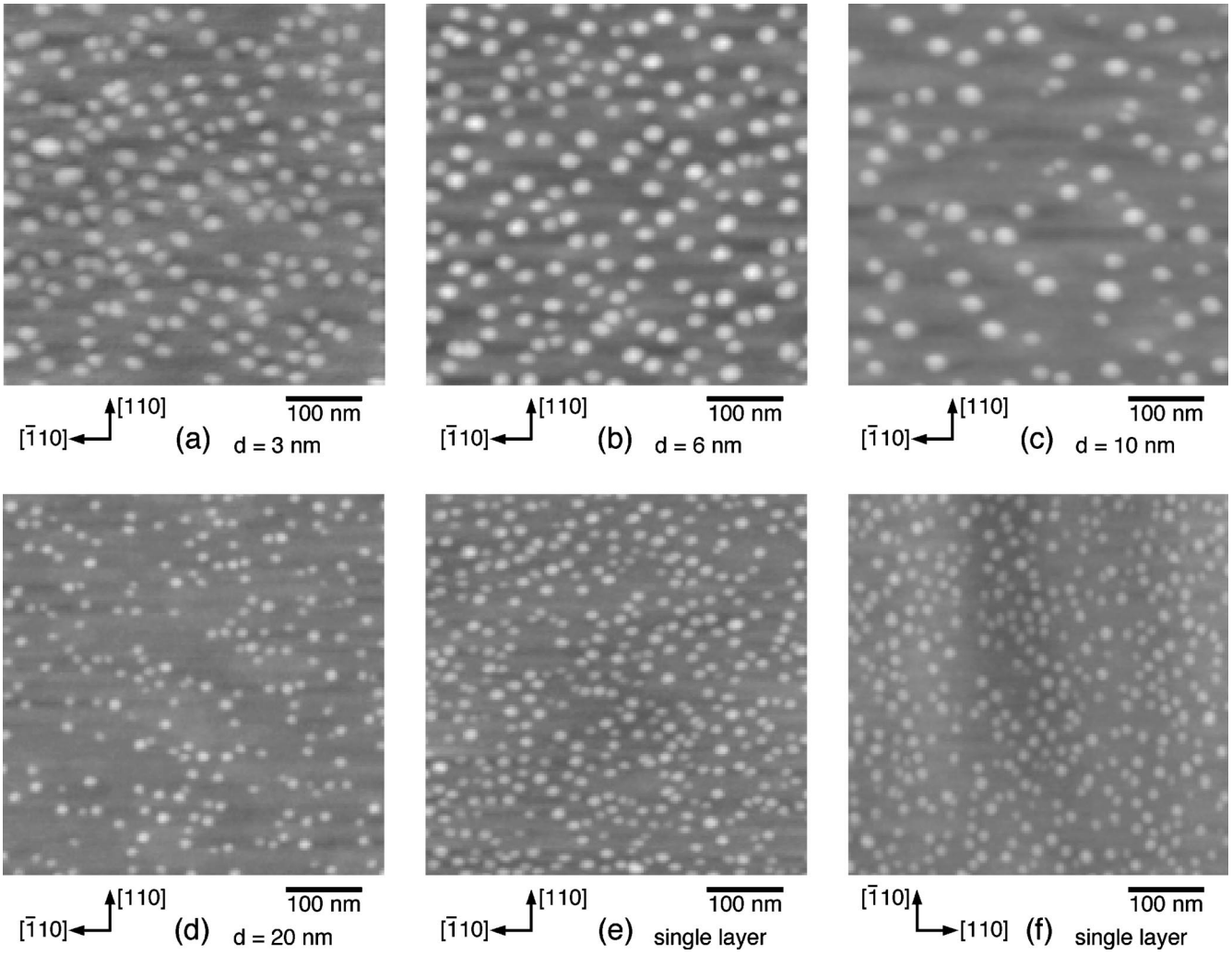

FIG. 3. Images observed by AFM of S–K islands of the multilayers with intermediate-layer thicknesses *d* of (a) 3, (b) 6, (c) 10, and (d) 20 nm, and (e)(f) the single layer. The scan direction was horizontal. The elliptical image in (f) was observed by 90° rotation of the same sample as (e), and its major axis was also rotated by 90°. Relations between contrast and height are different for respective images because of automatic scaling by AFM.

grown at 655 °C. The substrate temperature was decreased to 510 °C during growth interruption for 8 min. For multilayers, five InAs layers each of a thickness of 1.8 monolayers (ML) and four GaAs layers each of a thickness *d* were alternately grown with interruptions for 2 min before InAs growth except for the first layer, and for 1 min after InAs growth. The fifth InAs layer formed the surface. The substrate temperature was lowered after the last 1 min interruption. The growth rates were 0.1 ML/s for InAs, 0.77 μm/h for GaAs, and 1.1 μm/h for $Al_{0.3}Ga_{0.7}As$. The uncalibrated V/III ratios were 60 for InAs, and 30 for GaAs. For a single layer, InAs of 1.8 ML was grown on a $GaAs/Al_{0.3}Ga_{0.7}As/GaAs$ structure under the same conditions, and it formed the surface.

Cross-sectional observations of the multilayer with *d* of 10 nm and the single layer were carried out using a field-emission TEM (JEOL, JEM-2010F), operated at an acceleration voltage of 200 kV. Observed TEM samples were prepared by mechanical dimpling and thinning by Ar^+ irradiation with an acceleration voltage of 4 kV at an angle of 15° with an ion thinning machine (Gatan, model 600).

Surface topography of the samples was observed in air by AFM. A MultiMode SPM/NanoScope IIIa (Digital Instruments) was operated in contact AFM mode. A probe (Digital Instruments, NP-20) of Si_3N_4 was used. All the AFM images in the present article were obtained using the one probe alone. The scan area was 500×500 nm^2. The scan direction was the $[\bar{1}10]$ direction for Figs. 3(a)–3(e), and the [110] direction for Fig. 3(f) (from right to left for all the images). The scan rate was 3 scans/s (3 μm/s).

The obtained AFM images were processed by a ''flatten'' procedure in NanoScope IIIa software for reduction of line-like noise. The flatten procedure is the process whereby a fitted straight line is obtained for data of each scan and is subtracted from each pixel value. It sometimes causes an artifact, i.e., low stripes beside islands, as seen in Fig. 3.

The size and density of the islands were obtained from the AFM images. The images were analyzed using a public domain NIH Image program.[9] The density was determined from the number of islands in an image of 500×500 nm^2.

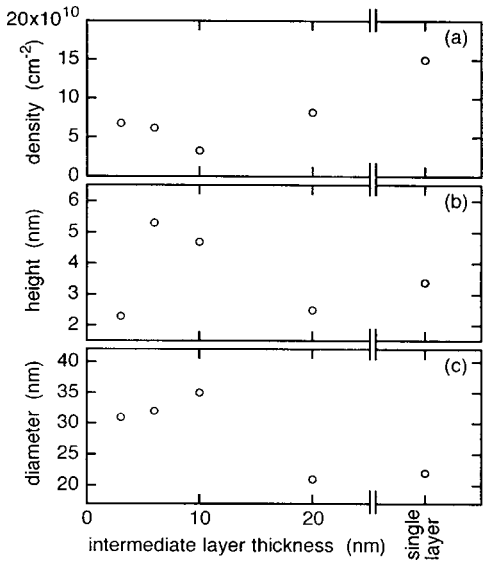

FIG. 4. Changes in (a) the area density, (b) height, and (c) in-plane diameter of S–K islands in the closely stacked multilayers with an intermediate-layer thickness *d*. The diameter is an average of the $[\bar{1}10]$ and [110] diameters. A statistical error due to the limited number, i.e., 30, of samples is nearly the extent of the plot symbol for the height and diameter.

The diameter was determined from $[\bar{1}10]$ and [110] profiles of 30 islands of each image. The height was determined only from $[\bar{1}10]$ profiles to avoid effects of the flatten procedure. The size determined using AFM can be different from the as-grown size because of native oxidation on the surface.

III. RESULTS

Figure 2 shows lattice images observed by cross-sectional TEM of the single layer (a), and the multilayers with *d* of 10 nm (b). Coherent islands were observed in both samples. The islands were on the surface in Fig. 2(a). The islands were aligned in the growth direction to form a column in Fig. 2(b). No lattice defects, e.g., dislocations or stacking faults, were observed in the course of the present study. The lattice image of Fig. 2(b) was somewhat distorted; the distortion was probably caused by a strain field around the column.

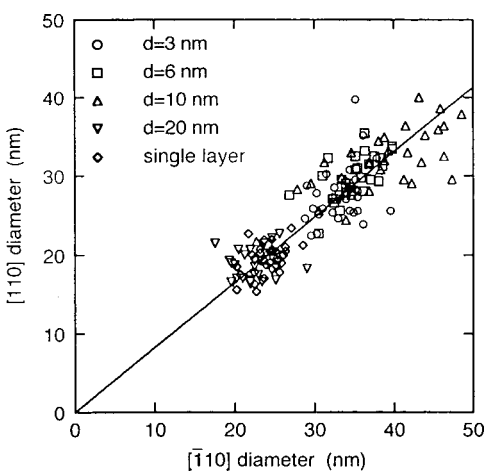

FIG. 5. Relation between the $[\bar{1}10]$ diameter and the [110] diameter of S–K islands in the closely stacked multilayers and the single layer. A fitted line was determined on the assumption that the [110] diameter is in proportion to the $[\bar{1}10]$ diameter.

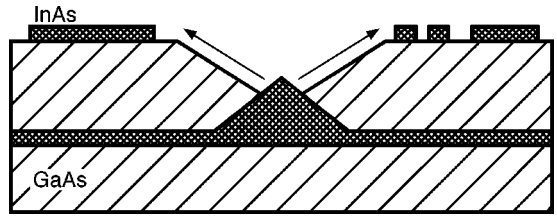

FIG. 6. Schematic illustration of In detachment from the S–K InAs island for a thin intermediate GaAs layer. Indium atoms detach from the top of the island to migrate away, and a partial wetting layer is formed on the GaAs layer. The scheme is based on a model of Ledentsov *et al.*[10]

Figure 3 shows AFM images of the multilayers (a)–(d) and the single layer (e), (f). The area density, height, and in-plane diameter of the islands, determined from Fig. 3, are shown in Fig. 4. The density of the multilayers with *d* of 20 nm was half that of the single layer. It showed the lowest value at *d* of 10 nm. It increased with smaller *d* although it is still smaller than that of the single layer. The island sizes for *d* of 20 nm were nearly equal to those of the single layer. However, the sizes for *d* of 6 and 10 nm were larger than those of the single layer. At *d* of 3 nm, the diameter was larger than that of the single layer although the height was smaller.

The observed AFM images of the islands were elliptical, of which the major axis was in the $[\bar{1}10]$ direction (Fig. 3). The elliptical, rather than circular, image reflected an anisotropy in the island shape. This was established by observation of the 90°-rotated sample; the obtained image, of which the major axis was rotated by 90° [Fig. 3(f)], implies that the elliptical image was not caused by an image drift and did not depend on the scan direction. Rotation of the other samples showed the same results (not shown here). Figure 5 shows the relation between the $[\bar{1}10]$ diameter and the [110] diameter. The data points concentrated around a straight line; this means that a ratio of the major axis length to the minor axis length has a constant value. The ratio was determined to be 1:0.8 from the slope of the fitted line of Fig. 5.

IV. DISCUSSION

The size and density of the S–K islands of the closely stacked multilayers changed with *d* as shown in Fig. 4. The change can be explained in terms of (i) change in the vertical pairing probability of islands, (ii) In detachment from the top of the island, and (iii) surface segregation of In. Before detailed discussion on the changes in the size and density, let us consider these effects in closely stacked InAs/GaAs multilayers.

First, an island in an upper layer is formed preferably above an existing island in a lower layer to form a pair in the growth direction as mentioned above, and the pairing probability changes with *d*. The pairing probability for two island layers was reported by Xie *et al.*[2] The pair correlation disap-

pears for $d>33$ nm. The probability increases with a decrease in d, and it is larger than 95% for $d<11$ nm.

Second, In atoms detach from the top of islands to form a wetting layer (WL) on GaAs after a thin GaAs layer is grown on the island layer.[10] The WL is 1 ML of InAs originating in the initial 2-D growth of the S–K mode. When GaAs is grown on InAs islands, GaAs does not prefer to grow on the islands.[11,12] Therefore, the top of an island is not covered with a thin GaAs layer. During interruption after GaAs-layer growth, In atoms detach from the top of the island to become adatoms, and they migrate to form a partial WL on GaAs (Fig. 6).[10] In Fig. 6, In atoms on the top of the island can decide whether to stay in the island or to migrate and to form a layer on GaAs. In such a case, InAs prefers to cover the GaAs surface; this can be understood from the fact that InAs formed a WL to cover the GaAs surface prior to the formation of islands when InAs was grown on GaAs originally. Therefore, an effective In supply of the second and successive layers is more than the nominal value in multilayer structures.[13] For a thick GaAs layer, islands are completely covered at some point; however, the effect can occur. Even for a thick GaAs layer, the top of an island is not covered at an early point of growth time of the GaAs layer. Then, In atoms can detach from the top of the island. In this case, the In atoms should be segregated to a surface over the rest of GaAs to participate in the next InAs layer.

Thirdly, surface segregation of In is expected at the growth temperature and the growth time in the present study, although its effect on growth is hard to determine quantitatively.

Let us return to the changes in the size and density of the islands with d (Fig. 4). Here we consider the single layer as a standard; we can regard it as multilayers with $d\rightarrow\infty$. The change in the density with d is in a V shape whose dip is at $d=10$ nm. Two factors, i.e., the change in the probability of forming a column, and the change in the probability of halfway disappearance of a column, affect the change in the density with d. The former is dominant for $d>10$ nm, and the latter is dominant for $d<10$ nm. Details can be explained as follows.

In the multilayers with d of 20 nm, the density was half that of the single layer, and the size was nearly equal. The islands act as if they were nucleating on a surface with no subsurface islands at d of 20 nm.[2,14,15] This would result in island sizes that are similar to the single layer as observed. The decrease in the density, reported in Refs. 5 and 6 also, is inferred to be caused by some effect from the lower island layer although its detail is unknown.[16] The effective increase in In supply for the second and successive layers, as mentioned above (ii), may be the reason for the decrease in the density.

In the multilayers with d of 10 nm, the density was smaller than that of the multilayers with d of 20 nm, and the size was larger. The decrease in the density with d can be explained as follows. The pairing probability[2] increases with a decrease in d and is larger than 95% at d of 10 nm. However, the islands are not formed on all the islands in the lower layer; i.e., some columns disappear halfway.[2,7,15,17] In the second and successive layers, nucleation hardly occurs without islands under there. In the early stage of growth, nucleation on islands in the lower layer is energetically favorable. In the late stage, In adatoms hardly nucleate because the adatom density decreases. Consequently the density of the top layer should decrease. The size should increase because all the In atoms assemble to the islands with reduced density. The effective increase in the In supply, (ii), and the In segregation, (iii), also contribute to the increase in the size.

In the multilayers with d of 3 and 6 nm, the densities were larger than that with d of 10 nm, and the sizes were smaller; although the height for d of 6 nm was higher than that for d of 10 nm. The pairing probability increases[2] from that for d of 10 nm; as a result, the density increases, and the size decreases.

An elliptical AFM image of S–K islands has been reported already for a single layer. These reports are consistent with the results of the present study. Mirin *et al.* reported that a (001) section of an island is elliptical, and the $[\bar{1}10]$ and $[110]$ diameters are 55 and 42 nm, respectively, on AFM observations of 13.3 ML of $In_{0.3}Ga_{0.7}As$.[18] Nabetani *et al.* reported that the island shape is anisotropic in plane, and the $[\bar{1}10]$ and $[110]$ diameters are 15 and 13 nm, respectively, on plan-view TEM observations of 2 ML of InAs.[19] The S–K island has been reported to have facets.[19–22] The islands of the present study also probably have facets, and the obtained elliptical AFM image can reflect this.

We can summarize our discussion as follows. The changes in the size and density of S–K InAs islands in closely stacked multilayers with d have been determined systematically and explained comprehensively in terms of the characteristics of S–K growth of InAs/GaAs multilayers. The observed elliptical AFM image has been discussed in relation to the shape of the islands.

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[1]D. Leonard, M. Krishnamurthy, C. M. Reaves, S. P. Denbaars, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[2]Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).
[3]G. S. Solomon, J. A. Trezza, A. F. Marshall, and J. S. Harris, Jr., Phys. Rev. Lett. **76**, 952 (1996).
[4]M. K. Zundel, P. Specht, K. Eberl, N. Y. Jin-Phillipp, and F. Phillipp, Appl. Phys. Lett. **71**, 2972 (1997).
[5]G. S. Solomon, S. Komarov, J. S. Harris, Jr., and Y. Yamamoto, J. Cryst. Growth **175/176**, 707 (1997).
[6]Y. Nakata, Y. Sugiyama, T. Futatsugi, and N. Yokoyama, J. Cryst. Growth **175/176**, 713 (1997).
[7]Z. R. Wasilewski, S. Fafard, and J. P. McCaffrey, J. Cryst. Growth **201/202**, 1131 (1999).
[8]G. S. Solomon, S. Komarov, and J. S. Harris, Jr., J. Cryst. Growth **201/202**, 1190 (1999).
[9]http://rsb.info.nih.gov/nih-image/.
[10]N. N. Ledentsov *et al.*, Phys. Rev. B **54**, 8743 (1996).
[11]Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).

[12]This is because S–K InAs islands are partially relaxed elastically and the potential energy of Ga adatoms is larger on the islands; see Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).
[13]Island formation is promoted by closely stacking. In the second and successive island layers, islands are formed at an earlier stage than those in the first layer. One of the present authors (Y.N.) has reported that the growth time, corresponding to In supply, required for islanding in the third and fifth layers is 63% of that of the first layer in a five-layer structure with *d* of 3 nm on the basis of the results of *in situ* reflection high energy electron diffraction observations [Y. Nakata, Y. Sugiyama, T. Futatsugi, and N. Yokoyama, J. Cryst. Growth **175/176**, 713 (1997)]. The promotion of islanding was ascribed to a strain field from existing islands in the lower layer and In surface segregation; [Y. Nakata, Y. Sugiyama, T. Futatsugi, and N. Yokoyama, J. Cryst. Growth **175/176**, 713 (1997)] in addition, the effective increase in In supply for the second and successive island layers, (ii), can also contribute.
[14]K. Shiramine, Y. Horisaki, D. Suzuki, S. Itoh, Y. Ebiko, S. Muto, Y. Nakata, and N. Yokoyama, Jpn. J. Appl. Phys., Part 1 **37**, 5493 (1998).
[15]M. A. Migliorato, L. R. Wilson, D. J. Mowbray, M. S. Skolnick, M. Al-Khafaji, A. G. Cullis, and M. Hopkinson, J. Appl. Phys. **90**, 6374 (2001).
[16]There is still an open question concerning conservation of volume.
[17]Y. Sugiyama, Y. Nakata, K. Imamura, S. Muto, and N. Yokoyama, Jpn. J. Appl. Phys., Part 1 **35**, 1320 (1996).
[18]R. Mirin, A. Gossard, and J. Bowers, Electron. Lett. **32**, 1732 (1996).
[19]Y. Nabetani, T. Ishikawa, S. Noda, and A. Sasaki, J. Appl. Phys. **76**, 347 (1994).
[20]H. Lee, R. Lowe-Webb, W. Yang, and P. C. Sercel, Appl. Phys. Lett. **72**, 812 (1998).
[21]T. Kaizu and K. Yamaguchi, Jpn. J. Appl. Phys., Part 1 **40**, 1885 (2001).
[22]J. Márquez, L. Geelhaar, and K. Jacobi, Appl. Phys. Lett. **78**, 2309 (2001).