Crystal shape of GaAs nanocrystals deposited on Si(100) by molecular beam epitaxy

Usui, Hiroyuki
Yasuda, Hidehiro
Mori, Hirotaro

(Citation)
Thin Solid Films,516(9):2487-2490

(Issue Date)
2008-03

(Resource Type)
journal article

(Version)
Accepted Manuscript

(URL)
https://hdl.handle.net/20.500.14094/90001070

Crystal shape of GaAs nanocrystals deposited on Si(100) by molecular beam epitaxy

Hiroyuki Usui[a,*], Hidehiro Yasuda[b] and Hirotaro Mori[c]

[a] *Venture Business Laboratory, Kobe University, Nada, Kobe 657-8501, Japan*

[b] *Department of Mechanical Engineering, Kobe University, Nada, Kobe 657-8501, Japan*

[c] *Research Center for Ultra-High Voltage Electron Microscopy, Osaka University, Yamadaoka, Suita, 565-0871, Japan*

*Corresponding author: +81-078-803-6121, fax number: +81-078-803-6155,
e-mail address: usui@mech.kobe-u.ac.jp

Abstract

The shape changes in GaAs nanocrystals deposited on Si substrate have been studied as a function of the coverage by transmission electron microscopic observations in order to see the growth mechanism from the viewpoint of the surface energy and the lattice strain energy between the nanocrystal and the substrate. When GaAs was deposited on the Si(100) surface, the shape of the GaAs nanocrystals changes from stepped mound, hut cluster to dome structure with increasing the coverage. The shape changes are responsible for decreasing the total free energy caused by the lattice strain energy with the substrate and surface energy depending on the crystal size.

Keyword: Crystal growth; Nanoscale material; TEM; GaAs/Si

1. Introduction

Heteroepitaxial growth of GaAs crystal on Si substrate is greatly required for developing important devices such as high-efficiency solar cells [1] and optoelectronic integrated circuits [2]. It is known that GaAs nanocrystals three-dimensionally grow on a flat Si surface because of elastic lattice strain caused by the lattice mismatch of 4% between GaAs and Si [3]. The elastic strain strongly affects growth mode and crystal shape of nanocrystals during the lattice-mismatched heteroepitaxial growth. It is very important for the realization of optoelectronic devices to make size and crystal shape of nanocrystals uniform. In these days, researches on the crystal size and shape have been carried out in epitaxially grown nanocrystals of InAs on GaAs [4-8], Ge on Si [8-11], and so on. However, systematic researches on the crystal size and shape of GaAs nanocrystals grown on Si are relatively limited. In this study, the shape changes in GaAs nanocrystals deposited on Si substrate have been studied as a function of the coverage by transmission electron microscopic (TEM) observations in order to see the growth mechanism from the view point of the free energy change in nanocrystals.

2. Experimental details

Exactly Si(100)-oriented substrates were used for GaAs deposition to avoid step-flow growth by dense step edges on vicinal surface. The Si substrates were annealed at 1100K for 5 minutes to remove native oxide from the surface in the vacuum chamber at the base pressure below 2.0×10^{-7} Pa. Further detailed annealing procedures have been described in our previous paper [12]. Surface reconstruction of (2×1) and (1×2) domains on Si(100) surface were observed by in situ reflection high-energy electron diffraction. GaAs nanocrystals were deposited on the Si surface by molecular beam epitaxy at the pressure of approximately 9×10^{-3} Pa. The growth temperature, growth rate and effective V/III pressure ratio were 673K, about 0.8 monolayer (ML)/min and 30, respectively. The coverage of GaAs was set to be 0.4, 4 and 17 ML to investigate changes in the crystal shape. After the deposition, TEM specimens for cross-sectional observation were prepared by sequential gluing of the Si substrates to Si blocks, mechanical polishing, dimpling, followed by argon ion milling to electron transparency. The morphology and structure of GaAs grown on the Si substrate were observed by bright-field image (BFI) and high-resolution image (HRI). The microscope used was Hitachi HF-2000, operating at an accelerating voltage of 200 kV.

3. Results and discussion

Figures 1(a) and (b) depict a cross-sectional BFI and HRI of GaAs grown up to 0.4 ML on the Si substrate, respectively. The incident beam direction is <011>. In the BFI (Fig. 1(a)), GaAs nanocrystals which have a triangular cross section are observed on the Si substrate. The width in the <011> directions on the basal plane of the nanocrystals is ranging from 20 to 30 nm, and the height of the nanocrystals is approximately 2 to 3 nm. The {011} lattice spacing in the GaAs nanocrystal measured by the lattice image in Fig. 1(b) is estimated to be 0.38_9 nm, which is approximately 2.5% smaller than that in bulk GaAs (i.e., 0.40_0 nm), and is almost similar to that in the Si substrate. The multi terraces of (100) planes are observed on the surface of the GaAs nanocrystal as indicated by white arrows in Fig. 1(b). The structure consisting of the multi terraces and single steps what is called "stepped mound" [13] appears on the substrate.

Figures 2(a) and (b) show a cross-sectional BFI and HRI of GaAs grown up to 4 ML on the Si substrate, respectively. The incident beam direction is <011> again. As shown in Fig. 2(a), the width in the <011> directions on the basal plane of the nanocrystals is ranging from 20 to 30 nm, and is equivalent to that at the coverage of 0.4 ML. The facet planes in all of the GaAs nanocrystals and the Si (100) surface make an angle of 55 degrees, which is equivalent to an angle of 54.4 degrees between {100} and {111} plane. On the other hand, the other important point to be noted here is the fact that all of the nanocrystals have a fixed height of approximately 14 nm. Fig. 2(b) shows a typical example of HRIs in GaAs nanocrystals. The facet structure consisting of facet planes of {111} type what is called “hut cluster” [14] is observed at this coverage, and is stabilized by the lowest {111} surface energy of GaAs which has the zinc-blende structure. Lattice mismatches on the interface and dense stacking faults are recognized in the nanocrystal [15]. The {011} lattice spacing in the GaAs nanocrystal measured by the lattice image was estimated to be 0.40_3 nm, which is almost equivalent to that in bulk GaAs.

Figures 3(a) and (b) present a cross-sectional BFI and HRI of GaAs grown up to 17 ML on the Si substrate, respectively. The incident beam direction is <011> again. As shown in Fig. 3(a), the width in the <011> directions on the basal plane of the nanocrystals significantly increases to 100-200 nm, and the height of the nanocrystals becomes 50-100 nm. The number density of the nanocrystals decreases because of a significant coalescence of the nanocrystals during the growth. Fig. 3(b) shows a typical example of HRIs in GaAs nanocrystals. Two kinds of facet planes observed in the nanocrystal and the Si(100) surface make angles of 55 and 26 degrees, which are equivalent to an angle of 54.4 degrees between {100} and {111} plane and an angle of 25.2 degrees between {100} and {311} plane, respectively. It was revealed that the nanocrystal has the "dome" structure consisting of {111} and {311} facet planes.

From these results, it was evident that when GaAs was deposited on the Si(100) surface, the shape of the GaAs nanocrystals changes from stepped mound, hut cluster to dome structure with increasing the coverage.

A mechanism of the shape changes in GaAs nanocrystals will be discussed from the viewpoint of the interfacial strain between the nanocrystal and the substrate and surface energy as follows. In the GaAs crystal grown on the Si substrate in which the lattice mismatch is approximately 4%, it is difficult due to the large interfacial lattice strain that the continuous GaAs thin film grows coherently on the Si substrate. An upper limit of the interfacial area appears in order to prevent the increase of the lattice strain energy caused by compressive strain accumulated in the coherently-grown GaAs nanocrystal, and consequently, the mound-type nanocrystalline nuclei consisting of the basal plane with the interfacial width ranging from 20 to 30 nm in the <011> directions are formed at the low coverage. The initially-formed mounds grow under a nearly constant interfacial area with increasing coverage. The angle between the facet planes on the mounds and the substrate surface gradually increases to form the subsequent {111} facet planes which have the lowest surface energy under As-rich environment [16]. When the GaAs nanocrystal has the structure consisting of the facet planes of {111} type and the basal plane limited by the interfacial area mentioned above, the nanocrystal becomes a hut cluster in which the height is fixed to be approximately 14 nm. The hut clusters coalesce into larger nanocrystals with increasing coverage. The {111} facet planes around the top of the nanocrystal change to the {311} facet planes to lower the total surface energy of the nanocrystals, and then the dome structure is formed. It is suggested that the shape of the GaAs nanocrystals deposited on the Si(100) substrate changes to decrease the total free energy which is responsible for the lattice strain energy with the substrate and surface energy depending on the crystal size.

4. Conclusion

The shape changes in GaAs nanocrystals deposited on Si substrate have been studied as a function of the coverage by TEM. When GaAs was deposited on the Si(100) surface, the shape of the GaAs nanocrystals changes from stepped mound, hut cluster to dome structure with increasing the coverage. The shape changes are responsible for decreasing the total free energy caused by the lattice strain energy with the substrate and surface energy depending on the crystal size.

Acknowledgements

This work was, in part, supported by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) Japan under “Grant-in-Aid for Young Scientists (B)”, “Grant-in-Aid for Scientific Research” and “Nanotechnology Support Project”.

References

[1] Y. Shimizu, Y. Okada, J. Cryst. Growth, 265(2004)99.

[2] I. Hayashi, Jpn. J. Appl. Phys., 32(1993)266.

[3] V. H. M.-García, A. P.-Centeno, M. López, Thin Solid Films, 433(2003)63.

[4] I. Mukhametzhanov, Z. Wei, R. Heitz, A. Madhukar, Appl. Phys. Lett., 75(1999)85.

[5] K. Yamaguchi, K. Yujobo, T. Kaizu, Jpn. J. Appl. Phys., 39(2000)L1245.

[6] T. Kaizu, K. Yamaguchi, Jpn. J. Appl. Phys., 40(2001)1885.

[7] T. Kaizu, K. Yamaguchi, Jpn. J. Appl. Phys., 42(2003)4166.

[8] G. Costantini, A. Rastelli, C. Manzano, P. A.-Diaz, G. Katsaros, R. Songmuang, O. G. Schmidt, H. V. Känel, K. Kern, J. Cryst. Growth, 278(2005)38.

[9] F. M. Ross, J. Tersoff, R. M. Tromp, Phys. Rev. Lett., 80(1998)984.

[10] G. M.-Ribeiro, T. I. Kamins, D. A. A. Ohlberg, and R. S. Williams, Mater. Sci. Eng. B, 67(1999)31.

[11] M. Kästner, B. Voigtländer, Phys. Rev. Lett., 82(1999)2745.

[12] H. Usui, K. Ishiji, H. Yasuda, H. Mori, J. Cryst. Growth, 289(2006)76.

[13] A. Rastelli, M. Kummer, H. Känel, Phys. Rev. Lett., 87(2001)256101.

[14] A. I. Nikiforov, V. A. Cherepanov, O. P. Pchelyakov, A. V. Dvurechenskii, A. I. Yakimov, Thin Solid Films, 380(2000)158.

[15] H. Usui, H. Yasuda, H. Mori, Appl. Phys. Lett., 89(2006)173127.

[16] N. Moll, A. Kley, E. Pehlke, M. Scheffler, Phys. Rev. B, 54(1996)8844.

Figure caption

Fig.1. (a) A cross-sectional BFI and (b) HRI of GaAs grown up to 0.4 ML on the Si substrate. The width on the basal plane of the nanocrystals is ranging from 20 to 30 nm. The structure consisting of multi terraces and single atomic height steps what is called “stepped mound” appears on the surface.

Fig.2. (a) A cross-sectional BFI and (b) HRI of GaAs grown up to 4 ML on the Si substrate. The width on the basal plane of the nanocrystals is ranging from 20 to 30 nm. The facet structure consisting of {111} type facets what is called “hut cluster” is observed on the Si surface.

Fig.3. (a) A cross-sectional BFI and (b) HRI of GaAs grown up to 17 ML on the Si substrate. The width on the basal plane of the nanocrystals is ranging from 100 to 200 nm. The GaAs nanocrystals have “dome” structure consisting of {111} and {311} facet planes.

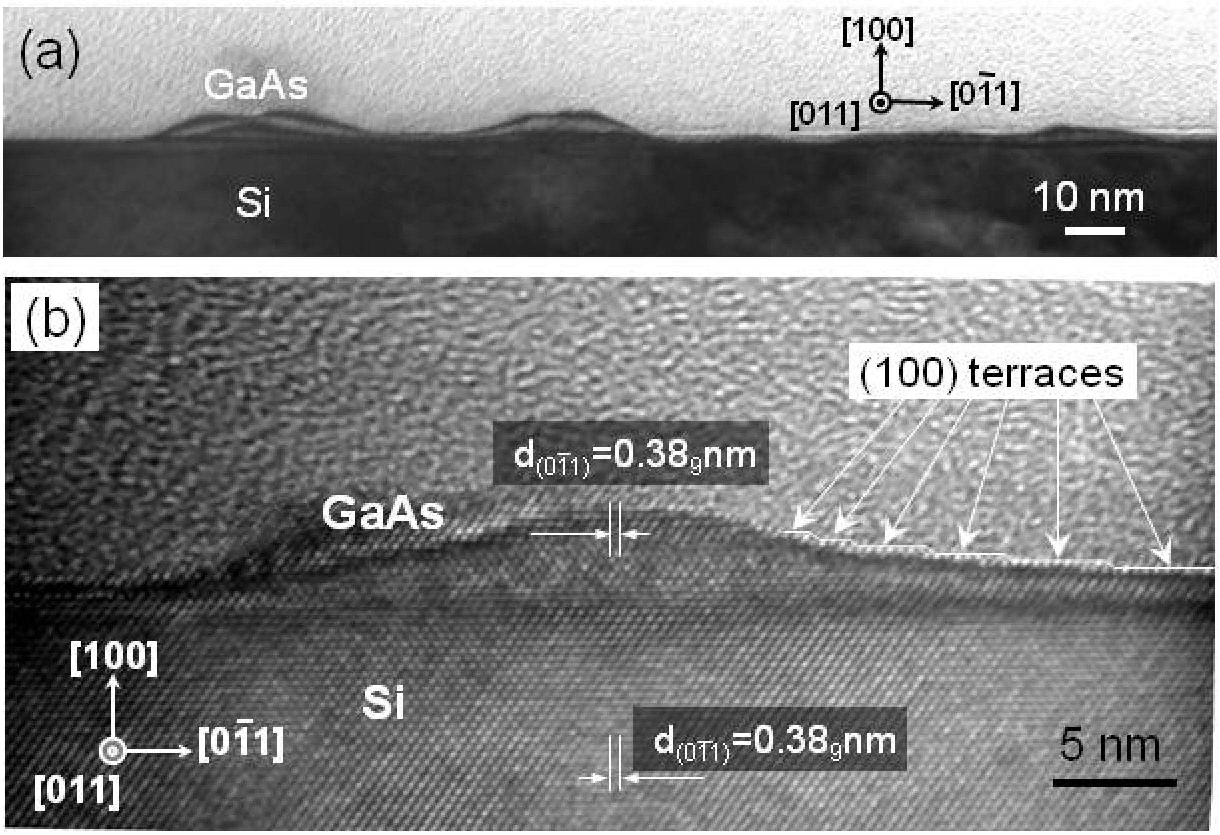

Fig. 1 H. Usui *et al.*

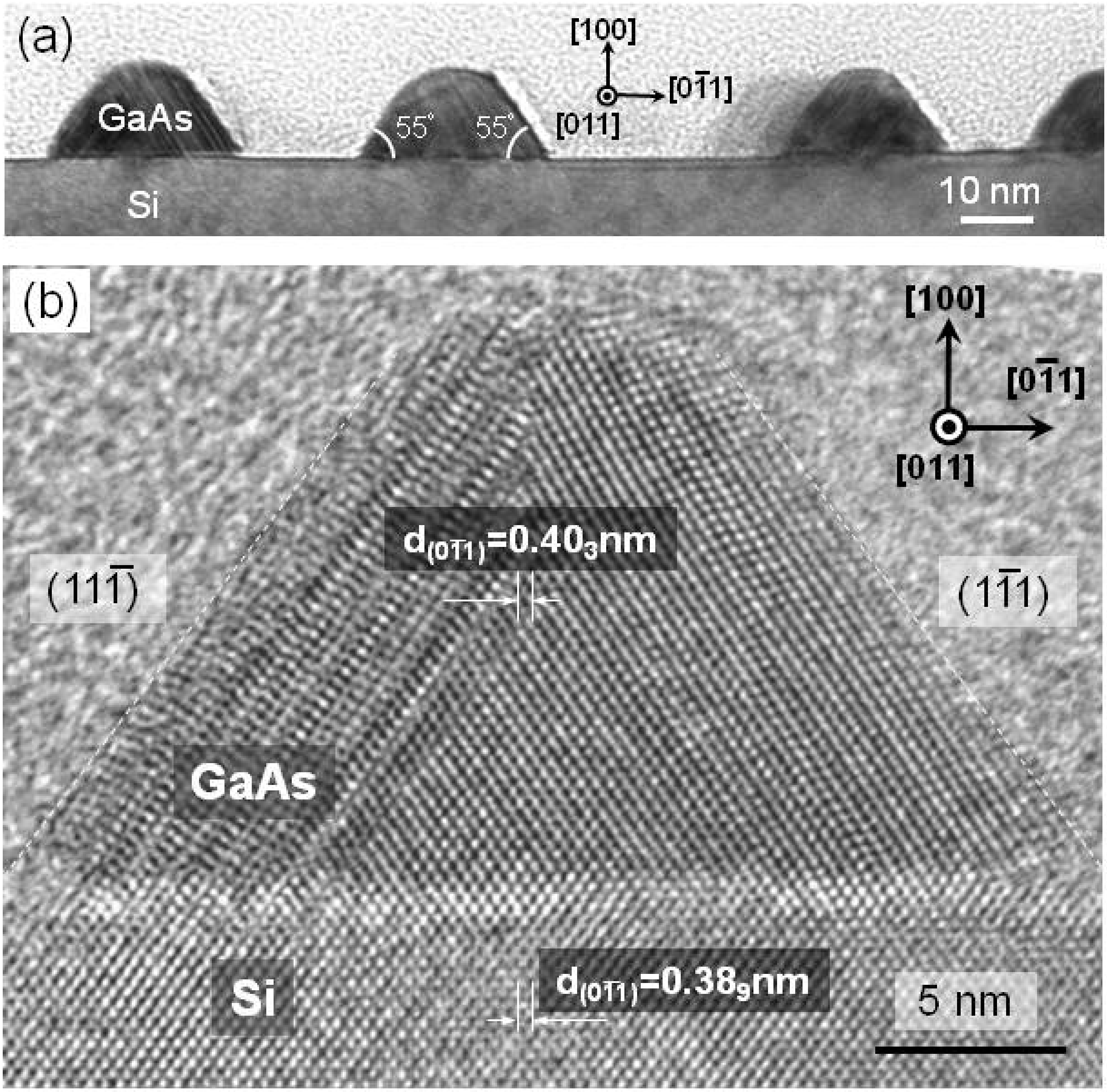

Fig. 2 H. Usui *et al.*

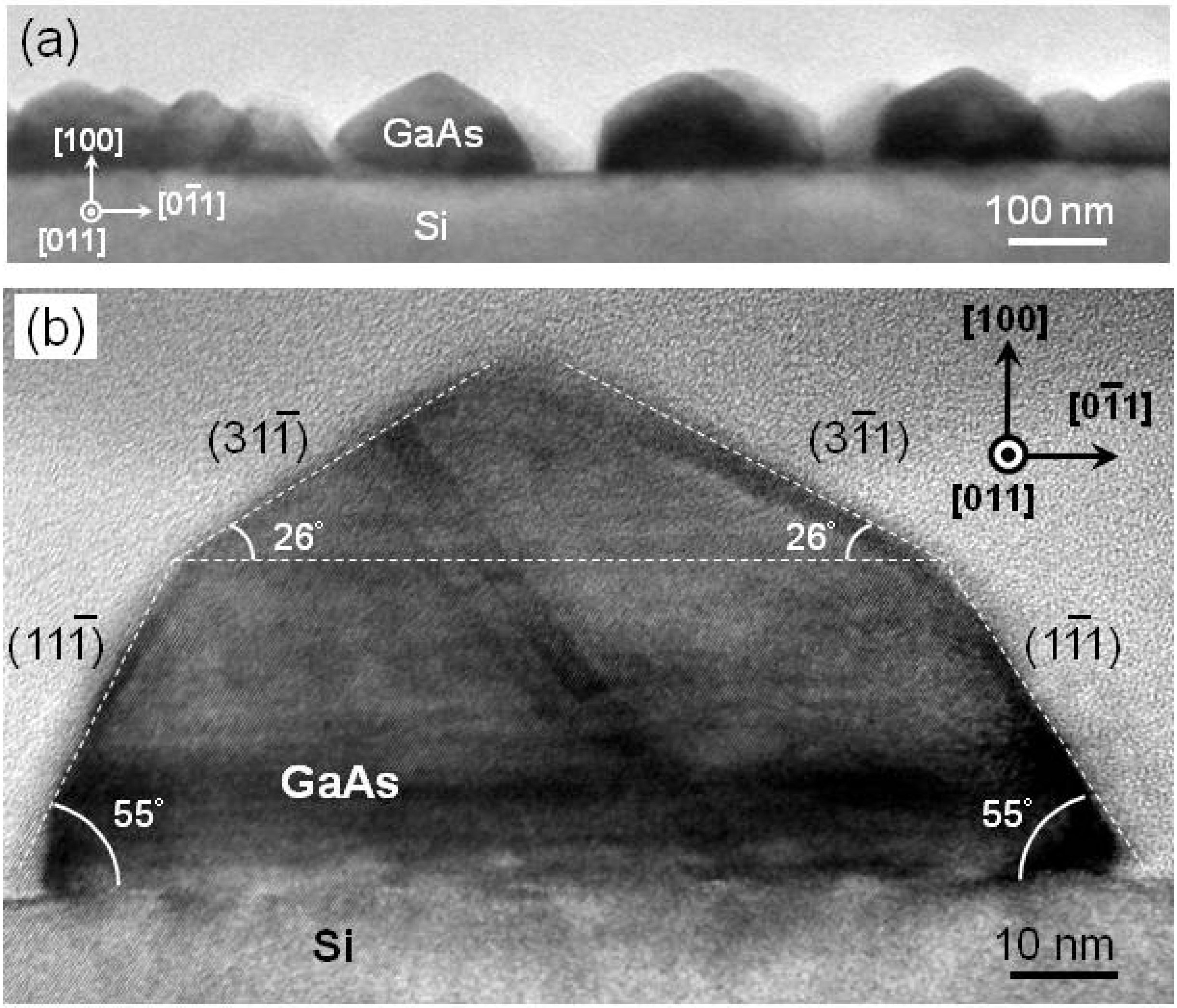

Fig. 3 H. Usui *et al.*