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Self-assembly of Ge quantum dots on Si(100)-2×1 by pulsed laser deposition

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Self-assembled Ge quantum dots are grown on Si(100)-2×1 by pulsed laser deposition. The growth is studied by *in situ* reflection high-energy electron diffraction and postdeposition atomic force microscopy. After the completion of the wetting layer, transient hut clusters, faceted by different planes, are observed. When the height of these clusters exceeded a certain value, the facets developed into {305} planes. Some of these huts become {305}-faceted pyramids as the film mean thickness was increased. With further thickness increase, dome clusters developed on the expense of these pyramids. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1949285]

Self-assembled Ge quantum dots (QD) grown on Si are used in fabricating several devices such as midinfrared photodetectors,[1–4] thermoelectric devices,[5] and enhanced performance Si solar cells.[6–8] From a basic physics point of view, Ge/Si is a model system to study the dynamics of the Stranski-Krastanow growth mode. Upon the completion of the wetting layer, which is estimated to be 4–6 monolayers (ML) ($1\ \mathrm{ML}=6.24\times10^{14}\ \mathrm{atoms/cm^2}$), three-dimensional nucleation starts by the formation of {105}-faceted "hut" or "pyramid" clusters. As the film coverage increases, multifaceted "domes," faceted by {113} and {102} planes, start to appear on the expense of the hut clusters. With further increase in thickness, large clusters "superdomes" start to appear. Ge QDs were grown on Si(100) by molecular beam epitaxy (MBE),[9–14] chemical vapor deposition (CVD),[15,16] and liquid phase epitaxy (LPE).[17,18] The shape of the initial islands was found to depend on the deposition technique as well as the deposition conditions. It is known that the physical properties of the QDs are controlled by their shape and size distribution. If Sb is used as a surfactant in the MBE growth of Ge/Si(100), the initial island shape changes from {105} faceted to {117} faceted.[19] If Ge is grown by liquid phase epitaxy, {115}-faceted islands are first observed instead of the {105}-faceted ones, and as the coverage increases, {111}-faceted pyramids are formed.[17,18] However, the growth dynamics of Ge on Si was not studied in the case of pulsed laser deposition (PLD).

PLD is a powerful deposition technique with several attractive features that can be particularly useful in the growth of quantum dot devices. Among its attractive features are the preservation of stoichiometry, the ease to grow multilayered films, the ability to grow thin films out of any material, the highly energetic deposited particles, and its periodic nature that allows for surface relaxation between pulses. The first two features enable the growth of multilayered devices of different materials or dopings without the need for residual gases or doping sources; just targets with the desired doping are used. In QD devices, tens of multilayers of doped QDs separated by a wetting layer are typically used. PLD is particularly suitable for the growth of these configurations. Using PLD, we have fabricated a Ge QD-based midinfrared photodetector that showed an absorption peak around 2 μm.[4]

In this Letter, we report on the growth of self-assembled Ge QD on reconstructed Si(100)-2×1 by PLD. Reflection high-energy electron diffraction (RHEED) is used to *in situ* monitor deposition and growth, while *ex situ* atomic force microscopy (AFM) is used to study the morphology of the grown quantum dots. We focus on the evolution of the Ge quantum dots and the change of their shapes at different film thicknesses. Small, elongated hut clusters are first formed, which transform into {305}-faceted ones, or large pyramids, as their size increases. After ~6 ML average thickness, dome clusters start to appear on the expense of the hut and pyramid clusters.

The growth is conducted in an ultrahigh vacuum chamber. The Si substrate is heated by direct current. The Ge target is mounted on a rotated holder with a variable rotation speed. Target rotation during PLD is necessary to minimize the formation of particulates. RHEED is used to monitor the deposition. A 15-keV electron gun is used for RHEED, while a phosphor screen displays the electron diffraction pattern, which is recorded by a charge-coupled devices (CCD) camera. Before loading into the vacuum, the Si(100) substrates are cleaned by chemical etching using a modification to the Shiraki method.[20] The vacuum system is then pumped down, baked at ~300 °C for at least 12 h, and flashed to 1100 °C in order for the 2×1 reconstruction to form. The chamber pressure is maintained $<1\times10^{-9}$ Torr. A Nd- yttrium aluminum garnet (YAG) laser (with a wavelength of 1064 nm, pulse width of 40 ns, and repetition rate of 10 Hz) is used to ablate the Ge target. The laser beam is focused on the rotating target with a spot size of 330 μm (measured at $1/e$ of the peak value).

A series of Ge samples is prepared under the same deposition conditions (substrate temperature of 400 °C, laser fluence of 0.24 J/cm^2, and a laser repetition rate of 10 Hz) but with different thicknesses. The thickness calibration is done in separate runs by means of a crystal thickness monitor placed at the target's location. Figure 1 shows the change in the RHEED pattern as a function of the film thickness. The Si(100)-2×1 diffraction pattern, Fig. 1(a), does not change during the first few seconds of deposition due to the epitaxial growth of the wetting layer as seen in Fig. 1(b) taken at ~3.3 ML. No change in the streak spacing is observed.

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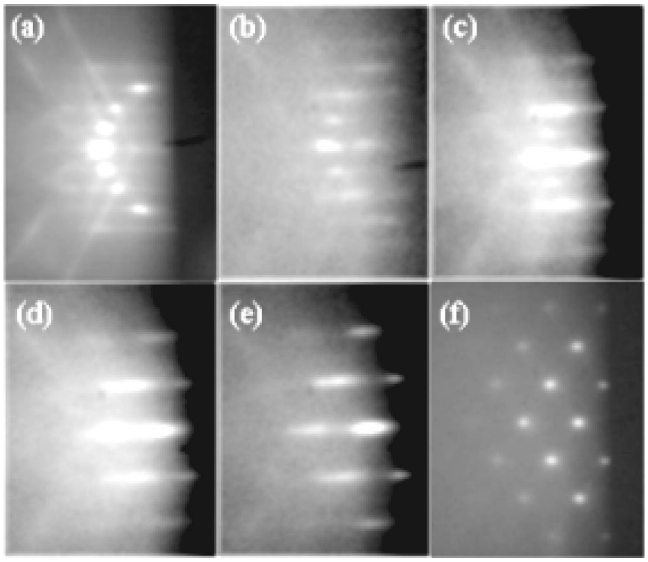

FIG. 1. RHEED patterns taken at different thicknesses for deposition at 400 °C, 23.5 J/cm^2, 10 Hz. (a) The substrate 2×1 reconstruction pattern is shown. (b) Growth starts epitaxially, as seen after the deposition of ~3.3 ML. (c) Around 4.1 ML, elongated transmission features with lines at the position of the second order strikes start to appear. (d) In the pattern at ~6 ML, the lines disappear while the elongation of the transmission features increases. As thickness increases, the transmission features become more well defined and the elongation decreases, (e) taken at ~9.3 ML. Later, (f) taken at ~13 ML, the transmission features become more round.

Around 4.1 ML, shown in Fig. 1(c), elongated transmission features with lines at the position of the second-order strikes start to appear. In MBE growth, this RHEED feature was shown to correspond to the formation of the {105}-faceted hut and pyramid clusters.[9,11,21] At ~6 ML, Fig. 1(d), the lines at the position of the second-order strikes disappear, while the elongated transmission features increase in intensity and elongation. As the film thickness is increased further, the transmission RHEED features split into well-defined features and their elongation starts to decrease, as shown in Fig. 1(e) taken at ~9.3 ML. Finally, around 13 ML, Fig. 1(f), the transmission features appear to be fully rounded. Such spotty transmission patterns, but with chevron lines due to faceting of the Ge clusters, were observed when multifaceted "macroislands" clusters were formed (domes and superdomes faceted by {113} and {102} planes).[9,22] We were not able to observe any chevron lines. The absence of such lines may be related to the lack of well-defined facets in the PLD-formed dome clusters.

Ex situ AFM is used to study the morphology and the shape of the quantum dots. Depending on the film thickness, three cluster shapes are observed; huts, pyramids, and domes. The hut clusters are seen to dominate at low film thicknesses with cluster sizes ranging between 90 and 400 nm, and heights ranging between 3 and 90 nm. Well-developed huts are facted by {305} planes, making angles of 31° with the (100) substrate. However, developing huts are seen to be facted by different "transient" planes making slopes with the (100) substrate that vary with the height of the cluster. Figure 2 shows two representative hut clusters formed after deposition of ~4 ML. The one on the left is a developing hut (with a major length of ~150 nm and a height of ~3.7 nm), while the one on the right is a developed one (with a major length of ~480 nm and a height of ~68 nm). The line profiles along the denoted *x* direction

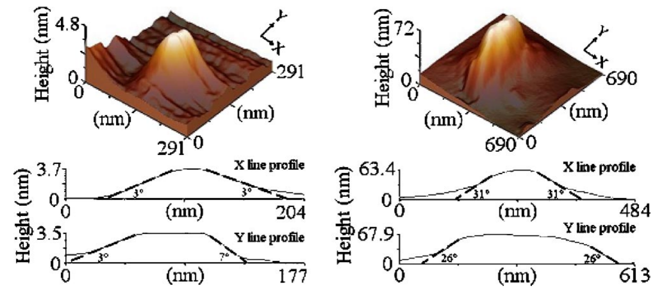

FIG. 2. (Color online) Hut clusters formed after the deposition of ~4 ML. (Left) A small developing cluster with line scans along the denoted *x* and *y* directions. (Right) A developed hut with line scans along the *x* and *y* directions. The line profile along the *x* direction shows that the hut is faceted by {305}.

show that the developing cluster is bounded by planes making ~3° with the (100) substrate, probably corresponding to {1 0 19}. The line scan in the same direction shows that the developed hut is faceted by {305} planes. Line profiles along the *y* direction show another faceting plane making ~7° with the substrate, probably corresponding to {108}, for the developing hut shown.

As the film thickness was increased, large pyramids in addition to the large huts are observed. These pyramids are slightly elongated. Figure 3(a) shows one of these large pyramids that form the minority of clusters at ~8 ML. The main faceting planes of such pyramids are also found to be {305}. The observation of the {305}-faceted huts and pyramids differs from the previous observations in the cases of MBE, CVD, and LPE. Thus, PLD produces cluster morphologies that can differ from those produced in other vapor phase deposition techniques.

With increased film thickness, the huts and pyramids transformed into large domes. Figure 3(b) shows a well-developed dome. Notice the smoothness of the domes, which could be seen as a continuous distribution of faceting planes. This morphology is consistent with the lack of observation of chevron lines in RHEED.[23] The chevron lines arise from the intersection of two diffraction patterns, each originating from one faceting plane.[21]

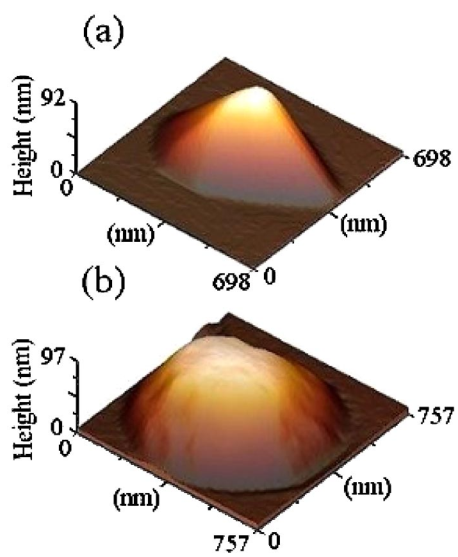

FIG. 3. (Color online) AFM images showing a (a) large pyramid and (b) a developed dome.

In summary, the growth of self-assembled Ge QDs on Si(100)-2×1 by PLD has been studied. We have focused on the evolution of the QDs and their shape change during growth. The growth is observed to start by the formation of elongated hut clusters faceted by planes that differ depending on the height of the cluster. The faceting planes become {305} when certain size is reached. At a larger film thickness, some large pyramids are also observed. With further deposition, large huts and pyramids are transformed into domes with smooth edges.

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[1]V. A. Egorov, G. É. Cirlin, A. A. Tonkikh, V. G. Talalaev, A. G. Makarov, N. N. Ledentosov, V. M. Ustinov, N. D. Zakharov, and P. Werner, Phys. Solid State **46**, 49 (2004).
[2]A. Elfving, G. V. Hansson, and W.-X. Ni, Physica E (Amsterdam) **16**, 528 (2003).
[3]M. Elkurdi, P. Boucaud, S. Sauvage, G. Fishman, O. Kermarrec, Y. Campidelli, D. Bensahel, G. Saint-Girons, G. Patriarche, and I. Sagnes, Physica E (Amsterdam) **16**, 450 (2003).
[4]M. S. Hegazy, T. F. Refaat, M. Nurul Abedin, H. E. Elsayed-Ali, Opt. Eng. Lett. 44, 059702 (2005).
[5]J. L. Liu, A. Khitun, K. L. Wang, T. Borca-Tasciuc, W. L. Liu, G. Chen, and D. P. Yu, J. Cryst. Growth **227–228**, 1111 (2001).
[6]A. Alguno, N. Usami, T. Ujihara, K. Fujiwara, G. Sazaki, K. Nakajima, and Y. Shiraki, Appl. Phys. Lett. **83**, 1258 (2003).
[7]H. Presting, J. Konle, H. Kibbel, and F. Banhart, Physica E (Amsterdam) **14**, 249 (2002).
[8]J. Knole, H. Presting, H. Kibbel, Physica E (Amsterdam) **16**, 596 (2003).
[9]V. Cimalla, K. Zekentes, and N. Vouroutzis, Mater. Sci. Eng., B **88**, 186 (2002).
[10]I. Goldfarb, P. T. Hayden, J. H. G. Owen, and G. A. D. Briggs, Phys. Rev. Lett. **78**, 3959 (1997).
[11]Y.-W. Mo, D. E. Savage, B. S. Swartzentruger, M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).
[12]A. I. Nikiforov, V. A. Cherepanov, O. P. Pchelyakov, A. V. Dvurechenskii, A. I. Yakimov, Thin Solid Films **380**, 158 (2000).
[13]O. P. Pchelyakov, V. A. Markov, A. I. Nikiforov, and L. V. Sokolov, Thin Solid Films **306**, 299 (1997).
[14]J. A. Floro, E. Chason, L. B. Freund, R. D. Twesten, R. Q. Hwang, and G. A. Lucadamo, Phys. Rev. B **59**, 1990 (1999).
[15]T. I. Kamins, E. C. Carr, R. S. Williams, and S. J. Rosner, J. Appl. Phys. **81**, 211 (1997).
[16]P. S. Chen, Z. Pei, Y. H. Peng, S. W. Lee, and M.-J. Tsai, Mater. Sci. Eng., B **108**, 213 (2004).
[17]M. Schmidbauer, T. Wiebach, H. Raidt, M. Hanke, R. Köhler, and H. Wawre, Phys. Rev. B **58**, 10523 (1998).
[18]W. Dorsch, S. Christiansen, M. Albrecht, P. O. Hansson, E. Bauser, and H. P. Strunk, Surf. Sci. **331–333**, 896 (1995).
[19]M. Horn-von Hoegen, B. H. Müller, A. Al Falou, and M. Henzler, Phys. Rev. Lett. **71**, 3170 (1993).
[20]M. S. Hegazy and H. E. Elsayed-Ali, J. Vac. Sci. Technol. A **20**, 2068 (2002).
[21]C. E. Aumann, Y.-W. Mo, and M. G. Lagally, Appl. Phys. Lett. **59**, 1061 (1991).
[22]J. W. Lee, D. Schuh, M. Bichler, and G. Abstreiter, Appl. Surf. Sci. **228**, 306 (2004).
[23]M. D. Kim, T. W. Kim, and Y. D. Woo, J. Cryst. Growth **265**, 41 (2004).