APPLIED PHYSICS LETTERS VOLUME 74, NUMBER 7 15 FEBRUARY 1999

In situ scanning tunneling microscopy study of C-induced Ge quantum dot formation on Si(100)

O. Leifeld,[a] E. Müller, and D. Grützmacher
Micro- and Nanostructures Laboratory, Paul-Scherrer-Institute, CH-5232 Villigen-PSI, Switzerland

B. Müller[b] and K. Kern
*Institut de Physique Expérimentale, Ecole Polytechnique Fédérale de Lausanne,
CH-1015 Lausanne, Switzerland*

(Received 15 May 1998; accepted for publication 16 December 1998)

Deposition of submonolayer coverages of C on Si(100) prior to Ge growth leads to the formation of Ge quantum dots below the critical thickness for Ge islanding on bare Si(100). *In situ* scanning tunneling microscopy reveals a high density of irregularly shaped islands for Ge coverages from 2.5 to 4 ML. Island sizes are broadly distributed between 10 and 25 nm. Keeping the C coverage constant and increasing the Ge coverage from 2.5 to 4 ML, the islands increase in height but their density remains constant ($\sim 10^{11}\,\mathrm{cm}^{-2}$). At a Ge coverage of 5.8 ML, formation of larger (105)-faceted islands is observed. Their density is reduced by a factor of 4 compared to smaller Ge coverages. Transmission electron microscopy shows that the nonfaceted islands are preserved after Si capping. © *1999 American Institute of Physics.* [S0003-6951(99)03107-1]

Integration of optical components onto a Si-based chip allows additional functionality and realization of new concepts. Consequently, various approaches to the engineering of optically active Si components have been discussed and are still under debate.[1] Among these, quantum dots have gained a lot of attention in recent years due to the discovery that structures containing Si particles of a few nanometers diameter can luminesce intensively.[2,3] However, in these structures the Si particles are surrounded by silicon oxide or nitride films leading to serious problems in contacting these films and in achieving optical confinement. Embedding a low-band-gap material like Ge into Si, it should be possible to overcome these difficulties. Deposition of Ge on Si(100) surfaces leads to a strain-induced spontaneous formation of hut clusters as soon as the Ge film exceeds a critical thickness of 3–4 ML.[4] Dot diameters can be reduced by lowering the growth temperature. However, at low temperatures, where the smallest dots can be fabricated,[5] the material quality suffers and no significant dot luminescence is detected. Recently, it has been shown that Ge islands with diameters as small as 10 nm can be produced on Si(100) surfaces precoated with a submonolayer of carbon at a growth temperature of 550 °C.[6] These islands show rather intense photoluminescence (PL)[6] and may have some potential in Si-based optoelectronics.

In this letter we focus our attention on the formation of these C-induced Ge dots on Si(100) studied *in situ* by ultrahigh-vacuum scanning tunneling microscopy (UHV-STM). The results are supported by transmission electron microscopy (TEM) of C-induced Ge dots capped with Si.

The samples were prepared by molecular beam epitaxy (MBE) using e-beam evaporation sources for Si and Ge. Carbon was sublimated from a graphite filament. The C coverage was calibrated by secondary ion mass spectroscopy (SIMS). The 4 in. Si substrates were wet-chemically cleaned and baked at 950 °C in the MBE chamber, leading to a well-defined 2×1-reconstructed surface as verified by reflection high-energy electron diffraction (RHEED) and STM. After a 200 nm wide Si buffer layer, 0.11 monolayers (ML) C, and subsequently, the Ge dot layer were deposited at a substrate temperature of 550 °C and a Ge growth rate of 0.16 ML/s. The 4 in. Si wafers were then transferred from the MBE into the STM chamber without braking UHV.[7] For the TEM investigations 1–3 C-induced Ge dot layers separated by 150 nm wide Si barriers were grown. Dot layers grown directly on top of the Si buffer and on top of the layered structure show no difference in the STM analysis.

Figure 1 compares the STM images obtained from (a) 2.5, (b) 4, and (c) 5.8 ML of Ge on a Si(100) surface precovered by 0.11 ML C. For Ge coverages up to 4 ML [Figs. 1(a) and 1(b)], basically a very rough three-dimensional (3D) growth front is observed. It consists of bumps (islands) and voids with random shape, which are formed from stacks of single atom high Ge terraces, as can be seen in the inset of Fig. 1(b). No formation of distinct crystal facets is detected in this stage. The growth front comprises already up to 7 atomic layers for 2.5 ML deposited Ge and up to 12 layers in the case of a 4 ML Ge deposit. It is remarkable, that 3D Ge islands are already observed after the deposition of 2.5 ML of Ge on Si(100) surfaces covered with fractions of a ML of C. In contrast, Ge on bare Si(100)-2×1 surfaces forms 3D islands (faceted hut clusters) only after the Ge thickness exceeds the critical thickness of 3–4 ML;[8] in this case, the driving force for the island formation is the strain relief of the Ge layer.[9] Since the submonolayer C coverage should compensate the overall amount of strain in the Ge layer, strain relief appears not to be the dominating force for the early onset of island formation. However, the submonolayer C coverage that is not uniformly distributed on the surface[10] may lead to an undulating surface strain field even before Ge deposition. It is likely that the Ge island formation is driven

---

[a] Also with the Institut de Physique Expérimentale, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland. Electronic mail: oliver.leifeld@psi.ch

[b] Present address: Surface- and Joining Technology, Swiss Federal Laboratories for Materials Testing and Research, CH-8600 Dübendorf, Switzerland.

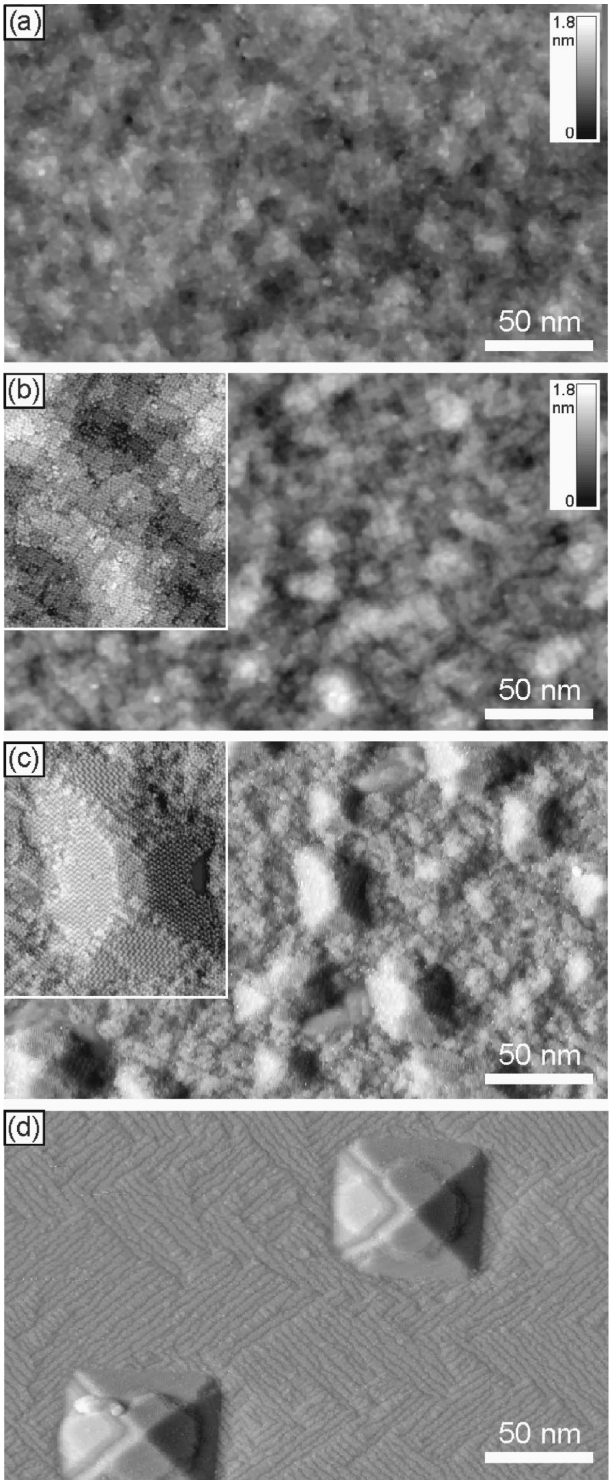

FIG. 1. Comparison of surface morphologies of different Ge coverages on Si(100) precovered with 0.11 ML C and on bare Si. For (a) 2.5 ML and (b) 4 ML Ge coverage irregularly shaped islands with stepped terraces are obtained on C-precovered Si. Their height increases with the Ge coverage. At 5.8 ML Ge {105} faceting of the Ge island occurs at the expense of island density, as depicted in (c). The size and height (about 3 nm) of the faceted island exceeds that of the stepped ones. Without C predeposition (d) at 5.8 ML Ge a low density of large hut clusters is obtained on top of a smooth two-dimensional wetting layer. [(c) and (d) have been taken in derivative imaging mode.]

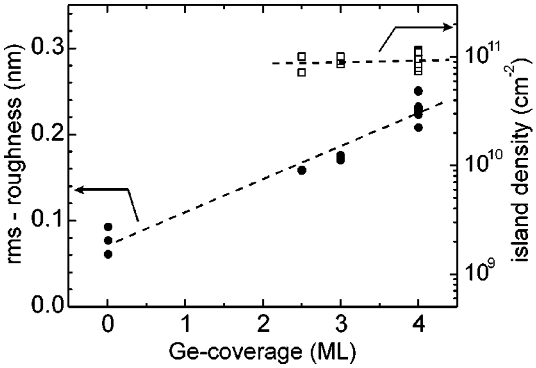

FIG. 2. Root-mean-square roughness (rms) and island density as a function of Ge coverage as deduced from STM images. The roughness increases monotonically while the island density remains constant.

by compensation of this strain field. Furthermore, the surface roughness introduced by the C deposition[11] and, hence, reduced diffusion length for Ge adatoms as well as the strong repulsive forces between C and Ge,[12] may contribute to the island formation in the low coverage range of 2–3 Ge ML.

Increasing the Ge coverage beyond a critical thickness leads to the spontaneous formation of faceted islands also on the C-covered Si surfaces [Fig. 1(c)]. At the same time, the surface between the islands smoothens and the irregular islands obtained at lower Ge coverage die out. The dominating facets are (105) side facets and a flat (100) top facet. Quadratic as well as rectangular shapes are found. Some clusters appear to be coalesced from islands created at neighboring nucleation centers. On the top (100) facet buckled Ge dimers with missing dimer rows are observed. Since these rows of missing dimers are an effective way for stress relaxation for the compressively strained Ge on Si,[13] this indicates that the islands are still strained at their apex. The similarities with the well-known strain-driven Ge ''hut clusters''[14] observed on bare Si surfaces [Fig. 1(d)] let us conclude that these islands are formed by a similar mechanism. However, the increased surface roughness leads to a reduction of the diffusion length of the Ge atoms and to a higher nucleation density, which in turn results in a higher density and smaller island size compared to islands grown on bare Si surfaces at the same temperature.

The density of the faceted islands on C precovered surfaces is about a factor of 4 lower than that of the irregular islands at lower Ge coverages but is still larger by more than an order of magnitude compared to the density of islands formed by 5.8 ML of Ge on bare Si surfaces [Fig. 1(d)]. The density of the irregular C-induced islands is determined to be about $10^{11}\,\mathrm{cm}^{-2}$. As illustrated in Fig. 2 for 2.5–4 ML of Ge, the island density appears not to depend on the actual Ge coverage, when counting all islands higher than the mean height in the STM images plus the root-mean-square (rms) roughness. Figure 2 also shows the dependence of the rms roughness obtained from the STM scans as a function of the Ge coverage. The rms roughness increases monotonously with the amount of Ge deposited from 0.08 nm, detected after the deposition of 0.1 ML C without Ge, to 0.22 ± 0.03 nm after additional deposition of 4 ML Ge. Figure 3 depicts the island height distribution of samples with Ge coverages of 3 ML (dark gray) and 4 ML (light gray). Island heights are given in the number of ML with respect to the level of the lowest voids in the images that are defined as zero. Only islands are counted that lie above a certain threshold value (5 ML, in this case, see the horizontal line in the

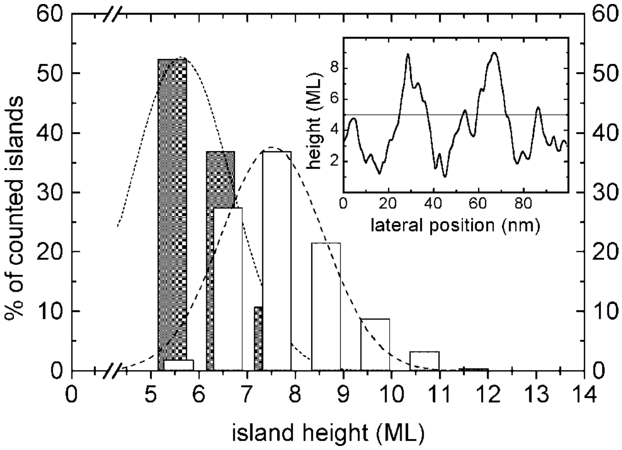

FIG. 3. Island height distribution for nominal Ge coverages of 3 ML (dark gray) and 4 ML (light gray). Mean island heights shift to higher values with increasing Ge coverage. The inset shows a typical line scan with the threshold for island count at 5 ML.

inset of Fig. 3) by a gray value discrimination and particle count algorithm. Counting below this threshold is meaningless, since around the mean value of heights between bumps and voids in the images one obtains only large areas formed by interconnected island bases from adjacent bumps. The average height of the islands shifts from 6.1 to 7.6 ML for the samples with 3 and 4 ML deposited material, respectively. At the same time the height distribution becomes broader with increasing thickness, again showing that the growth front contains an increasing number of atomic layers, in agreement with the increasing rms value.

A detailed analysis of an island diameter distribution on this randomly rugged surface does not seem to be useful, since the definition of the island bases and, hence, of the island diameters, always remains somewhat arbitrary. Nevertheless, one can state that the size distribution is quite broad with the irregularly shaped bumps having diameters ranging from less than 10 nm up to 25 nm.

The growth of the islands in the regime of 2.5–4 ML Ge can be clearly seen in TEM cross sections of capped C-induced dot layers. Figure 4(a) shows a C-induced island layer (dark) of 2.5 ML Ge embedded in Si (light). The islands are quite shallow, with an approximate width of 10–20 nm and a height of 14 atomic layers. In the case of 4 ML Ge [Fig. 4(b)], the islands are more pronounced, in particular, the height has grown to about 18 atomic layers. Like the STM images of the surface dots, the TEM data of the corresponding islands embedded in Si show no well-defined facets. An overall change of island shape due to capping with Si, which has been demonstrated for initially (105)-faceted dots,[14] cannot be detected for the initially nonfaceted islands. Still, the islands are undoubtedly affected by the overgrowth with Si; the slightly larger island height observed by TEM compared to the STM data might be attributed to the segregation of Ge during capping. One should keep in mind that capping is always necessary when studying optical properties of dot layers in order to prevent recombination at surface states of the nanostructures.

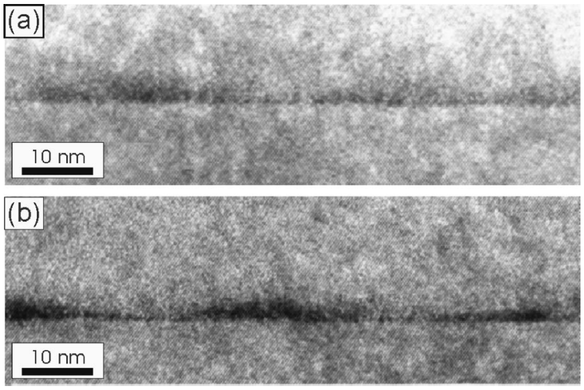

FIG. 4. TEM cross-sectional images of overgrown samples with (a) 2.5 and (b) 4 ML Ge coverage. Islands become higher and more pronounced with increasing Ge coverage.

In the TEM images the islands seem to be connected by a narrow dark line. However, this dark line is not very homogeneous and, hence, is not necessarily a hint for a Ge wetting layer. It can either be just the projection of the irregular depressions in the dot layer across the TEM foil, or it can be the Si–C alloy or both. A detailed study of the Ge nucleation in the submonolayer regime on the C-precovered Si(100) surface would be helpful to clarify this point. The TEM shows no extended defects like dislocations in the grown layer although the crystal contains carbon in the area of the dot layer. It is worth mentioning that we have observed intense photoluminescence for the capped C-induced dot layers[11] in perfect agreement with Ref. 6.

In conclusion, the formation of C-induced Ge dots has been *in situ* investigated by STM. At a carbon precoverage of 0.11 ML, small irregularly shaped islands with a constant density of $10^{11}\,\mathrm{cm}^{-2}$ are observed for 2.5–4 ML Ge. They show no facets. Dot height and, hence, rms growth front roughness increase with Ge layer thickness, indicating pure three-dimensional growth. The existence of a Ge wetting layer cannot be undoubtedly derived from our data. Faceting occurs at about 5.8 ML Ge at the expense of quantum dot density. TEM investigations show no essential change of island shapes after capping for the nonfaceted dots, although some Ge segregation takes place.

[1] S. S. Iyer and Y.-H. Xie, Science **260**, 40 (1993).

[2] S. Schuppler, S. L. Friedmann, M. A. Marcus, D. L. Adler, Y.-H. Xie, F. M. Ross, Y. J. Chabal, T. D. Harris, L. E. Brus, W. L. Brown, E. E Chaban, P. F. Szajowski, S. B. Christman, and P. H. Citrin, Phys. Rev. B **52**, 4910 (1995).

[3] E. F. Steigmeier, R. Morf, D. Grützmacher, H. Auderset, B. Delley, and R. Wissicken, Appl. Phys. Lett. **69**, 4165 (1996).

[4] Y.-W. Mo, D. E. Savage, B. S. Swartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990).

[5] G. Abstreiter, P. Schittenhelm, C. Engel, E. Silveira, A. Zrenner, D. Meertens, and W. Jäger, Semicond. Sci. Technol. **11**, 1521 (1996).

[6] O. G. Schmidt, C. Lange, K. Eberl, O. Kienzle, and F. Ernst, Appl. Phys. Lett. **71**, 2340 (1997).

[7] O. Leifeld, B. Müller, D. A. Grützmacher, and K. Kern, Appl. Phys. A: Mater. Sci. Process. **66A**, S993 (1998).

[8] D. J. Eaglesham and M. Cerullo, Phys. Rev. Lett. **64**, 1943 (1990).

[9] B. J. Spencer and J. Tersoff, Mater. Res. Soc. Symp. Proc., 283 (1996).

[10] O. Leifeld, D. Grützmacher, B. Müller, K. Kern, E. Kaxiras, and P. Kelires, Phys. Rev. Lett. (to be published).

[11] O. Leifeld, R. Hartmann, E. Müller, B. Müller, K. Kern, and D. Grützmacher, Nanotechnology (to be published).

[12] P. C. Kelires and E. Kaxiras, J. Vac. Sci. Technol. B **16**, 1687 (1998).

[13] U. Köhler, O. Jusko, G. Pietsch, B. Müller, and M. Henzler, Surf. Sci. **248**, 321 (1991).

[14] E. Mateeva, P. Sutter, J. C. Bean, and M. G. Lagally, Appl. Phys. Lett. **71**, 3233 (1997).