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Atomic force microscopy study of self-organized Ge islands grown on Si(100) by low pressure chemical vapor deposition

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In this letter, we present an atomic-force-microscopy investigation of the Stranski–Krastanov growth of Ge on Si(100). Upon increasing the base width of the islands, two morphology transitions are found. The first transition occurs at a base width of ~50–60 nm and marks the evolution from few-monolayer-thick terraces to square-base pyramidal islands. In the second transition, which takes place when the base width exceeds ~300 nm, the island shape changes from square base pyramids to tetragonal truncated pyramids. Both transitions are brought about by the need for the system to minimize the elastic energy. © *1997 American Institute of Physics.* [S0003-6951(97)02104-9]

Recently the physical properties of quantum confined semiconductor heterostructures have attracted wide attention both from the experimental and theoretical point of view.[1] The main reason is the demonstrated capability of tailoring the optical and electronic properties of such heterostructures by controlling the growth at the atomic scale. In the case of the fourth group semiconductors, we deal with indirect band-gap materials exhibiting poor luminescence efficiency. It would be attractive, therefore, to enhance the carrier radiative recombination efficiency by means of quantum confinement effects, in order to realize Si-based optoelectronic devices. In particular, quantum dots (QDs) are expected to give a significant increase of the quantum efficiency.[2]

In the literature, a variety of recipes for realizing QDs has been reported.[3–7] Among them, the self-organization of islands seems to be one of the most promising ways to produce zero dimensional quantum confined systems. A possible way to achieve self-organization is to take advantage of the spontaneous formation of clusters under the Stranski–Krastanov (SK) growth conditions. In this way, it is possible to produce QDs of good quality with a narrow distribution of sizes and with an increase of the optical quantum efficiency.[8] These promising experimental findings require detailed knowledge of the SK growth dynamics.

In this letter, we report an atomic force microscopy (AFM) study of the evolution of SK islands in Ge grown by UHV low pressure chemical vapor deposition (LPCVD) on Si(100). Atomic force microscopy measurements were performed in contact mode with a Park Scientific Instruments CP microscope equipped with a high aspect ratio conical (80° sidewall angle) Ultralever™ tips with 5 nm minimum radius.[9] We have done standard tests and scan calibrations on freshly cleaved mica in order to obtain atomic imaging and to avoid tip imaging misleading information.

The Si(100) substrates used in this study were treated with an *ex situ* chemical etch in and immediately loaded into a UHV growth chamber, whose basic pressure was in the low 10^{-10} Torr range, and then heated up to 950 °C in H_2 atmosphere. As determined by x-ray photoemission spectroscopy, reflection high-energy electron diffraction, and AFM the surfaces were clean (C and O contamination below 0.1%), 2×1 reconstructed and flat (RMS roughness ~1 ML). Moreover, photoluminescence measurements showed that radiative dislocations were absent.

The Ge films were deposited by LPCVD from high purity germane without carrier gas at pressures, P_{dep}, in the range 0.5–2 mTorr. The samples considered in the present investigation grew according to the Stranski–Krastanov mode. Under our experimental conditions, this growth mode was observed for deposition temperatures, T_{dep}, in the range 400–650 °C. For $T_{dep}<400$ °C, a layer-by-layer Frank–Van der Merwe growth mode occurred, while for $T_{dep}>650$°C the samples grew according to the Volmer–Weber mode.

In Fig. 1, we show AFM topographies of samples grown at $T_{dep}=600$ °C and $P_{dep}=1$ mTorr for four different deposition times. We see in Fig. 1(a) that 0.5 min deposition time is enough to develop on top of the wetting layer a large number of small islands characterized by a narrow distribution of sizes. Longer deposition times [Figs. 1(b)–(d)] bring about an increase of the average island size, a spreading of the size distribution, and a slight decrease of the number of the islands in agreement with recent theoretical predictions.[10–12] These observations suggest that the new material deposited contributes to enlarge the existing islands. In such conditions, the deposition rate was estimated to correspond to a Ge effective thickness of 3 nm per min.

A detailed analysis of the size distributions and shapes shows that the island evolution follows a well defined and regular pattern. Concerning the size and shape evolution, this can be illustrated with reference to Fig. 2, where the island height, *h*, as a function of the square root of the base area, i.e., the base width *b*, is plotted for different growth times. We see [points with $b<\sim50$ nm and negligible *h* in Figs. 2(a) and 2(b)] that at the initial stage, the island heights are very small as compared to the corresponding bases. Accordingly, the islands should be considered, more properly, as few-monolayer-thick terraces. These terraces expand laterally without an appreciable vertical growth until they reach a critical value b_c of 50–60 nm for the base width.

At the critical width b_c, a vertical growth begins until

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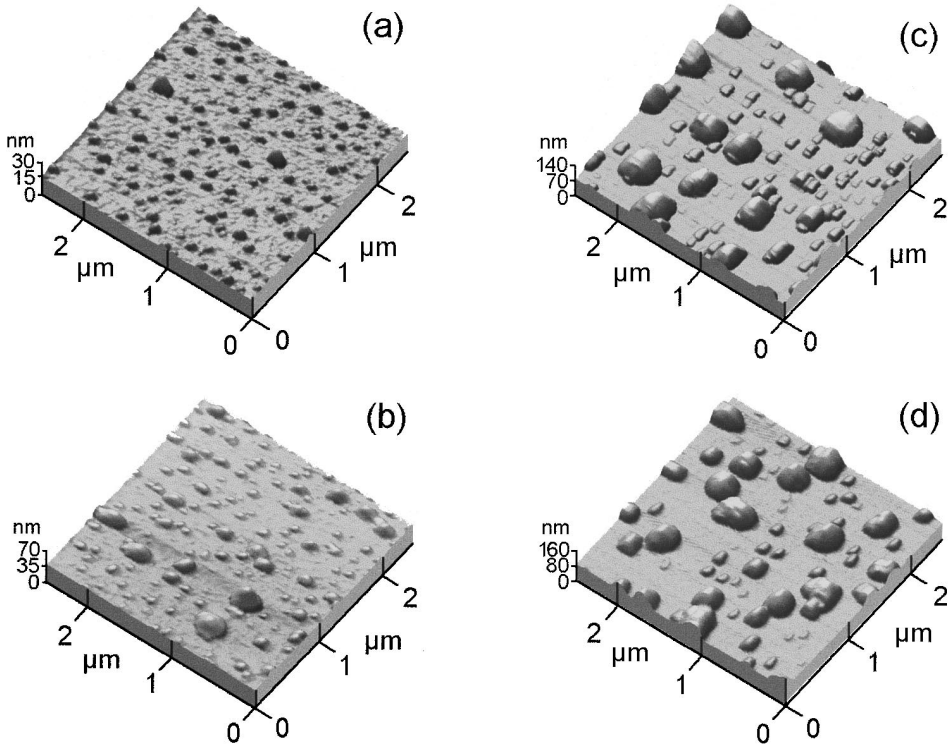

FIG. 1. AFM topographies of four samples grown for different deposition times: (a) 0.5, (b) 1, (c) 3, and (d) 4 min. Note the variation of the vertical scale from (a) to (d).

the terraces become islands with an aspect ratio $h/b \sim 0.25$. These islands now have the shape of square-base pyramids with {311} facets. The direct estimation of the facet orientation was performed by calculating the gradient in each point of the 256×256 mesh used to acquire AFM topographies. From this analysis, we found that the facet angle increases from a value close to zero for $b < b_c$ to $\vartheta \sim 25$ deg for $b > b_c$. This value is very close to that between the {311} and the {100} planes, i.e., 25.2 deg. It is worth noting that the aspect ratio of an ideal square pyramid with {311} facets is 0.24, in good agreement with our experimental finding of $h/b = \sim 0.25$. It is worth noticing that the {311} facets were evaluated[13] to exhibit the minimum surface energy for the growth of Ge on Si(100). The base edges of the islands are parallel to the [011] and [011] directions. These orientations were determined by referring to the cleaving directions of the Si(100) substrates. Possible artifacts on the AFM images due to the raster scan direction were ruled out by changing the scanning direction relative to the sample orientation.

It has been pointed out[14,15] that the island formation results from the possibility to relax laterally for the lattice planes that are compressed in a coherently grown planar film. In this way, the elastic energy is reduced. Therefore, the above experimental observations show that $b_c = 50-60$ nm represents the critical width for the planar growth on the wetting layer under the growing conditions used in the present work. For $b > b_c$, the new Ge atoms impinging on the terrace find energetically more favorable to form successive layers at whose periphery the lateral lattice compression can relax than to expand the bottom terrace itself. The evolution we are observing is in good agreement with the theoretical predictions of Tersoff and Tromp,[10] who found that,

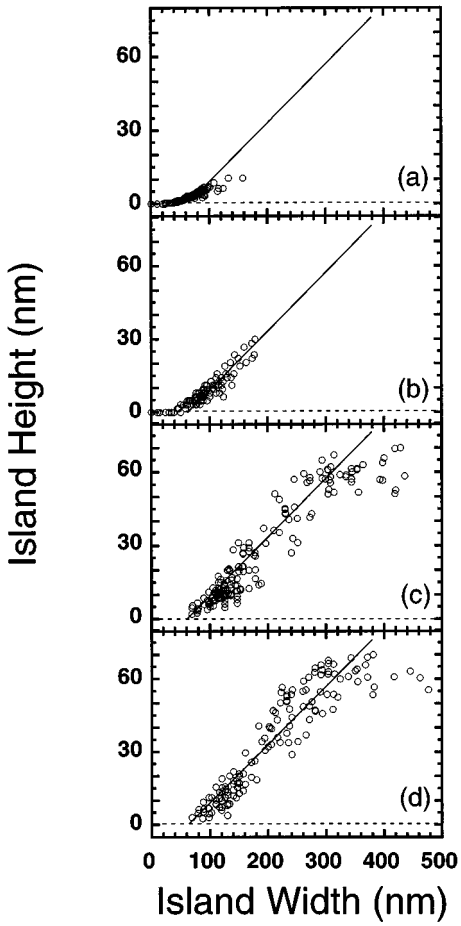

FIG. 2. Island height h vs base-width b for the samples shown in Fig. 1. The continuous line shows the $h/b = 0.24$ linear dependence, i.e., the growth according to a fixed 0.24 aspect ratio.

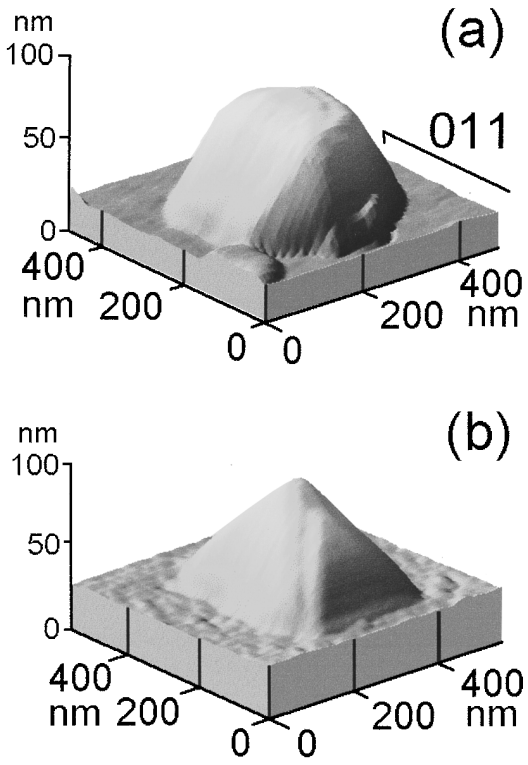

FIG. 3. (a) View of a typical island with the shape of tetragonal truncated pyramid with rectangular base (h=60 nm and b=320 nm). (b) View of a square-base pyramidal island (h=55 nm and b=300 nm) on which a dislocation has been just inserted, forming a new growing ledge. The vertical magnification, shading and azimuth are the same for both images. The scan direction is aligned in the [011] sample direction.

as the islands grow, they become triangular in cross section. Following their model, we find that, in our case, the critical width for the development of pyramidal islands is $b_c \sim 40$ nm.

For $b > b_c$, the islands increase in size without varying their shape, keeping fixed the aspect ratio at $h/b \sim 0.25$. This is illustrated in Figs. 2(b)–2(d) where a linear relationship between h and b is evident for values of b in the range 70–300 nm. Furthermore, the island distribution is quite narrow with $\Delta b/b_{\text{mean}} \sim 0.3$, where Δb is the standard deviation of the Gaussian distribution of the base widths and b_{mean} their mean value. These data show unambiguously that the island growth proceeds in the ledge-by-ledge mode as recently suggested.[15]

The increase of the island width beyond ~300 nm brings about a variation in the shape and a change of the growth mode. Referring to Figs. 2(c) and 2(d), we see that the island height tends to a limiting value $h_c \sim 55$ nm. The island shape becomes that of truncated pyramids with rectangular base, as shown in Fig. 3(a). Concomitantly, there is an increase in the width of their size distribution, becoming $\Delta b/b_{\text{mean}} \sim 0.5$. Thus, a second morphology transition occurs at $b \sim 300$ nm. We believe that this lateral growth is originated by the insertion of dislocations in order to relax the stress accumulated in the islands. Indeed, we have observed that at the beginning of the lateral growth, i.e., when h and b are close to the values at the transition point, the islands exhibit a shape like that reported in Fig. 3(b). A triangular growing front, 20 nm thick, and oriented on the (311) direction, is clearly visible on one side of the pyramidal island. This observation fits nicely within the growth cycle model recently proposed,[16] which the relaxation of the strain in the islands occurs by the insertion of dislocations that nucleate at the edge of the islands themselves, becoming a preferential site for the attachment of Ge. In particular, every time a dislocation is created, the island grows by ~20 nm in the direction of the strain relief. We conclude, therefore, that the growing fronts we have observed are originated by the insertion of dislocations. Moreover, the growth of the front occurs in a ledge-by-ledge mode from the base upwards in order to minimize the interaction between the surface steps.[15] After the insertion of a certain number of dislocations, the typical shape of the islands becomes the truncated pyramid reported in Fig. 3(a) with {311} lateral facets and a (100) top facet. The appearance of the (100) top facet confirms the insertion of dislocations.[17] We point out that the critical height at which the morphological transition occurs is in good agreement with the critical value of 50 nm for the maximum height of dislocation free SK islands found by Eaglesham and Cerullo.[14] Under these conditions, the subsequent lateral increase of the islands is much faster than the vertical one, so that h remains nearly constant. Moreover, the lateral facets orientation remains {311}.

In conclusion, we have analyzed the evolution of the SK Ge islanding on Si(100). We demonstrated that two morphology transitions occurred upon increasing the base width of the islands. The first transition marks the evolution from few-monolayer-thick terraces to square-base pyramidal islands. In the second transition the island shape changes from square-base pyramids to tetragonal truncated pyramids. Both transitions are brought about by the need for the system to minimize the elastic energy.

[1] F. Capasso and S. Datta, Phys. Today, 74 (1990).
[2] T. Takahagara and K. Takeda, Phys. Rev. B **46**, 15 578 (1992).
[3] J. R. Heath, J. J. Shiang, and A. P. Alivisatos, J. Chem. Phys. **101**, 1607 (1994).
[4] J. Brunner, T. S. Rupp, H. Gossner, R. Ritter, I. Eisele, and G. Abstreiter, Appl. Phys. Lett. **64**, 994 (1994).
[5] N. Usami, T. Mine, S. Fukatsu, and Y. Shiraki, Appl. Phys. Lett. **64**, 1126 (1994).
[6] H. Sunamura, N. Usami, S. Fukatsu, and Y. Shiraki, Appl. Phys. Lett. **66**, 3025 (1995).
[7] R. Apetz, L. Vescan, A. Hartmann, C. Dieker, and H. Lüth, Appl. Phys. Lett. **66**, 445 (1995).
[8] E. Palange, G. Capellini, L. Di Gaspare, and F. Evangelisti, Appl. Phys. Lett. **68**, 2982 (1996).
[9] The tip sidewall angle of 80° ensures that convolution problems could be important only for structures having growing angles $\vartheta \geqslant 70°$.
[10] J. Tersoff and R. M. Tromp, Phys. Rev. Lett. **70**, 2782 (1993).
[11] C. Priester and M. Lannoo, Phys. Rev. Lett. **75**, 93 (1995).
[12] A. L. Barabási (unpublished).
[13] D. J. Eaglesham, F. C. Unterwald, and D. C. Jacobson, Phys. Rev. Lett. **70**, 966 (1993).
[14] D. J. Eaglesham and M. Cerullo, Phys. Rev. Lett. **64**, 1943 (1990).
[15] D. E. Jesson, K. M. Chen, and S. J. Pennycook, Mater. Res. Bull. **21**, 31 (1996).
[16] F. K. Le Goues, M. C. Reuter, J. Tersoff, M. Hammar, and R. M. Tromp, Phys. Rev. Lett. **73**, 300 (1994); M. Hammar, F. K. Le Goues, J. Tersoff, M. C. Reuter, and R. M. Tromp, Surf. Sci. **349**, 129 (1996).
[17] M. A. Lutz, R. M. Feenstra, P. M. Mooney, J. Tersoff, and J. O. Chu, Surf. Sci. **316**, L1075 (1994).