Cyclic Growth of Strain-Relaxed Islands

F. K. LeGoues, M. C. Reuter, J. Tersoff, M. Hammar, and R. M. Tromp
IBM Research Division, T. J. Watson Research Center, Yorktown Heights, New York 10598
(Received 8 March 1994)

Growth of Ge islands on Si(001) is observed in real time with high spatial resolution, using UHV transmission electron microscopy. We are able to monitor the formation of successive strain-relieving dislocations. The shape of the island oscillates as it grows, with each cycle corresponding to the introduction of one dislocation. Such growth cycles are shown to be a general feature of the growth of strain-relaxed islands, occurring even in equilibrium.

PACS numbers: 68.55.Jk, 68.35.Fx, 68.35.Ja

The growth and relaxation of strained layers are classic issues which have become critical in microelectronics [1]. The crucial problem is to understand and control the formation of strain-relieving dislocations. Transmission electron microscopy (TEM) has been the technique of choice to determine the structure, nature, and location of the dislocation network, in part because it can probe the microstructure of an interface through a thick overlayer with high spatial resolution. However, most TEM experiments are limited to a "post mortem" analysis, wherein the sample is removed and observed at different stages of growth. It is then sometimes possible to infer the mechanism and kinetics of growth and relaxation [2, 3].

This approach has several limitations. First, in order to obtain a detailed description of the growth process, many samples have to be investigated, at small increments of thickness. Second, this method only catches successive "snapshots," missing any phenomena happening faster than the intervening time interval. Third, it is necessary to stop growth, cool the sample, and prepare it for observation, which may introduce artifacts. Finally, dislocations interact strongly with each other, and it is often impossible to infer a formation mechanism from the final tangled network.

Ideally one would like to study the thin film as it grows, observing the nucleation of each dislocation before it interacts with neighboring ones. *In situ* techniques such as UHV scanning electron microscopy [4] and low energy electron microscopy [5] can provide real-time images of growth. But these techniques cannot match TEM for the combination of high spatial resolution with the ability to penetrate a thickness of material (which is crucial for interface studies). Early work by Pashley *et al.* [6] demonstrated the advantages of *in situ* TEM (non-UHV). They studied the growth of metals, and discovered a remarkable "liquidlike" behavior of the coalescence of islands, which could only be observed by an *in situ* technique. More recent work by Ross and Gibson [7], though directed at a surface phenomenon, showed the enormous potential of a UHV TEM. By leaking oxygen into the microscope column during observation at high temperature, they obtained real-time images of the oxidation and etching of silicon.

Here we make use of the full power of this technique for interface studies, by growing Ge islands on a Si substrate inside the UHV TEM. We can thus observe every dislocation as it forms, and determine its character. A fascinating cyclic growth is observed. The island exhibits no detectable lateral growth for long periods, punctuated by sudden spurts of growth each time a dislocation is introduced.

We introduce a simple model to illustrate why this punctuated lateral growth occurs. For a fixed number of dislocations in the islands, growth is primarily vertical. However, each time a new dislocation forms, there is a sudden change in the preferred island shape, which appears as abrupt lateral growth.

We have modified a Hitachi 9000 UHV TEM, so that we can introduce both oxygen and germane in the column. The background pressure in the column is 2×10^{-10} Torr, regardless of whether the electron beam is on or off. Further, we have constructed a double-tilt heating stage, capable of heating samples up to 400 °C during observation. The double tilt enables us to achieve precise dark field conditions, critical to the study of dislocations. The evolution of the microstructure is recorded in real time on video tape.

The samples are prepared by mechanical thinning to about 00 μm, followed by chemical cleaning and thinning to electron transparency. The sample is flashed in the TEM to about 300 °C, and then oxygen etched, to further clean the surface and obtain thinner viewing areas. The cleanliness of the sample is demonstrated by the observation of surface truncation and reconstruction reflections visible in diffraction mode, and by the lack of any defects such as carbides. The surface reflections are monitored throughout the experiment, in order to ensure that the surface remains clean. Ge is deposited by leaking a 0:1 He-GeH_4 mixture into the column to a pressure of about 10^{-6} Torr, while the sample is at a temperature of 650 °C. The growth rate in these conditions is of the order of a few monolayers per hour.

0031-9007/94/73(2)/300(4)$06.00

All images were obtained in a weak-beam dark field condition, using the [220] reflection. This gives rise to the typical "black-white" contrast observed in Fig. 1, which shows an array of fully coherent (i.e., dislocation-free) strained islands. This condition is ideal to study the formation of dislocations, which will appear as bright lines on the dark half of the island (see Fig. 2). However, it is not possible to observe dislocations forming on the white side of the island.

Figure 2 is a series of images from the video recording of Ge islands growing at 650 °C. While a few images cannot capture the wealth of information contained in a real-time videotape, they illustrate the main results. Figure 2(a) shows the island after the formation of one dislocation. The dislocation has formed near the edge of the island, and the island has grown by about 200 Å past that dislocation. We did not observe the formation of this dislocation in real time, and found it very difficult to ever observe the formation of the first dislocation, because all of the strained islands quickly reach a uniform size, and it is impossible to guess which one will dislocate first (Fig. 1). However, once a single dislocation has been introduced, that island becomes more fully relaxed and hence a preferential site for Ge attachment, so it grows faster than the surrounding strained islands. We can then follow the growth of this island, and observe the formation of further dislocations in real time.

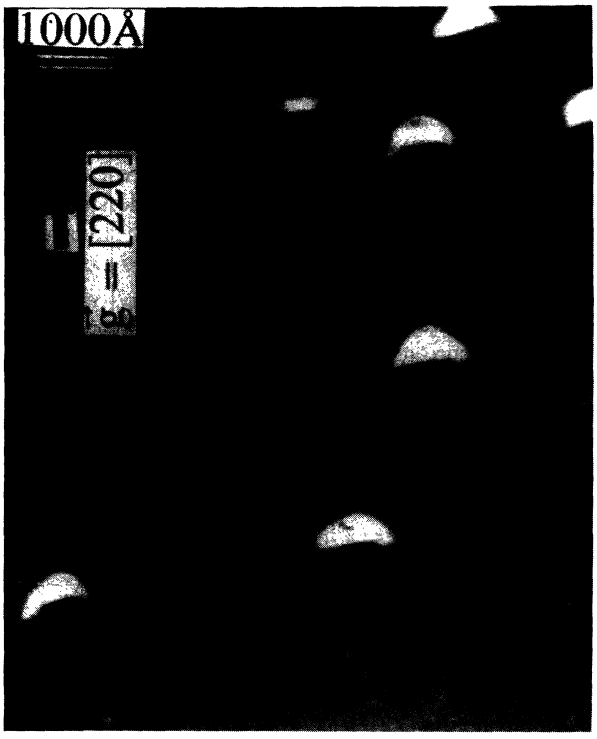


FIG. 1. Strained Ge island on Si(001). The "black-white" contrast is typical of the weak-beam dark field condition on the [220] reflection used to image the island. This microstructure was obtained after 30 min of growth.

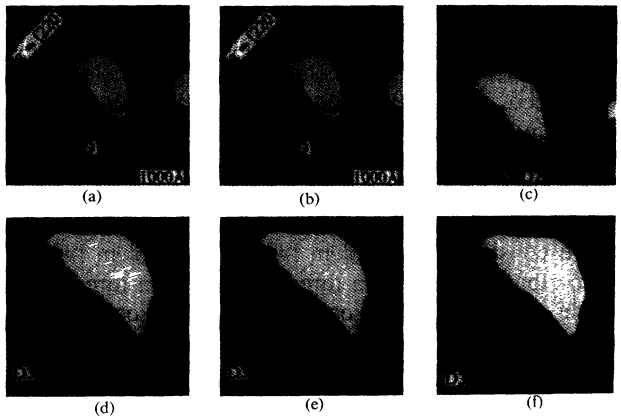


FIG. 2. Series of frames grabbed from the video recording of the growth, at the following time intervals (starting from beginning of growth). Note the change of magnification between (a)–(c) and (d)–(f). (a) $t = 59$ min, (b) $t = 94$ min, (c) $t = 96$ min, and (d)–(f) $t = 140$ min; these three pictures were taken within about 1 s of each other.

The "black side" of the island in Fig. 2(a) did not grow detectably for 35 min after we began watching it. (A dislocation was introduced on the white side, explaining the apparent growth on the top left of the island, but we will not discuss these dislocations, because we cannot observe their formation.) Then, a second dislocation ($D2$) formed very quickly, in about 1 s, and the island grew past that dislocation by about 200 Å in a few seconds [(Fig. 2(b)]. A $\mathbf{g} \cdot \mathbf{b}$ analysis shows that these are 60° dislocations, which can relieve the strain in both the [220] and the $[2\bar{2}0]$ directions, depending on their line direction. $D2$ thus extended more slowly from the [220] direction toward the $[2\bar{2}0]$ direction [Fig. 2(c)], allowing the island to grow in this direction too.

At this point, the apparent growth of the island stopped again for 45 min. Figures 2(d)–2(f) show three frames grabbed within 1 s of each other, where the introduction of dislocation $D3$ is observed. These highlight the advantage of the *in situ* approach, where the dislocation can be observed as it forms. It shows that, when a segment of the dislocation is nucleated, the island immediately grows past that segment by about 100 Å [Fig. 2(d)]. As the dislocation extends sideways, the island, which had not grown detectably for about 45 min, quickly extends past the dislocation by 200 Å [Figs. 2(e) and 2(f)]. This behavior was observed for the formation of four additional dislocations on this island. Each time, the lateral growth was undetectable for a period of 30 to 45 min, and then "jumped" by order of 200 Å each time a dislocation was formed.

This apparent stop-start behavior is due to oscillations in the shape of the island, which occur, we believe, because the energetically preferred island shape depends on

the number of dislocations. Dislocations can occur only in integer numbers, so in the equilibrium limit, the shape changes discontinuously each time another dislocation is introduced. While the actual growth conditions are far from equilibrium, we can illustrate the basic nature of the oscillations with a mean-field model for the equilibrium shape of a relaxed island, which includes surface, strain, and dislocation energies [8].

We consider an island in two dimensions (equivalent to an elongated island in three dimensions), having mean width w. With n dislocations, the mean strain ε in one direction is reduced from ε_0 (0.04 for Ge on Si) to $\varepsilon_0 - nb/w$, where b is the misfit component of the Burgers vector. The strain energy $M\varepsilon^2/2$ (where M is an elastic constant) is correspondingly reduced. For simplicity, *elastic* relaxation of the island is not included in the model. Each dislocation is assigned an energy λ per unit length (we neglect the logarithmic dependence on boundaries), and the extra surface energy per island length is assumed to be proportional to the height, as for a trapezoidal island (this shape is justified by *ex situ* cross-sectional TEM studies of relaxed Ge islands; see Ref. [9] for examples). Numerical values are estimates for Ge on Si(100); details are given in Ref. [8].

In the limit of slow growth, at each volume the island will adopt the height, width, and number of dislocations which minimizes the energy. The result, within our model, is shown in Fig. 3. The island grows vertically at constant width until it becomes energetically favorable to introduce another dislocation. At that point, the island height drops discontinuously, though never becoming as low as at the beginning of the previous cycle. The shape (the ratio of height to width) oscillates with each period, corresponding to the introduction of one dislocation.

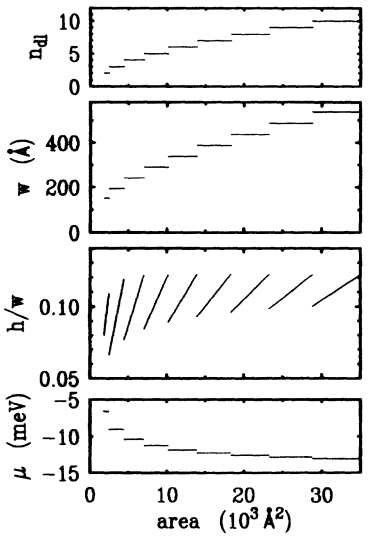


FIG. 3. Calculated island properties versus island size (cross-sectional area), from the mean-field model described in text. From top, properties are number n_{dl} of dislocations, island width w, height-to-width ratio h/w, and chemical potential μ, where zero corresponds to a uniform strained layer. In contrast with experiment, no islanding occurs prior to the introduction of dislocations, because the model does not include elastic strain relaxation.

This matches exactly what is observed experimentally. The experiment only determines the width of the island, not its height. However, since the flow of Ge is continuous, and the island is an effective sink for Ge throughout the growth cycle [10], it is reasonable to assume that the volume of the island increases linearly with time. Consequently, when we see no lateral growth, it means that the height of the island is increasing. Then, when fast lateral growth is observed (when each dislocation is introduced), the island must be growing laterally by the transport of atoms from its top to its edge, in order to optimize its shape. After that, lateral growth stops, and the height of the island again begins to slowly increase, until another dislocation is introduced.

Certain features of the observed growth, however, cannot be understood from the viewpoint of equilibrium energetics. Evidently nucleation of dislocations occurs at the edges of the island. Since the dislocations cannot glide on the (001) plane, they cannot move once they are at the interface. Thus there is no way for dislocations to reach the center of the island, and the island center remains free of dislocations even after considerable growth has occurred [Fig. 2(e)]. Furthermore, the islands initially relax elastically, which results in nucleation of dislocations being delayed compared to the simple model used here.

In conclusion, we have found that strain-relaxed islands grow in a remarkable fashion. Growth is almost entirely vertical, except for periodic lateral "jumps" associated with sudden changes in island shape each time a dislocation forms. This behavior can be understood based on the energetics of strain-relaxed islands. In addition, this study highlights the advantages of *in situ* electron microscopy in unraveling the complex process of dislocation formation and strain relief in epitaxial growth systems.

It is a pleasure to thank John Ott for his contributions made to the installation and construction of the electron microscope and associated equipment.

[1] For a review of recent literature on the subject, see F. K. LeGoues, in "Encyclopedia of Advanced Materials" (Pergamon, Oxford, to be published).
[2] F. K. LeGoues, B. S. Meyerson, J. F. Morar, and P. D. Kirchner, J. Appl. Phys. **71**, 4230 (1992).
[3] D. J. Eaglesham, E. P. Kvam, D. M. Maher, C. J. Humphreys, and J. Bean, Philos. Mag. **59**, 1059 (1989).
[4] M. Krishnamurthy, J. S. Drucker, and J. A. Venables, J. Appl. Phys. **69**, 6461 (1991).
[5] R. M. Tromp, A. W. Denier van der Gon, F. K. LeGoues, and M. C. Reuter, Phys. Rev. Lett. **71**, 3299 (1993).

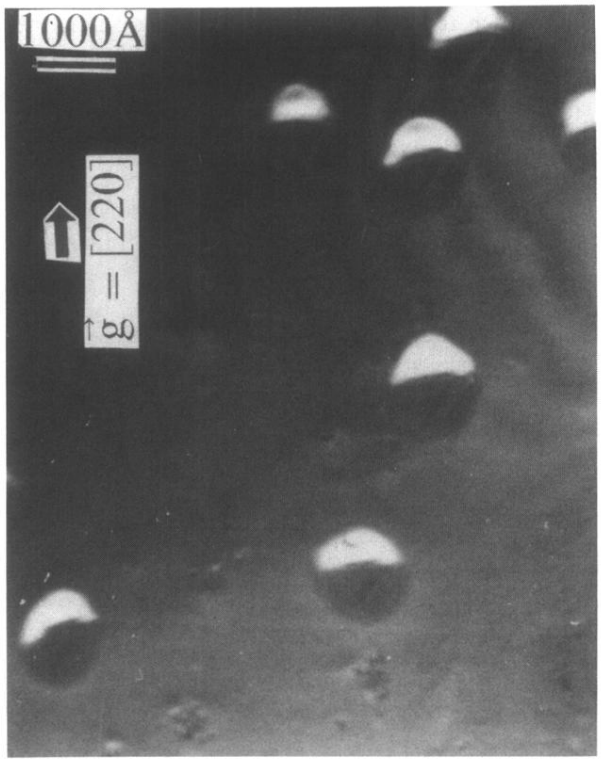
[6] D. W. Pashley, M. J. Stowell, M. H. Jacobs, and T. J. Law, Philos. Mag. **10**, 571 (1964); D. W. Pashley, Adv. Phys. **14**, 327 (1965).

[7] F. M. Ross and J. M. Gibson, Phys. Rev. Lett. **68**, 1782 (1992).

[8] For convenience, the dislocation core energy can be related to a more accessible quantity, the equilibrium critical thickness. For further details, see J. Tersoff, Appl. Phys. Lett. **62**, 693 (1993). We assume a critical thickness of 8 Å. The surface energy is evaluated assuming a trapezoidal shape, as in J. Tersoff and R. M. Tromp, Phys. Rev. Lett. **70**, 2782 (1993). We take a surface energy of 0.1 eV $Å^{-2}$, and a (311) orientation for the island sides. Changing these values, or including other effects such as elastic relaxation and anisotropy of the surface energy, would obviously not change the basic qualitative behavior.

[9] F. K. LeGoues, M. Copel, and R. M. Tromp, Phys. Rev. B **42**, 11 960 (1990); D. J. Eaglesham, F. C. Unterwald, and D. C. Jacobson, Phys. Rev. Lett. **70**, 966 (1993).

[10] From Fig. 3, the chemical potential of the island is always seen to be much lower than that for an unrelaxed island. Thus at any point in the oscillation the island will irreversibly trap diffusing adatoms. The growth rate is therefore limited only by diffusion of incoming atoms, and remains constant throughout the oscillatory growth.



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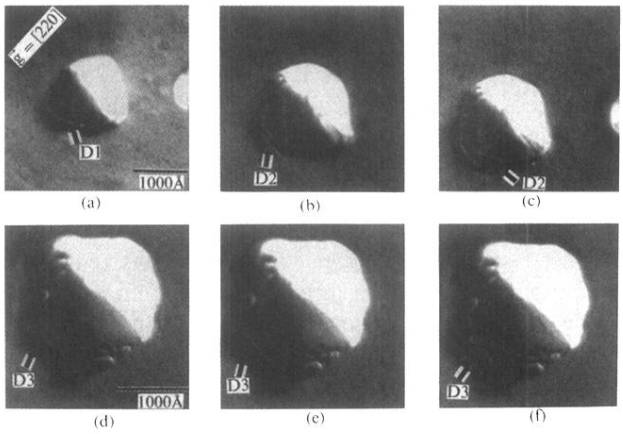

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