

Chapter 3. Step Structures and Epitaxy on Semiconductor Surfaces

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3.1 Introduction

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At present, there is considerable effort directed towards the fabrication and utilization of nanoscale semiconductor objects. This is, in part, to investigate the behavior of electrons within structures of sufficiently small dimensions to produce quantum confinement. Such so-called *quantum dots and quantum wires* have dimensions of typically a few hundred Angstroms. In addition, quantum dots and wires have been proposed as a route to develop many new device applications.¹

Recently, it has emerged that nanoscale structures on certain semiconductor surfaces may spontaneously self-assemble into more or less ordered patterns. Over the last year, we have been pursuing investigations of two examples of this remarkable phenomenon. Our goal is to achieve a microscopic and predictive understanding of the factors that determine the nanostructure. We also want to understand how it may be controlled and exploited in the creation of novel nanoscale surface structures, for example, by decoration schemes to create chemically heterogeneous surfaces. Specifically, we have pursued studies of the self-assembly of nanoscale grooves on stepped silicon surfaces. In addition, we have initiated a study of Ge quantum dots on Si.

3.2 Self-assembly of Nanoscale Gratings on Silicon

We have found that certain stepped silicon surfaces spontaneously form a remarkable mesoscopically grooved morphology. One side of each groove is a step-free (113) facet and the other side is a step bunch. Individual grooves are a few hundred Angstroms across and exceed one micron in length. The grooves self-assemble to form a nearly-periodic nanoscale grating. Associated with this grating are first-order diffraction peaks. The peak locations are determined by the grating periodicity and the peak widths by the degree of order of the grating. Figure 1 shows typical scattering profiles obtained at several times up to 1200 s, following a temperature quench from 1248 K, where surface steps are uniformly separated, to 1171 K, where step bunches form, in turn forming grooves. The peak at $Q_y = 0$ corresponds to specular reflection (zeroth-order diffraction peak). The two peaks, symmetrically located about the specular peak, constitute first-order diffraction peaks from the grooves. With increasing time, these peaks grow in intensity, shift to smaller wavevectors, corresponding to an increase in the groove period, and become narrower, corresponding to an increasingly ordered structure.

The peak positions and widths (HWHM) of the first-order diffraction peaks are shown on a log-log scale versus time for three representative final temperatures in figure 2. The variation of the peak position versus time is consistent with a 1/6 power-law behavior for times between one and several hundred seconds. This result has inspired a theory of the growth of grooves, which reproduces the 1/6 power-law and identifies the rate-limiting step as the close approach of neighboring step bunches.² However, at longer times the position approaches a

¹ C. Weisbuch and G. Vinter, *Quantum Semiconductor Structures* (Boston: Academic Press, 1991).

² S.T. Milner (unpublished).

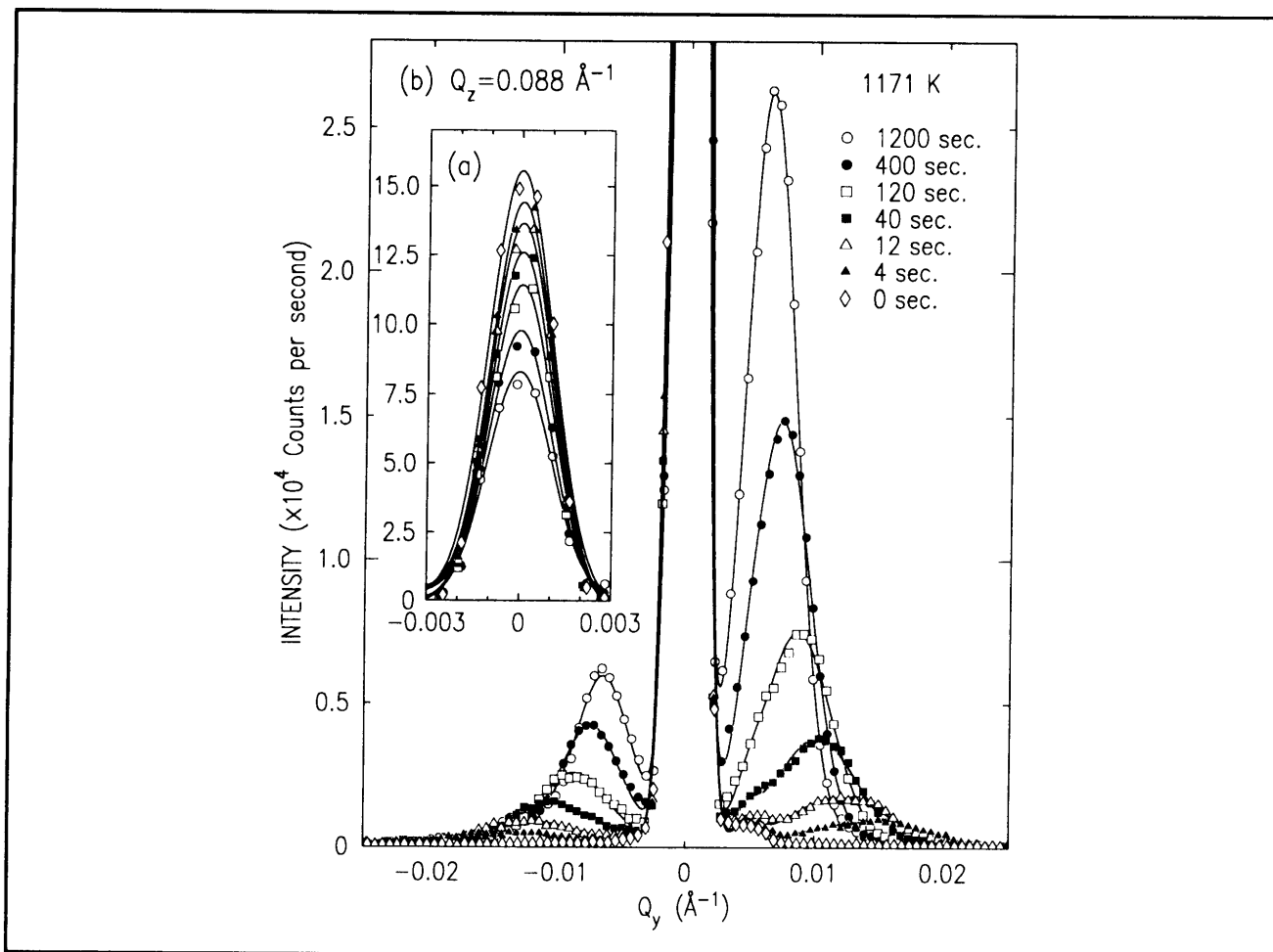


Figure 1. Time evolution of the first-order diffraction peaks of the groove superstructure, following a temperature quench from 1248 to 1171 K.

constant value, corresponding to a limiting groove size. By contrast, the peak width continues to decrease even at long times, indicating that the surface morphology evolves to an ordered equilibrium state. Notice that, by a second quench to room temperature, we may prepare a surface with any periodicity between 300 and 1200 Angstroms at will.

Shown in figure 3 is the limiting facet size plotted versus the angle between the sides of the groove for two samples with different misorientations. It seems especially surprising that the limiting facet size depends nonmonotonically on the angle. However, this may be understood qualitatively on the basis of the work done by Marchenko³ and Alerhand,⁴ where it is proposed that the lowest

energy configuration of a faceted surface corresponds to a periodically grooved morphology. The special stability of the grooved structure originates in a reduction of surface-stress-induced elastic energy in the bulk that occurs for mesoscopic facets as compared to macroscopic facets. The period is determined by balancing the reduction in elastic energy against the energetic cost of maintaining edges between the coexisting phases. The theory predicts that the groove period is minimized when the angles between each side of the groove and the macroscopic surface are equal, as found experimentally for the two differently misoriented samples. Thus, we have achieved an in-depth understanding of the mechanism responsible for nanoscale grooves on stepped Si(113) surfaces. As a result, it may become possible to engineer the

³ V.I. Marchenko, *Zh. Eksp. Teor. Fiz.* 81: 1141 (1981); *Sov. Phys. JETP* 54: 605 (1981).

⁴ O.L. Alerhand, D. Vanderbilt, R. Meade, and J.D. Joannopoulos, *Phys. Rev. Lett.* 61: 1973 (1988).

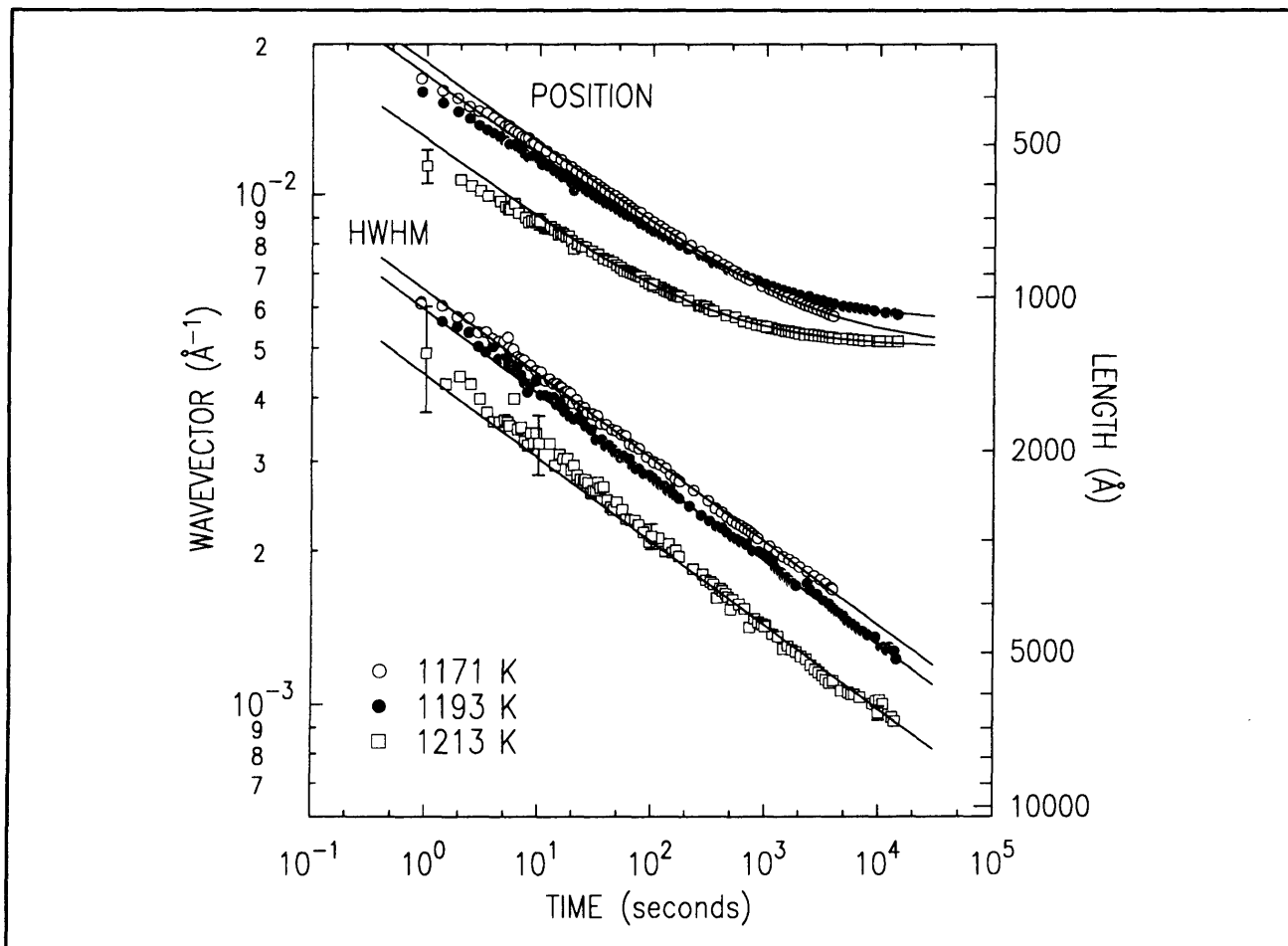


Figure 2. Log-log plot of the position and HWHM for three representative quench temperatures.

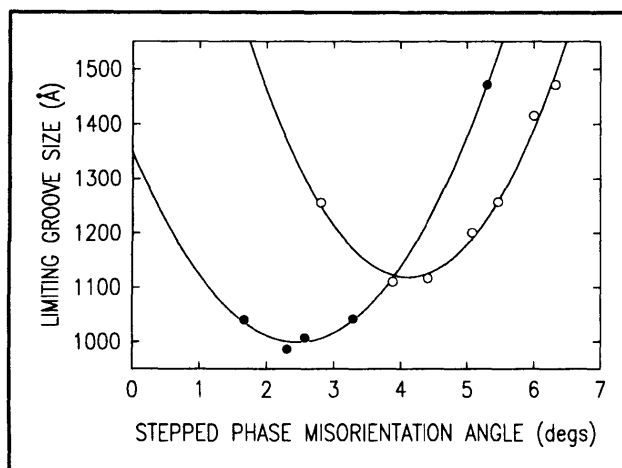


Figure 3. Limiting groove size vs. the groove angle.

equilibrium groove size. For example, an adsorbed species might lower the edge energy, allowing us to tune the groove periodicity with coverage.

The results described in this section refer to a single azimuthal orientation of the steps. In this case the surface is tilted away from (113) towards (001). We plan to investigate the behavior for other azimuthal orientations. In particular, we have initiated studies of surfaces tilted towards $(1\bar{1}0)$ (90° -rotated with respect to the surfaces studied previously by 90°). Figure 4 shows an AFM image of this surface, which was quenched to 300 K after equilibration at 900 K. The surface morphology for the azimuth of figure 4 is evidently more complicated than simple linear grooves. Specifically, straight steps are not stable; instead, there are kinks between two stable step orientations, giving rise to a two-dimensionally patterned substrate. Further AFM measurements of this surface quenched from other temperatures and x-ray measurements versus temperature are called for to further elucidate this behavior.

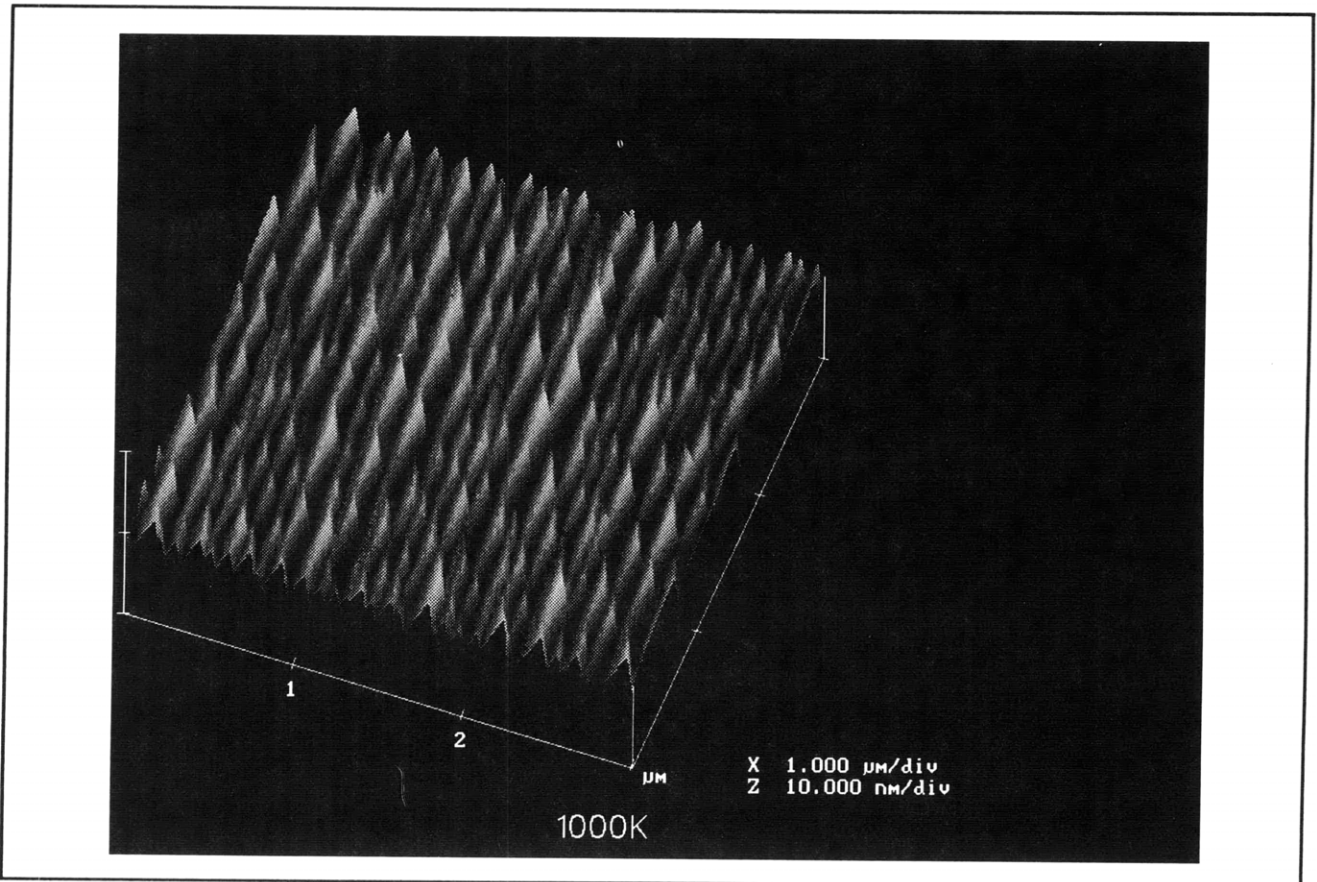


Figure 4. AFM image of the surface morphology of a stepped Si(113) surface tilted towards $(1\bar{1}0)$, quenched from 900 K.

3.3 Self-assembly of Quantum Dots

The manufacture of nanostructure semiconductor objects is exceedingly difficult, because these structures are tiny. For example, it is difficult to achieve the requisite sizes with lithography. Schemes which directly build quantum-sized objects during growth are possible alternatives. Such schemes may include cluster formation in the gas phase and subsequent cluster deposition and growth on patterned substrates such as stepped surfaces.

An especially promising and interesting approach is the self-assembly of a nanostructure during strained heteroepitaxial growth on a flat substrate. Strained heteroepitaxial growth, involving a large lattice mismatch, generally proceeds via the formation of islands after the initial growth of a few flat layers. Recently, Krishnamurthy et al.,⁵ Leonard et al.,⁶ and

Moison et al.⁷ have found that it is possible to prepare near-uniformly sized and spaced islands of dislocation-free Ge on Si(001), InGaAs on GaAs(001), and InAs on GaAs(001), respectively. It is this apparent self-organization of defect-free quantum dots that is so exciting, both from technological and scientific viewpoints. However, a fundamental understanding of quantum dot formation—whether it is kinetic or thermodynamic in origin—is presently lacking.

In the earlier studies, it was not possible to examine *in-situ* island formation during the deposition and annealing processes. By contrast, we have initiated an *in-situ* investigation of the self-assembly of quantum dots of germanium on silicon as it occurs. We are using both synchrotron x-ray scattering techniques to monitor how the surface morphology develops in real-time and the atomic force micro-

⁵ M. Krishnamurthy, J. Drucker, and J.A. Venables, *J. Appl. Phys.* 69: 6461 (1991).

⁶ D. Leonard et al., *J. Vac. Sci. Technol. B* 12: 1063 (1994).

⁷ J.M. Moison et al., *Appl. Phys. Lett.* 64: 196 (1994).

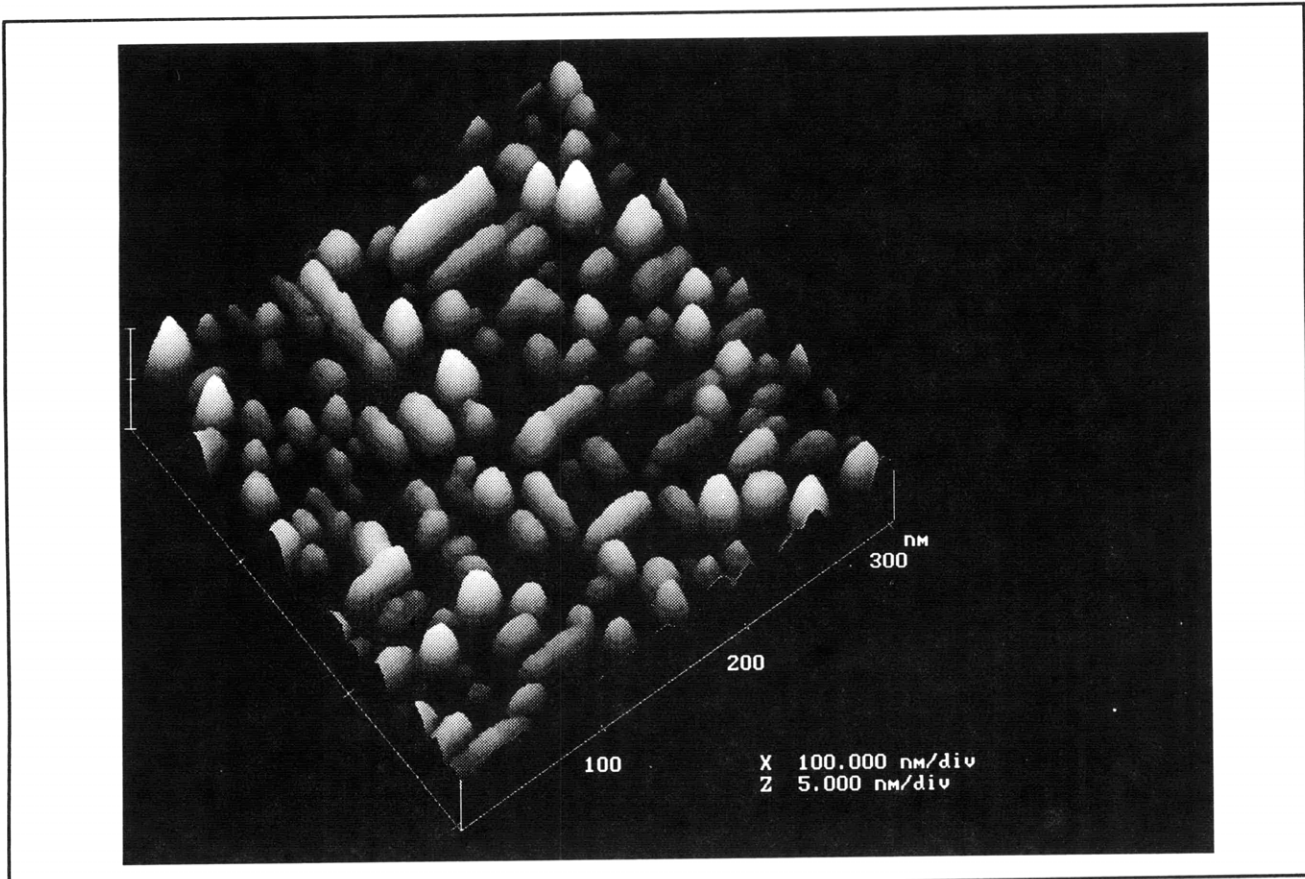


Figure 5. AFM image of Ge quantum dots on Si(001).

scope to definitively determine “snap shots” of the surface morphology by means of rapid temperature quenches. In this way, we will achieve a microscopic and predictive understanding of the factors that determine the final morphology. As result, we expect to learn how to control the nanostructure.

We have chosen to study initially the Ge-on-Si system, in part, because of the earlier work of Krishnamurthy et al., and also because of the potential utility of Ge-Si heterostructures in device applications. In addition, the topic of the present research was inspired by the detailed understanding of the role of elasticity in promoting nanoscale self-assembly. We have gained this knowledge from our own measurements (made within the Joint Services Electronics Program at MIT) of the self-assembly of a nanoscale grating on stepped silicon surfaces described above.⁸

Figure 5 illustrates an AFM image of a surface upon which is deposited the equivalent of approxi-

mately eight monolayers of Ge at a substrate temperature of 300 K, which we obtained. Three monolayers grow in a layerwise fashion, but subsequent deposition occurs in the form of three-dimensional islands, i.e., quantum dots, which may be clearly seen in the image more or less uniformly spaced across the surface. This figure demonstrates that we can prepare quantum dots for study. As may be seen, some of the dots are elongated along two orthogonal directions, which are high symmetry directions of the substrate. This interesting aspect is not understood.

The development of the morphology seen in figure 5 may be monitored *in situ* and in real time using synchrotron x-ray scattering. In this way, we will investigate the evolution of the surface morphology versus time at different deposition temperatures and for different Ge coverages and temperatures subsequent to deposition. Our goal is to gain a quantitative microscopic understanding of Ge quantum dots on Si(001) and how they form. On the basis of the

⁸ S. Song and S.G.J. Mochrie, *Phys. Rev. Lett.* 73: 995 (1994); S. Song and S.G.J. Mochrie, *Phys. Rev. B* 51: 10068 (1995); S. Song, S.G.J. Mochrie, and G.B. Stephenson, *Phys. Rev. Lett.* 74: 5240 (1995).

behavior observed for stepped Si(113), we are lead to speculate that Ge quantum dots on Si may themselves self-assemble under the right conditions into a *periodic* superlattice as a result of elastic effects. It would then be possible to grow Si on top of such a structure. Following on from layered superlattices, such a three-dimensionally patterned material would be very interesting.

3.4 Publications

Mochrie, S.G.J., S. Song, M. Yoon, and D.L. Abernathy. "Faceting of Stepped Si(113) Surfaces: Self-assembly of Nanoscale Gratings." *Physica B*. Forthcoming.

Song, S., M. Yoon, and S.G.J. Mochrie. "Faceting, Tricriticality, and Attractive Interactions Between Steps in the Orientational Phase Diagram of Silicon Surfaces Between [113] and [5 5 12]" *Surf. Sci.* 334: 153 (1995).

Song, S., S.G.J. Mochrie, and G.B. Stephenson. "Faceting Kinetics of Stepped Si(113) Surfaces: A Time-Resolved X-ray Scattering Study." *Phys. Rev. Lett.* 74: 5240 (1995).

Song, S., and S.G.J. Mochrie. "Attractive Step-step Interactions, Tricriticality, and Faceting in the Orientational Phase Diagram of Silicon Surfaces Between [113] and [114]." *Phys. Rev. B* 51: 10068 (1995).