SiGe Coherent Islanding and Stress Relaxation in the High Mobility Regime

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Real-time stress measurements during Si_8Ge_2/Si(001) heteroepitaxy, combined with *ex situ* microscopy, are used to examine islanding dynamics under conditions of relatively low strain and high adatom mobility, where morphological evolution bypasses dislocation formation. We show that growth in this regime proceeds similarly to growth of Ge/Si(001) (i.e., at high strain, low temperature), but with the length scales expanded by the reduced strain. This greatly facilitates measurement of the coupled kinetics of morphological evolution and stress relaxation. [S0031-9007(97)04479-7]

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Under conditions of relatively high misfit coherency strain ($\varepsilon_{\text{coh}} > 2\%$), heteroepitaxial thin films often grow via the Stranski-Krastanov (S-K) mode, i.e., the first 1–3 monolayers (ML) of film grow as a two dimensional (2D) wetting layer, but subsequent adatoms cluster to form small three dimensional (3D) islands. These islands can retain coherent interfaces with the underlayer, but nonetheless some portion of the net strain energy is relaxed by the 2D-3D transition—we will call this the geometric relaxation. This behavior is governed by the energetics of the strain field and the surface [1], subject to the kinetics of adatom transport that regulate nucleation and evolution of islands, in competition with dislocation introduction [2–6]. Although the reduction of strain energy is the driving force for coherent island formation, the degree of overall relaxation due to islanding is not well characterized.

In this Letter we report the first real-time measurements of the stress relaxation kinetics due to coherent island formation. The experiments are performed in a low strain, high temperature growth regime [Si_8Ge_2/Si(001) at 760 °C, $\varepsilon_{\text{coh}} = 0.8\%$] where dislocation formation is slow relative to adatom transport. Even at this low strain, we show that film growth proceeds similarly to that of pure Ge on Si(001) ($\varepsilon_{\text{coh}} = 4.2\%$), but with the length scales expanded an order of magnitude, thereby facilitating measurement of the detailed dynamics of island evolution. We observe formation of [501]-faceted "hut clusters" [7] via the S-K mode, followed by shape-invariant island growth and coarsening. The dense hut array relaxes 20% of the coherency stress. At larger thicknesses, an island shape transition occurs that relaxes nearly half the coherency stress while kinetically bypassing dislocation introduction by modifying the substrate surface.

Si_8Ge_2 films were grown at 760 °C in ultrahigh vacuum on a clean Si(001) buffer layer by electron-beam evaporation at a total growth rate of 0.1 Å/s. Details have been given elsewhere [8,9]. The stress in the film was determined in real time by measuring the curvature $\kappa(t)$ of the substrate using a sensitive multibeam optical deflection technique [8,9]. The curvature is proportional to the product of the film stress and the mean film thickness, here called the "stress-thickness." We use the real-time measurement both to quantify the stress relaxation during islanding, and to efficiently identify the interesting growth and thickness regimes in which to apply *ex situ* analysis.

Figure 1 shows the stress-thickness vs mean film thickness measured during Si_8Ge_2/Si growth. For "ideal" growth of a coherent film with uniform composition and a planar surface, a linear response is expected, since the coherency stress is constant and the film thickness increases linearly with time. The nonlinear response observed upon initiation of growth results from surface segregation of Ge [10]. Subsequently, a brief linear regime is observed whose slope, shown as a dashed line in Fig. 1, is within 3% of the expected value (1.51 GPa) for coherently strained Si_8Ge_2/Si.

From 30–65 Å deposited film thickness, the measured stress-thickness curve exhibits a slope transition region, labeled as $T1$ in Fig. 1, in which stress relaxation is occurring. Note that this is below the equilibrium critical thickness for introduction of misfit dislocations. Subsequent to $T1$ the stress-thickness evolves with a constant slope of 1.15 ± 0.06 GPa. From about $h_f = 140$–210 Å, a second transition region, labeled at $T2$ in Fig. 1, is observed. The slope in the regime after $T2$ is only 0.45 ± 0.03 GPa.

To evaluate the corresponding morphological evolution, further depositions were performed under identical growth conditions, terminating growth at thicknesses between 25 and 275 Å. The real-time curvature evolution is highly reproducible in all cases. The films were imaged *ex situ* using tapping-mode atomic force microscopy

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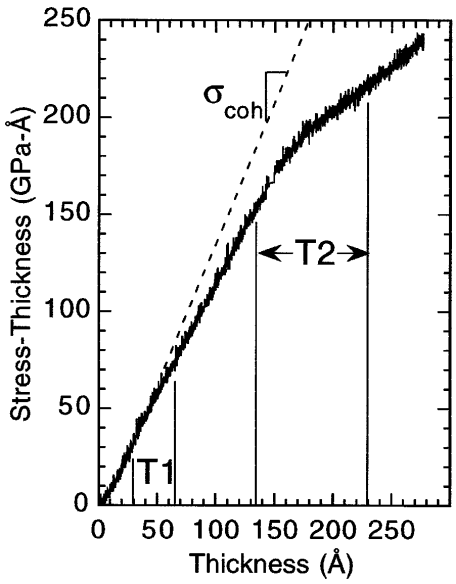

FIG. 1. The stress-thickness vs deposited thickness, determined from measurement of the substrate curvature during Si_8Ge_2/Si molecular-beam epitaxy (MBE) growth at 760 °C. The dashed line is a fit to the planar growth stage. The slope of the curve represents the instantaneous stress of the system.

(AFM), scanning electron microscopy (SEM), and transmission electron microscopy (TEM), both in plan-view (PTEM) and cross-section (XTEM) electron microscopy.

At 25 Å, just prior to $T1$, the film surface consists of extensive planar regions, with a sparse population of small features that may be incipient islands [Fig. 2(a)]. Root-mean-square roughness is approximately 6 Å, i.e., the film is still essentially flat. The islandlike features are clearly not laterally assembled and cooperative nucleation is not observed [11].

At 100 Å, in the linear stress-thickness regime between $T1$ and $T2$, the film consists entirely of dislocation-free pyramidal islands bound by [501] facets, with a narrow size distribution, and periodically assembled along {100} in-plane directions [Fig. 2(b)]. This is the well-known hut cluster morphology that has been observed in growth of pure Ge [5,7,12]; however, the mean island size for dense arrays of Si_8Ge_2 huts is an order of magnitude larger than in the case of pure Ge growth due to the lower strain.

We grew a sample 65 Å thick, to coincide with the end of islanding transition $T1$, and capped the sample *in situ* with amorphous Si to prevent oxidation. XTEM of this sample resolves the presence of a 20–30 Å thick planar alloy wetting layer (Fig. 3). This corresponds to the thickness at the beginning of the islanding transition, suggesting that the film grows 2D for about 25 Å (≈18 ML), but further deposition results in adatoms clustering into 3D islands on top of the wetting layer. This is similar to growth of pure Ge on Si(001), which grows in the S-K mode, but with a wetting layer thickness of only 3 ML [5,7,12].

Between $T1$ and $T2$ in Fig. 1, the stress-thickness evolves linearly. This behavior is consistent with growth

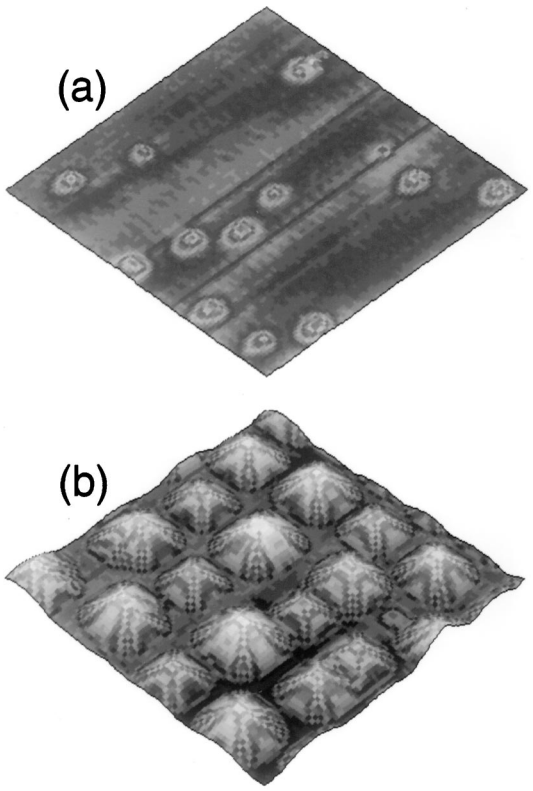

FIG. 2. (a) AFM image of a 25 Å thick Si_8Ge_2/Si film. (b) AFM image of a 100 Å thick Si_8Ge_2/Si film. Both AFM images are 2×2 μm, scanned along {100}, and have the same vertical scale (1000 Å).

of islands with constant shape. Specifically, the curvature evolution during coherent islanding will be given by

$$\kappa(t) \propto \sigma_{\mathrm{coh}} h_w(t) + \langle \sigma_{\mathrm{isl}} \rangle [Rt - h_w(t)] + \Delta \overline{F}(t), \quad (1)$$

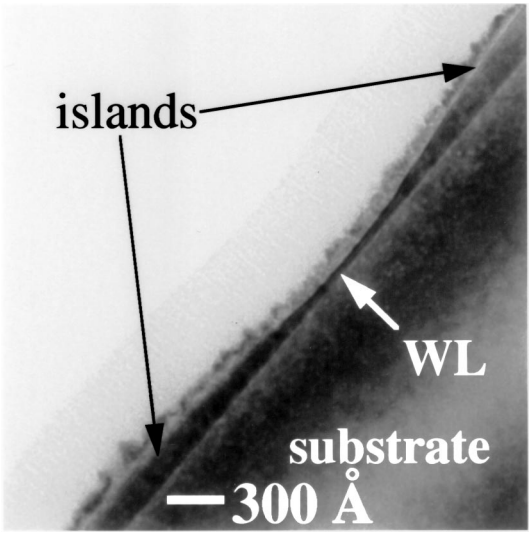

FIG. 3. XTEM image of the 65 Å Si_8Ge_2/Si film showing the wetting layer (WL) underlying the island array. The "structure" above the Si_8Ge_2 film arises from the (nominally) amorphous Si cap layer.

where σ_{coh} is the coherency stress of the planar film, $\langle\sigma_{\mathrm{isl}}\rangle$ is the effective stress of a coherent island array [13], h_w is the thickness of an underlying wetting layer, R is the deposition rate, and $\Delta\overline{F}(t)$ is the change in the mean surface stress associated with the islanding process. The surface stress term in Eq. (1) is estimated to be essentially constant between $T1$ and $T2$ since the change in surface area is less than 1% in this regime. Assuming the islands all have the same shape (but not size) at any given film thickness [14], the island stress can be written as $\langle\sigma_{\mathrm{isl}}\rangle = g[\mathcal{A}(t)]\sigma_{\mathrm{coh}}$, where $g[\mathcal{A}(t)]$ is the fractional geometric relaxation of an island with aspect ratio (i.e., height-to-width ratio) $\mathcal{A}(t)$. According to Eq. (1), linear curvature evolution requires that $g(\mathcal{A})$ be constant, i.e., that the island shape is time invariant [15]. AFM, SEM, and XTEM measurements of films grown between $T1$ and $T2$ support this conclusion.

The linear curvature evolution also suggests that the wetting layer thickness must be constant or varying slowly relative to the deposition rate. From the data of Fig. 1 between $T1$ and $T2$, combined with XTEM measurements of the wetting layer evolution, and using Eq. (1), we find that hut clusters relax $20 \pm 2\%$ of the effective stress in the film [$g(\mathcal{A}) = 0.8 \pm 0.02$], where the uncertainty arises from uncertainty in the measurement of the wetting layer thickness as a function of film thickness. This degree of relaxation for hut clusters is consistent with results of 2D finite element calculations [15].

Steinfort *et al.* recently reported x-ray measurements of the strain distribution in Ge hut clusters [16]. Integrating their distribution gives a 15% effective relaxation, slightly less than the $20 \pm 2\%$ value for Si_8Ge_2 huts reported here. We have grown Ge on Si(001) under conditions similar to Ref. [15] while measuring the curvature in real time [9], and we obtain an effective relaxation of 20%, consistent with our alloy measurements. Also, our results for Ge/Si(001) suggest that the [501] facet has a large intrinsic surface stress, which would tend to stabilize this facet under compression by reducing the surface energy [17], perhaps contributing to the strong preference for [501] formation in the $Si_{1-x}Ge_x/Si(001)$ system.

During growth between $T1$ and $T2$, we observe that the hut clusters not only increase in size, but the island periodicity increases from 3150 Å at 65 Å film thickness to 4250 Å at 104 Å. Thus the island array coarsens during growth, while maintaining a fixed island shape (huts). Nucleation of new islands does not occur in this regime since the areal coverage of the island array at the end of $T1$ (65 Å) is large enough such that most islands interact with their neighbors through the local strain and diffusion fields. Finite element calculations show that there is a short-range repulsive strain interaction between islands due to the stress concentration along the basal perimeters [6,18]. After 65 Å, the array is sufficiently dense that nucleation of new islands will be suppressed in favor of coarsening of the existing array, as long as adatom diffusion lengths are comparable to the island lateral length scales [19]. The local strain interactions may also lead to lateral self-assembly, cf. Figs. 2(a) and 2(b).

We also performed a static coarsening experiment, in which a film 104 Å thick was grown at 760 °C and then held at the growth temperature for 40 min. The mean island spacing increases from 4200 to 6000 Å during the anneal and we observe that the wetting layer is consumed [20]. This clearly indicates that the original wetting layer thickness is not energetically determined, but rather is determined by the growth kinetics [21].

As the hut clusters enlarge during high temperature growth, the stored elastic energy per unit area linearly increases with film thickness. Eventually, shape-invariant island growth and coarsening breaks down in region $T2$ (Fig. 1). SEM shows that at 130 Å film thickness, coarsening can no longer keep up with growth, and hut impingement occurs. Rather than undergo island coalescence, which is energetically unfavorable, new islands (that we refer to as dome clusters [12]) nucleate and grow to consume the hut cluster population [Fig. 4(a)]. The domes have nearly twice the aspect ratio of the huts and consequently a much larger geometric strain relaxation. While $T2$ lies above the critical thickness for dislocation formation, inspection over approximately a 2500 μm^2

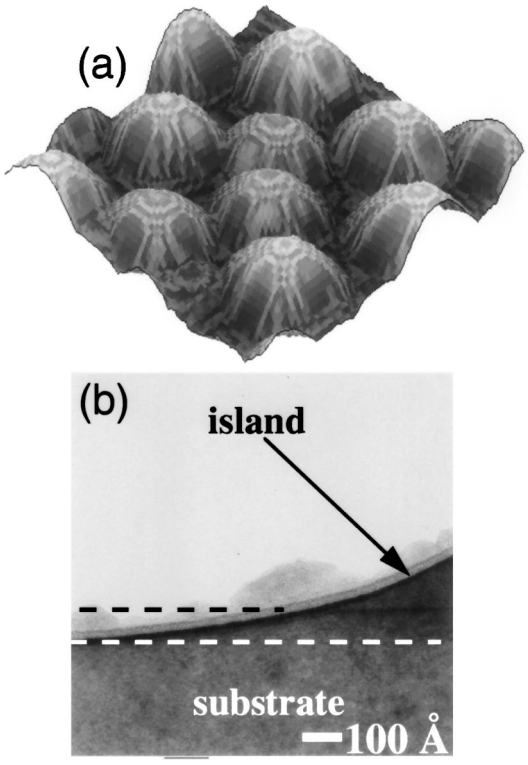

FIG. 4. (a) AFM image of a 275 Å film, same display parameters as in Fig. 2. (b) XTEM image showing the local removal of the substrate near an island basal perimeter. The distance between the black line (island/Si interface) and the white line (Si surface) is 73 Å.

surface area using weak-beam imaging in PTEM revealed only one island containing a dislocation. Thus the 46% stress relaxation observed at 275 Å results solely from the increased island aspect ratio [22]. This is in contrast to observations by Hammar *et al.* during real-time TEM studies of Ge growth of Si(001) [5,23]. They also observed nucleation of high-aspect-ratio islands amidst an array of hut clusters, but their islands always contained high densities of misfit dislocations introduced at the basal perimeters of the islands where the stress concentration is maximum. For Si_8Ge_2/Si at 760 °C, the low strain and high surface mobility allows the system to kinetically bypass dislocation formation through modification of the *substrate* surface. XTEM indicates that the interface between the island and the substrate (no wetting layer remains) lies above the surrounding Si surface [Fig. 4(b)]. The modification of the substrate surface reduces the contact angle between the dome and the substrate, as well as the local volume of material experiencing a stress concentration, thereby suppressing dislocation introduction. Similar depressions around pure Ge islands on Si(001) were reported by Kamins [24].

In conclusion, using real-time stress measurements of coherent island formation during deposition to guide *ex situ* microscopic analysis, we find that growth of a relatively low strain heteroepitaxial system, Si_8Ge_2/Si(001) under high adatom mobility conditions mimics growth of Ge on Si(001), but with greatly expanded layer thickness and island size scales. The increased length scales follow from the competition between surface energy and strain energy density (e.g., the critical nucleus size will vary as $\Gamma/\sigma_{\mathrm{coh}}^2$, where Γ is the difference in surface energy between the planar surface and the island facet [1]). This permits us to observe behavior that would be difficult to observe in Ge, such as hut cluster coarsening driven (at least partly) by local strain interactions, consumption of the kinetically determined wetting layer, and modification of the substrate surface morphology to bypass dislocation formation. Lateral assembly is obtained in these films directly through growth of dense island arrays at high temperature. We find that hut clusters geometrically relax 20% of the coherency stress, while dome clusters relax nearly half the effective stress in the film.

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