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$Si_{1-x}Ge_x$ growth instabilities on vicinal Si(001) substrates: Kinetic vs. strain-induced effects

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A comparative study of kinetically and thermodynamically driven instabilities on vicinal Si(001) surfaces during overgrowth with $Si_{1-x}Ge_x$ is reported. We mapped out a wide range of the multidimensional growth parameter space and found, in contrast to previous reports, no evidence for strain-induced step bunching. At low Ge concentrations strain is insufficient to promote strain-induced step bunching, and the modified surface kinetics in the presence of segregated Ge leads to a smoother rather than rougher morphology. High Ge concentrations around 50% could be expected to provide enough strain, but near equilibrium hut cluster formation is the more effective strain-relaxation mechanism. We found the characteristically rippled step-bunching morphology only in a kinetically limited growth regime, where strain is of limited relevance, and in experiments where the SiGe layers replicate an underlying ripple morphology.

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For most device applications semiconductor heterointerfaces have to be as smooth as possible to suppress interface roughness scattering or fluctuations in quantum confinement energies.[1,2] On the other hand, growth conditions that cause well-defined surface corrugations or three-dimensional (3D) islands have gained wide-spread interest for the implementation of self-assembled 1D and 0D nanostructures.[3] Commonly, corrugated surfaces have been associated with the built-in strain in lattice-mismatched heteroepitaxial layers,[4–6] but the relevance, and occasional dominance of the growth kinetics[7–10] becomes more and more obvious.

Here, we concentrate on the Si/SiGe heterosystem, which provides a maximum lattice mismatch of 4.2% between the pure constituents. In previous studies, single SiGe layers,[11] and Si/SiGe superlattices,[12] grown pseudomorphically on slightly vicinal Si(001) substrates were found to develop a rippled surface morphology, which was attributed to the interplay between kinetic and strain-induced effects. In an attempt to model these observations Tersoff *et al.* found that a compressively in-plane-strained layer can minimize its free energy by a bunching instability of the atomic height steps present on a vicinal surface.[5] Meanwhile, strain-induced step-bunching has been invoked to explain complex growth phenomena such as oblique replication of rippled Si/SiGe interfaces,[13] or the ordering of Ge dots on Si/SiGe superlattices.[14]

Recently, we found that a similar ripple morphology can develop under kinetic growth conditions on unstrained homoepitaxial Si layers.[10] This growth instability was shown to occur under MBE (solid source molecular-beam epitaxy) conditions that were frequently employed for the deposition of Si buffer layers and subsequent Si/SiGe layer sequences.

The morphological appearance of the two mechanisms is indistinguishable, but the involved strain fields, and hence the influence on subsequent heterolayers, are entirely different. Here we report, to the best of our knowledge, a first concise attempt to separate kinetic and strain-induced step-bunching in the accessible and application-relevant areas of a multidimensional growth parameter space. To study the influence of the growth parameters, vicinality (sample miscut), strain, and temperature, we investigated four sets of samples (Table I): set 1 to study the kinetic regime in SiGe layers, set 2 to asses the strain-dominated regime on substrates with small miscut angle (this condition is most frequently employed in the literature), set 3 for the strain regime with larger, but still commercially employed substrate miscut, and set 4 to investigate the influence of a kinetically corrugate Si buffer on the behavior of a subsequently deposited SiGe layer.

The detailed experimental conditions are described in Ref. 10. In brief: We cut samples from production Si(001) wafers with defined miscuts of 0.66° along [110], and 4.34° along [100]. A HF-free RCA procedure was employed for chemical pre-cleaning. The samples were radiatively heated in the MBE machine for 6 min to 1000 °C for *in situ* oxide desorption. This leads to atomically flat surfaces as confirmed by atomic-force microscopy (AFM). The active layers were then grown at 0.33–0.5 Å/s. Immediately after growth, the surface morphology was imaged on air with a Park Scientific AFM in contact mode.

For Set 1, 1000-Å-thick $Si_{1-x}Ge_x$ layers with Ge contents ranging from 0–25% Ge were deposited at 400, 450, and 490 °C on atomically flat, 0.66° miscut Si substrates. The rather thick layers are necessary to make kinetic step bunching easily observable,[10] but they limit the composition range due to the critical thickness for strain relaxation by misfit dislocations. Under the same growth conditions, homoepitaxial Si layers show a pronounced ripple morphology[10] with a mean amplitude of 10 Å (inset in Fig. 1). To rule out that Ge-induced modifications of the growth kinetics simply shift the temperature range where a maximum of the kinetic corrugations is to be expected, we varied the growth by almost 100 °C, but found little influence of the growth temperature on the morphology in this range. For very small Ge contents the $Si_{1-x}Ge_x$ layers develop a similar ripple pattern, but the mean peak-to-valley height of the ripples and the root-mean-square (rms) roughness decrease strongly with increasing Ge content (Fig. 1). Obviously, even minor Ge concentrations drastically affect the surface kinetics toward a suppression of kinetic step bunching. We attribute this to changes of the surface reconstruction,[15] which affects both adatom diffusion and step-edge incorporation. Ge segregation, which is near

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TABLE I. Growth parameters for the different sample sets used.

Set No.	Sample No.	Miscut Θ	T_{Growth}	Ge content	Surface morphology
2	*A*15	0.66°	550 °C	50%	
	*B*15[a]	0.66°	550 °C	50%	elongated huts
	*C*15	0.66°	650 °C	50%	square huts
	*C*14	0.66°	650 °C	40%	mounds + square huts
3	*A*45	4.34°	550 °C	50%	ripples
	*B*45[a]	4.34°	550 °C	50%	elongated huts
	*C*45	4.34°	650 °C	50%	elongated huts
4	*D*15	0.66°	490 °C	50%	ripple replication
	*E*15[b]	0.66°	490 °C	50%	hut clusters

[a]*In situ* post growth anneal at 550 °C for 1 h.
[b]*In situ* post growth anneal at 490 °C for 1 h.

its maximum at the growth temperatures employed,[16] explains why small volume concentrations affect the kinetics so strongly.

Surprisingly, this experiment shows that kinetic growth conditions and moderate Ge concentrations lead to smoother rather than rougher surface morphologies as compared to unstrained Si homoepitaxial growth. Strain-induced step-bunching, which should go the other way, does not play any role in the temperature and strain range ($\epsilon \leqslant 1\%$) investigated with set 1.

Critical thickness limitations do not allow an extension of the experiments of set 1 to higher compositions. With set 2 we therefore switched directly to growth conditions that have so far been associated with strain-induced step-bunching. For sample *A*15 we choose growth temperature (550°), Ge content (50%), layer thickness (25 Å), and substrate miscut (0.66°) to match as closely as possible the growth conditions employed in Ref. 11, where both single SiGe layers and superlattices were reported to exhibit rippled surfaces. Unfortunately, the superlattice results in Refs. 11 and 12 are not conclusive, because composition (strain) and growth temperature (kinetics) were changed simultaneously. Also, in superlattices grown under kinetic conditions the accumulated thickness of the Si spacer layers may dominate the final surface morphology. Therefore, we concentrate here on the single layer results in Ref. 11. Since strain-induced step-bunching is supposed to be an equilibrium phenomenon,[5] we approached the thermodynamically stable configuration by depositing the layer sequence of sample *A*15 at higher growth temperatures (650 °C, sample *C*15), or by *in situ* post-growth annealing (*B*15, 1 h at 550 °C). For comparison, sample *C*14 was grown at 650 °C with reduced strain ($x = 40\%$). To rule out kinetic ripple formation on the starting surface, 1000 Å Si buffer layers were grown at 550 °C and 0.5 Å/s, and then annealed *in situ* for 1 h at 750 °C. Control experiments confirmed that this procedure provides flat Si buffer morphologies,[10,11]

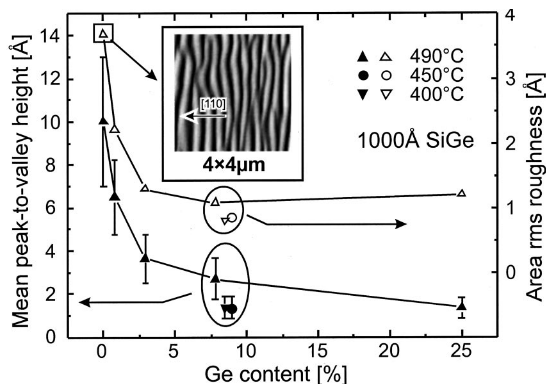

FIG. 1. Influence of increasing Ge content on surface morphology of 1000 Å thick $Si_{1-x}Ge_x$ layers deposited on a Si(001) substrate with 0.66° miscut. The inset shows the morphology of kinetic Si step bunching.

2×2 μm images of the four samples are depicted in Fig. 2. Sample *A*15 exhibits irregular corrugations of ~3 Å height and a spacing of ~700 Å. Upon post-growth annealing at the growth temperature (*B*15) elongated hut clusters with {015} facets can be identified. Growth at 650 °C directly results in square shaped hut clusters (*C*15). Both mounds and mostly square shaped hut clusters are found for a Ge content of 40% at 650 °C (*C*14).

The irregular ripples of *A*15 are not stable [Figs. 2(a) and 2(b)]. The observed morphology must therefore be the result of either a kinetic step-bunching mechanism,[7] or kinetically suppressed hut cluster formation. The elongated hut clusters of *B*15 [Fig. 2(b)]are also not stable.[17] Theoretical calculations showed that hut clusters with a square base minimize the total free energy,[18] whereas elongated hut clusters form under kinetic conditions.[19] This is corroborated by sample *C*15 [Fig. 2(b)], where a higher growth temperature resulted in a square base hut cluster morphology. Recent AFM (Ref. 17) and LEEM (Ref. 20) growth studies revealed that in SiGe layers with a low Ge content the transition to 3D islands takes place by nucleationless mounding rather than by heterogeneous nucleation events. We believe that Fig. 2(d) captures a situation where both these routes toward 3D growth are present simultaneously.

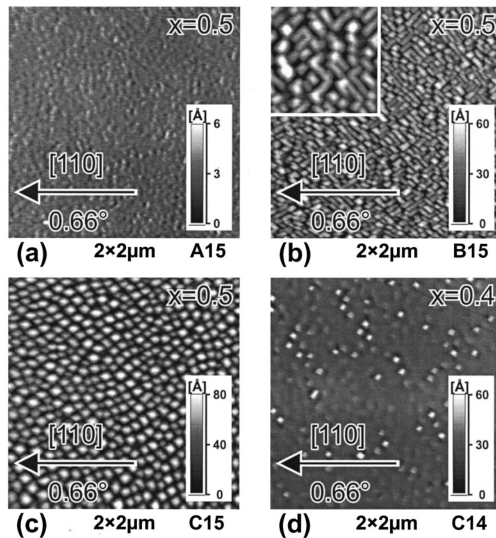

FIG. 2. Surface morphology of 25-Å-thick strained $Si_{0.5}Ge_{0.5}$ layers grown on 0.66° [110] miscut substrates. Sample *A*15 deposited at 550 °C (a); *B*15 deposited at 550 °C and annealed for 1 h at 550 °C (b); *C*15 deposited at 650 °C (c). Sample *C*14 as-grown at 650 °C with 40% of Ge (d). Arrows indicate miscut directions. The inset in (b) shows an enlarged image of 0.5×0.5 μm size.

Obviously, the small miscut angles investigated so far do not support a thermodynamically stable formation of step-bunches. Strain-induced step-bunching may, however, become observable upon decreasing the initial step spacing, i.e., by increasing the miscut angle. For that purpose we grew set 3 on substrates with a miscut of 4.34° along [100], under the same conditions as set 2. The rotation of the miscut direction is motivated by the propensity of strained SiGe layers to form {015} facets. Since their base lines are along the ⟨100⟩ directions, a miscut in the [100] direction should provide the most favorable conditions for strain-induced step bunching.

The results are shown in Fig. 3 together with AFM line scans along the miscut direction. Enhanced roughening is observed on *A*45 [Fig. 3(a)], where pronounced, 10 Å high ripples with a period of ~350 Å are found. However, this step-bunching appearance transforms during a 1 h anneal at the growth temperature into strongly elongated hut clusters [*B*45, Fig. 3(b)] with a preferential alignment parallel to the substrate steps. Thus, the ripples of *A*45 are again unstable. The line scans show that this transition is not just a breaking-up of the ripples: Additional upward steps against the miscut slope are introduced to form the favored {015} facets. At higher growth temperatures, hut clusters form already during growth with an apparent tendency toward a square base [*C*45, Fig. 3(c)].

None of our experiments gives any evidence for the formation of stable ripples on strained SiGe layers on substrates with miscuts ≤4.35°. This is in contrast to the experimental results in Ref. 11, where ~10 Å high ripples with a period of 4000 Å were reported for single, 25-Å-thick $Si_{0.55}Ge_{0.45}$ layers on a substrate with a rather small miscut of 0.4°, which was even somewhat smaller than our otherwise identical sample *A*15 of set 2. The only *way* we were able to reproduce a rippled surface as reported in Ref. 11 was by deposition of a 25-Å-thick $Si_{0.5}Ge_{0.5}$ layer, similar to the one of sample *A*15, onto a 1000-Å-thick, kinetically step-bunched Si buffer grown at 490 °C without any post growth anneal (*D*15, set 4 in Table I). Under these conditions we found in large area AFM scans the SiGe layer to basically replicate the kinetic roughness of the underlying buffer [Fig. 4(a)]. However, high-resolution AFM micrographs [insert in Fig. 4(a)] show that the as-grown SiGe layer already develops a weak small-scale roughness superimposed on the kinetic step-bunching morphology of the underlying Si buffer. Again, this morphology is unstable upon annealing at the growth temperature of 490 °C for 1 h [Fig. 4(b)]. The SiGe layer disintegrates, as all the other layers we investigated, into hut clusters, whereas the ripple morphology of the underlying Si buffer survives the annealing step. Figure 4(b) clearly demonstrates that kinetic step bunching of the Si buffer dominates the morphology, whereas the strained SiGe layer transforms into the energetically favorable hut clusters morphology, without any apparent influence of the underlying step bunches. Hence, not even a (kinetically) rippled starting surface, which could be expected to cause ideal starting conditions, promotes strain-induced step bunching.

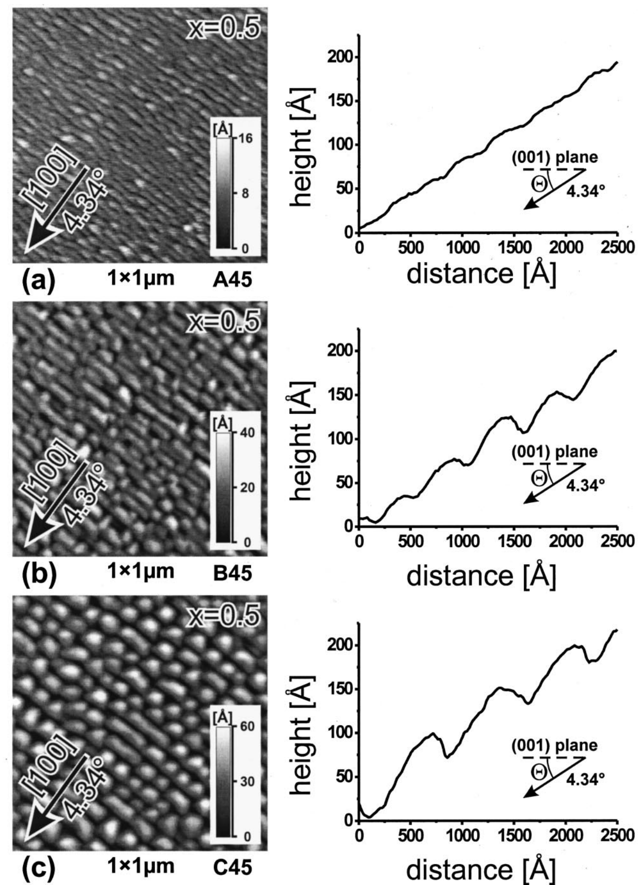

FIG. 3. Surface morphology of 25-Å-thick strained $Si_{0.5}Ge_{0.5}$ layers grown on 4.34° [100] miscut substrates. Sample *A*45 deposited at 550 °C (a); *B*45 deposited at 550 °C and annealed for 1 h at 550 °C (b); *C*45 deposited at 650 °C (c). Line scans were taken along the miscut direction.

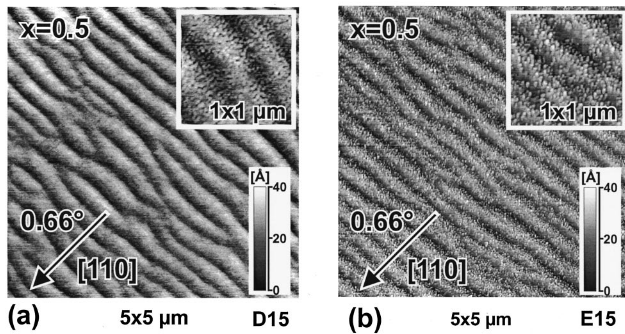

FIG. 4. Surface morphology of 25-Å-thick strained $Si_{0.5}Ge_{0.5}$ layers grown on kinetically step-bunched Si buffer layers. Both layers were grown at 490 °C on 0.66° [110] miscut substrates. Sample *D*15 as deposited (a); *E*15 additionally annealed for 1 h at 490 °C (b).

The outcome of our experiments strongly narrows the parameter space where strain-induced step bunching could affect the surface morphology. For $x \leqslant 40\%$ strain appears to be insufficient to cause step bunching,[10,11] and, in addition, even small Ge volume concentrations strongly affect the growth kinetics, and lead to smoother surfaces in the kinetic step-bunching regime. Under the well-controlled conditions of our experiments strain-induced step bunching is also absent in a strain ($\epsilon \approx 2\%$) and miscut ($<1°$) range, where its very existence has so far been claimed.[11,12] Moreover, we demonstrated that at such high strain levels near thermal equilibrium the competing mechanism of 3D hut cluster formation dominates over step bunching. By increasing the symmetry-breaking miscut to 4°, we were at least able to produce at 550 °C a surface morphology that resembles what has as yet been attributed to strain-induced step bunching. But again, the ripples are thermally unstable, and thus of kinetic origin. Finally, growing a strained SiGe layer onto a kinetically corrugated Si buffer layer leads to an evident separation of the kinetic and the strain-driven morphologies: After mild annealing the buffer remains corrugated, whereas the strained SiGe layer disintegrates into hut clusters.

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