Bending of nanoscale ultrathin substrates by growth of strained thin films and islands

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Mechanical bending is ubiquitous in heteroepitaxial growth of thin films where the strained growing film applies effectively an "external" stress to bend the substrate. Conventionally, when the deposited film is much thinner than the substrate, the bending increases linearly with increasing film thickness following the classical Stoney formula. Here we analyze the bending of ultrathin (nanometer range) substrates induced by growth of coherently strained thin films. The behavior deviates dramatically from the classical linear dependence: when the film thickness becomes comparable to the substrate thickness the bending decreases with increasing film thickness. This complex bending behavior can be understood by considering evolution of strain sharing between the film and substrate. We demonstrate experimentally such counterintuitive bending of a nanoscale thin Si substrate induced by a coherently strained Ge film, in the form of islands, grown on silicon-on-insulator substrate. Larger dome islands, representing a thicker film, induce much less bending of the substrate than smaller hut islands, representing a thinner film, in direct contrast to their behavior on thick Si. We explain these observations by properly considering the island shape and strain relaxation within the island.

Electronic and optoelectronic devices are often made from layered thin films whose mechanical properties play an important role in determining the performance, reliability, and lifetime of the device. As the size of devices shrinks toward the nanoscale, one can expect thin film mechanical properties to begin to differ from their macroscopic counterparts.[1–4] Bending of thin films, a fundamental form of mechanical deformation, is one case where differences may be significant.

Bending induced by lattice and/or thermal expansion mismatch between a thin film and a substrate is ubiquitous.[1–13] In conventional growth of a strained thin film on a bulk substrate, bending is generally calculated from the classical Stoney formula,[14] which is valid for the limit of small film thickness. This is because, in order to remain coherent (i.e., unrelaxed), the growing film in the conventional system must always be below the critical thickness for dislocation formation, i.e., much thinner than the substrate.

Figure 1 shows the predicted bending curvature (κ) from Stoney's formula, as a function of the ratio of film thickness to substrate thickness β, induced by a compressively strained nanometer-thin film on a micrometer-thick substrate. The parameters correspond to Ge film (up to 20 nm thick) growth on Si(001) (1 μm thick). The bending increases linearly with increasing film thickness; a thicker film always induces larger bending than a thinner film. A similar result would be obtained for the epitaxial growth of a tensilely strained film, except, of course, that the bending would be in the opposite direction. In both cases, before long the critical thickness for dislocation formation in the film is exceeded, and the problem is no longer an elastic relationship. Then, of course, Stoney's formula cannot be used, but, because this critical thickness occurs at a very small ratio of film to substrate thickness, no relationship other than Stoney formula was ever needed.[5,6]

If the same compressively strained film were instead deposited on an ultrathin substrate (e.g., one that is 1 – 100 nm thick) achievable in a realistic system, for example, by selec-

tive etching to release thin layers,[1] or by using thinned silicon-on-insulator substrate patterned into a free-standing cantilever,[15] Stoney's formula no longer applies, because the film thickness becomes comparable to and then larger than the substrate thickness. Using an energy minimization scheme based on continuum mechanics,[4] a general formula of bending curvature, valid for the whole range of film thicknesses, can be derived as

$$\kappa = -\frac{6(C_f \epsilon_m)t_f}{C_s t_s^2}\gamma,\qquad (1)$$

with

$$\gamma = \frac{1+\beta}{1+4\alpha\beta+6\alpha\beta^2+4\alpha\beta^3+\alpha^2\beta^4},\qquad (2)$$

where $\alpha = C_f/C_s$, C_f and C_s are, respectively, elastic constants (related to Young's modulus and Poisson ratio) of the film and substrate, and $\beta = t_f/t_s$ is the ratio of film thickness (t_f) and substrate thickness (t_s). Note for $\beta \ll 1.0$, $\gamma = 1.0$ and Eq. (1) reduces to Stoney's formula.

Figure 2 shows the calculated bending curvature for a film grown on a very thin substrate, using Eqs. (1) and (2). It deviates dramatically from the classical linear behavior. Initially, for $\beta \lesssim 1.0$, the bending curvature, κ, increases nearly linearly with increasing film thickness (i.e., with increasing β), the same as for growth on a thick substrate. However, because the substrate is now thinned to the nanoscale, the film thickness very soon becomes comparable to the substrate thickness, while still maintaining an epitaxial (i.e., strained, not relaxed) relationship to the substrate. Figure 2 shows that the curvature κ does not continue to increase monotonically, but instead reaches a maximum and then decreases. If the substrate is thin enough, it is possible to reach and exceed the maximum in curvature without reaching the critical thickness for dislocation formation. [The position of the maximum, here $\beta \sim 0.6$, depends on the weighting func-
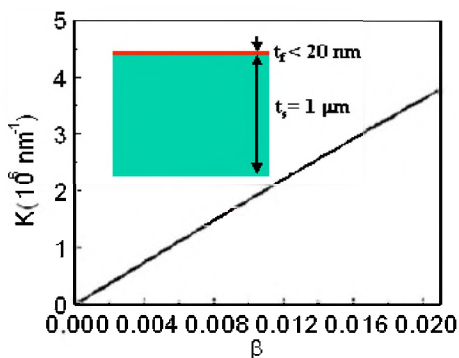
FIG. 1. (Color online) Bending curvature induced by a nanometer-thick strained film grown on a micrometer-thick substrate, calculated from Stoney's formula. A thicker film always induces larger bending than a thinner film. Notice that β is always very small.
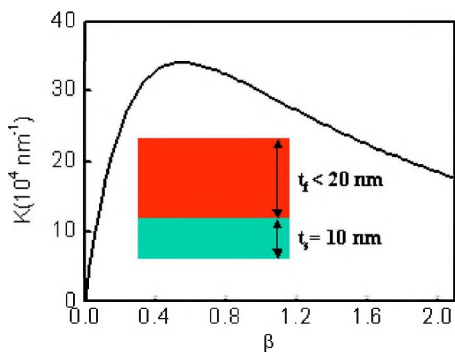

FIG. 2. (Color online) Bending curvature induced by a compressively strained film grown on an ultrathin substrate, calculated from Eq. (1). In contrast to Fig. 1, a thicker film may induce smaller bending than a thinner film.
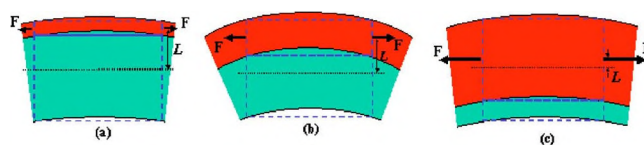

FIG. 3. (Color online) Schematic illustration of the force (F) and torque $(F \times L)$ applied by a compressively strained film (red) grown on a substrate (green) for a different range of film-to-substrate thickness ratio (β).

tion, $\gamma(\beta)$ in Eq. (2).] Such a dependence, for thin substrates, of bending curvature on film thickness, in particular the existence of a maximum, makes the bending induced by a sufficiently thick (but still epitaxial) film smaller than that induced by a thinner film.

The bending behavior described in Fig. 2 does not occur for conventional film growth on thick substrate because the film to substrate thickness ratio must remain small to keep film coherency. But it does have a classical analog in bending of bimetal strips, where the thicknesses of the two metal strips can be comparable. The same general formula [in a slightly different form from Eqs. (1) and (2)] had been derived by Timoshenko[16] for bending of bimetal strips over the whole range of thickness ratio. However, the complex dependence of bending on thickness ratio, in particular the counterintuitive sequence of the bending-to-unbending with increasing film thickness, was not discussed, although it is implicitly contained in the formula.

The bending-to-unbending occurs only when the film thickness becomes comparable to substrate thickness. This requires use of ultrathin substrates to maintain the film coherency, which has become only possible with the recent development of thin film growth technology, such as by growth of very thin semiconductor bilayer films released

from the selective etching process.[1,2] One such experiment[1,2] on rolled-up GaAs/InAs nanotubes has, in particular, shown direct evidence in support of the theoretical prediction in Fig. 2. The diameter of the roll-up GaAs/InAs tubes increases (i.e., the bending curvature decreases) with increasing β when 1.4-ML- to 17.4-ML-thick GaAs film is deposited onto an ultrathin 1.4-ML-thick InAs substrate (see Fig. 4 in Ref. 2). This behavior for large β (1–12) is in direct contrast to that for small $\beta(\ll 0.01)$[5,6] where the bending curvature increases with increasing β. However, we have not found an experiment to cover the whole range of β for both $\beta \ll 1.0$ and $\beta > 1.0$ regions so that the existence of a maximum bending curvature can be located.

A better understanding of bending of nanoscale thin films has important practical implications in thin-film nanotechnology. For example, the principle of nanoscale thin film bending can be used for producing a variety of nanostructures, such as nanotubes, via nanomechanical architectures.[15] While Eq. (1) gives the dependence of nanotube radius (inverse of curvature) on film-to-substrate thickness ratio, Fig. 2 actually tells that there exists a smallest nanotube radius (corresponding to the maximum bending curvature) one can produce for a given set of film and substrate combination. It will be interesting for future experiments to confirm such a limit.

Although the complex bending behavior is implicitly contained in the classical bending formula of bimetal strips[16] and has been partly shown in the bending of ultrathin bilayer semiconductor films,[2] a good understanding of the physical mechanism underlying the bending-to-unbending sequence is still lacking, which we discuss below.

When a compressively strained film grows on a substrate, its lattice relaxes outward, which applies both a *force* (*stress*) and a *torque* (*moment*) to the substrate, the former tending to expand the substrate uniformly and the latter to bend the substrate downward. The magnitude of the force is proportional to the misfit strain and the film thickness $(F \sim \epsilon t_f)$; the magnitude of torque equals force (F) times the distance (L) from the mean position of the applied force (middle of the film) to the line of the center of the whole system [middle of the (film+substrate)], as illustrated in Fig. 3.

For very small β $(t_f \ll t_s)$, the film applies a relatively small force but a large torque because of a large L, as shown in Fig. 3(a). Consequently, the film bends the substrate without much expansion. The bending increases with increasing film thickness (i.e., force) as $L \approx t_s/2 \gg t_f$ remains almost constant. For very large β $(t_f \gg t_s)$, the film applies a large force but a small torque because of a very small L, as shown in Fig. 3(c). Consequently, the film expands the substrate
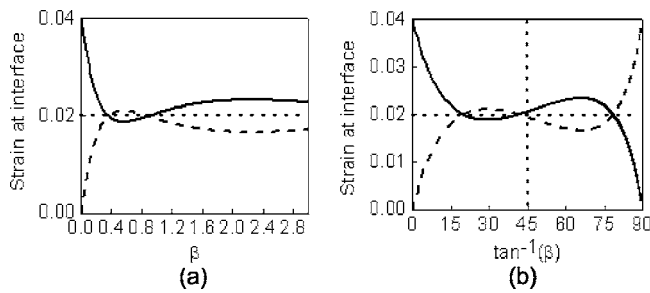
FIG. 4. (a) Strain in the film (solid) and in the substrate (dashed) at the film/substrate interface as a function of β. The initial increase of strain sharing at the interface up to $\beta \sim 0.6$ correlates closely with increasing bending in Fig. 2. The later increase of strain sharing is achieved mainly through uniform expansion. (b) The same plot as in (a) but as a function of $\tan^{-1}(\beta)$, showing the role of reversal of film and substrate beyond $\beta = 1$.

uniformly without much bending. The bending decreases with increasing film thickness because the torque decreases as force is applied more and more closely to the center of the whole system (diminishing L). At intermediate β ($t_f \sim t_s$), the film bends as well as expands the substrate, as shown in Fig. 3(b). Because the bending increases at very small β but decreases at very large β, with increasing β there must exist a maximum bending at an intermediate β.

We may understand further the bending behavior in terms of strain relationships (sharing). The compressive strain in the film is relaxed at the expense of introducing a tensile strain in the substrate, i.e., by sharing strain with the substrate, through both bending and uniform expansion. Initially, at low coverage, strain is mostly shared by the mechanism of bending, while later, at high coverage, it is by the mechanism of uniform expansion. To illustrate this point quantitatively, we plot in Fig. 4 the strain sharing at the film/substrate interface, i.e., the amount of compressive strain reduction in the film and the amount of tensile strain increment in the substrate at the film/substrate interface. The same would be true for a tensile film grown on a thin substrate, except the bending would be in the opposite direction.

As a model, we chose values corresponding to Ge film growth on Si(001). In this case, the compressive strain in the film at the interface starts (i.e., at zero thickness) at ~ -0.04, much larger than the tensile strain in the substrate (~ 0). As the film thickness increases, the compressive strain in the film (absolute value) decreases while the tensile strain in the substrate increases. By comparing Figs. 2 and 4, we see that for very thin films (below $\beta \sim 0.6$), bending curvature and strain sharing increase in parallel with increasing β, indicating that strain is shared mostly through bending, corresponding to Fig. 3(a). For thicker films (above $\beta \sim 2.4$), the bending continues to decrease but the strain sharing increases again, indicating that at later stages of large β, strain is shared mainly through increasing uniform expansion, corresponding to Fig. 3(c). In the intermediate range of β ~ 0.6–2.4, strain at the interface remains almost constant due to the interplay between bending and uniform expansion, corresponding to Fig. 3(b).

Beyond $\beta = 1.0$, the film becomes thicker than the substrate and in effect the roles of (conventional) substrate and
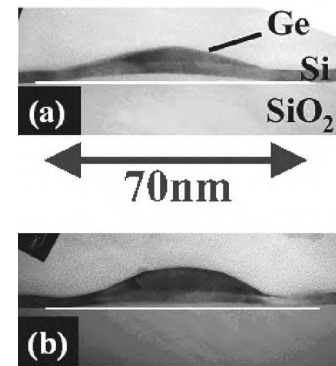

FIG. 5. TEM images of growth of coherent 3D Ge islands on SOI substrate, demonstrating the different bending induced by (a) a hut and (b) a dome. The bending induced by the hut is one order of magnitude larger than that by the dome.

film become reversed. The effect of this reversal on the trend of strain sharing can be seen more clearly by plotting the strain at the interface as a function of $\tan^{-1}(\beta)$ instead of β, as shown in Fig. 4(b). The curves are almost symmetric about the point of $\tan^{-1}(\beta) = 45°$, where the magnitude of strain in the film equals that in the substrate (perfect strain sharing). (The curves would be exactly symmetric if the elastic constants of Ge and Si were the same.)

Next, we illustrate such counterintuitive bending phenomenon in the context of growth of strained Ge islands on silicon-on-insulator (SOI) substrate with an ultrathin Si template layer. The bending of the Si template layer induced by domes is much smaller than that induced by huts, even though the average thickness of the strained layer is greater for the dome. We explain this behavior by extending the continuum-mechanics calculations to include the effect of island shape and strain relaxation within the island.

The growth of Ge film on Si is a classical example of Stranski-Krastanov growth, in which the deposited film does not have to be a film *per se*: as Ge is deposited on Si(001) first small coherent three-dimensional (3D) islands called "huts" form,[17] which transform to larger coherent islands called "domes" as the deposited amount increases.[17–19] When growing on a thick Si substrate, domes, representing a thicker film, induce larger bending than huts,[5,6] representing a thinner film.

Recently, it has been shown[3,4] when Ge hut islands are grown on thin SOI substrates, they induce local bending of the Si template layer underneath each hut rather than bending the template layer uniformly, as they do on bulk substrates. This local bending is large enough to be easily observable with transmission electron microscopy (TEM) on cross-section samples and therefore we are able to obtain quantitative measurements of the bending curvature. Domes can be analyzed in the same manner. The experiment and growth conditions have been described earlier.[4] Figures 5(a) and 5(b) show TEM images of a hut and a dome, respectively, grown on a SOI substrate,[20] with a Si template layer thickness of 10 nm. The images demonstrate the highly reduced bending induced by the dome relative to the hut.

The height of the hut is 10 nm, corresponding, because of its shape, to a local nominal Ge film thickness of ~ 3 nm
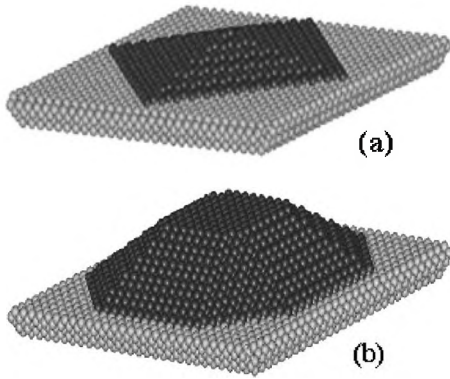
FIG. 6. Atomic model of simulating a Ge island (black) on a Si (001) substrate (gray). (a) Hut with bases along (100) directions. (b) Dome with bases in (100) and (110) directions.
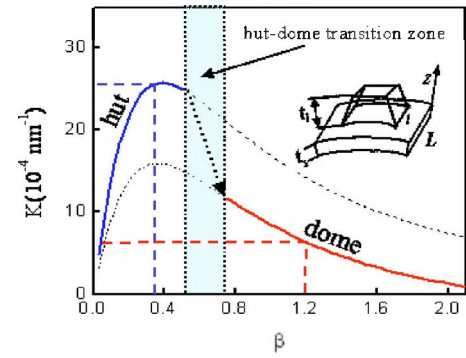

FIG. 7. (Color online) Average bending curvature induced by a Ge hut (solid blue line) and a Ge dome (solid red line), when they grow on a 10 nm thin Si substrate. The dashed red and blue lines mark the bending induced by a dome and a hut, respectively, if they had the observed sizes in Fig. 5. The dashed black lines indicate what bending would be if the dome or hut could have formed in these regions. The inset shows the dome geometry used for the calculation.

above the Ge wetting layer, which may itself be ~ 0.5 nm thick; the height of the dome is ~ 20 nm, corresponding to a local nominal Ge film thickness of ~ 12 nm. Conventional wisdom (Stoney's formula) would predict that, based on the corresponding nominal thicknesses, domes induce a local bending ~ 4 times larger than huts. In contrast, we estimate from Fig. 5 that the dome induces an average local bending curvature almost one order of magnitude smaller, $\kappa \sim 5 \times 10^{-4}$ nm^{-1}, versus $\kappa \sim 3 \times 10^{-3}$ nm^{-1} for the hut.

Therefore the relative bending induced by dome vs hut on an ultrathin Si substrate behaves in an opposite way to that on a thick bulk Si substrate.[5,6] This is qualitatively confirmed by atomistic simulations,[21] which were performed using the same interatomic potentials and simulation scheme as in the previous studies of SiGe thin films.[4,21,22] Figures 6(a) and 6(b) show the atomic models of simulation systems containing a hut and a dome sitting above a Si(001) film, respectively. The hut, a pyramid, is square-based having four (105) side facets,[17] the dome is octagon-based having a (001) top facet, four (113) and four (102) side facets.[18,19] We choose an island height of ~ 7 Å for the hut [Fig. 6(a)] and ~ 14 Å for the dome [Fig. 6(b)] and an ~ 6 Å thick Si film to mimic closely the relative scales of experiment and to capture the physical condition that the thickness of Ge islands and Si film is comparable. The lateral dimension of the Si substrate is chosen as 110 Å $\times$ 110 Å. These amount to ~ 5000 and ~ 8000 atoms for the hut [Fig. 6(a)] and the dome system [Fig. 6(b)], respectively.

The equilibrium bending curvatures induced by the hut and dome are determined by total energy minimization, relaxing the whole system until forces diminish on all atoms. Quantitatively, the average bending curvatures induced by the hut, in the cross-sectional plane through the center of island along the island base in three high-symmetry (110), ($\bar{1}$10), and (100) directions (see caption of Fig. 6), are calculated to be 1.8, 2.7, and 4.7×10^{-4} nm^{-1}, respectively. The corresponding values for dome are 1.6, 2.0, and 3.5×10^{-4} nm^{-1}. Evidently, the smaller hut induces consistently a larger bending curvature than the larger dome in all directions.

The atomistic simulations were limited to selected islands scaled down to much smaller sizes, according to relative ex-

perimental sizes. For a more realistic quantitative comparison, the bending behavior of dome vs hut can be understood based on the above analysis of bending of an ultrathin substrate induced by growth of strained film of comparable thickness, except the film is represented in the form of islands. The observed large bending curvature induced by the hut [Fig. 5(a)] indicates that the Si substrate behaves effectively like a freestanding film on SOI substrate;[3,4,20] it can be estimated by continuum-mechanics calculations using a modified Timoshenko's formula by taking into account the actual island geometry and strain relaxation.[4]

Strain relaxation within an island can be treated approximately by assuming the island is completely coherent at the interface ($z=0$) and completely relaxed at the top, $z=t_i$, and misfit strain gradually decreases from ϵ_m (misfit strain between Ge and Si) at $z=0$ to 0 at $z=t_i$ (see inset of Fig. 7). The average bending curvature induced by a pyramidal hut with quadratic strain relaxation[23] has been derived to follow the same formula as Eq. (1), but with a different weighting function γ for hut as[4]

$$\gamma = \frac{9/10 + (6/5)\beta - (3/20)\alpha\beta^2}{1 + 4\alpha\beta + 9\alpha\beta^2 + (54/4)\alpha\beta^3 + (81/20)\alpha^2\beta^4}, \quad (3)$$

where $\beta = t_f/t_s$ is the ratio of nominal Ge film thickness (t_f) to Si template layer thickness (t_s): For a pyramidal island shape, $t_f = t_i/3t_s$, where t_i is the island height.

Similarly, the average bending curvature induced by a dome island can be estimated. As is well-known, domes, due to their steeper facets, relax strain more effectively than huts.[5,6] We therefore assume a linear rather than a quadratic strain relaxation within the dome. For simplicity, we consider the dome having a truncated pyramidal shape with base dimension of $l \times l$, top dimension of $\frac{1}{2}l \times \frac{1}{2}l$, height t_i, and contact angle of $\sim 25°$ corresponding to ($\bar{1}$13)/(102) facet, as shown in the inset of Fig. 7. We obtain a different weighting function γ for the dome as

$$\gamma = \frac{0.61 + 0.56\beta - 0.26\alpha\beta^2}{1 + 4\alpha\beta + 8.08\alpha\beta^2 + 8.06\alpha\beta^3 + 2.62\alpha^2\beta^4}, \quad (4)$$

where $\beta = t_f/t_s$, for the given truncated pyramidal island shape, $t_f = 7t_i/12t_s$. We note that simplification of the actual island shape and different assumption of strain relaxation within the island will not qualitatively change the results and discussion below.

Figure 7 shows the calculated bending curvature as a function of β, the ratio of nominal film thickness to substrate thickness, for the film represented by a hut (upper curve) vs by a dome (lower curve). For a direct comparison to experiment, we use a Si layer thickness of 10 nm and the elastic constants of Si and Ge (Ref. 24) and calculate the curvatures of deposited-film thicknesses from zero up to twice as large as the substrate thickness ($\beta = 2$).

The average bending induced by either hut or dome is qualitatively the same as that induced by a film, as shown in Fig. 2. Initially, i.e., for $\beta \lesssim 0.1$, the bending curvature, κ, increases nearly linearly with increasing film thickness (i.e., with increasing β), the same as for growth on a thick substrate. However, because the substrate is now thinned to the nanoscale, the nominal film thickness very soon becomes comparable to the substrate thickness. Therefore the curvature κ does not continue to increase monotonically, but instead reaches a maximum and then decreases. [The position of the maximum, here $\beta \sim 0.4$, depends on the complex weighting function, $\gamma(\beta)$.] Such a dependence, for thin substrates, of bending curvature on overlayer film thickness, in particular the existence of a maximum, makes the bending induced by a larger dome island (or a thicker film) smaller than that induced by a smaller hut island (or a thinner film).

Notice that huts do not exist over the whole range of β of interest, in fact, only a small part of low β. They transform into domes. Conversely, domes do not exist at low β. As the film thickness reaches $\beta \sim 0.5 - 0.8$, the film becomes represented by domes that have transformed from huts. Also, dome and hut cannot be put on the same bending curve because of different island shape and different degree of strain relaxation within the island. Figure 7 shows that the dome bends the thin Si template much less than does the hut. Using the experimental island heights in Fig. 5, we estimate from Fig. 7 that the hut induces a bending curvature of $\kappa \sim 2.5 \times 10^{-3}$ nm^{-1} at $\beta \sim 0.33$ while the dome induces a bending curvature of $\kappa \sim 5.5 \times 10^{-4}$ nm^{-1} at $\beta \sim 1.2$, in good agreement with experiments.

The fact that domes can relax more strain within the island cannot explain the observation that a dome induces less bending than a hut. In fact, on a thick substrate, domes always induce larger bending than do huts.[5,6] Our calculations confirm this observation. For example, using Eqs. (1), (3), and (4) and a *hypothetical* Si layer thickness of 1 μm, we calculated bending curvatures as a function of β induced by a hut (quadratic relaxation) and a dome (linear relaxation), as shown in Fig. 8. The bending increases linearly with increasing β for both the hut and dome; the dome line has a somewhat smaller slope because a dome produces a greater amount of strain relaxation. Because the dome exists only in the regime of larger β, it is always observed to induce a
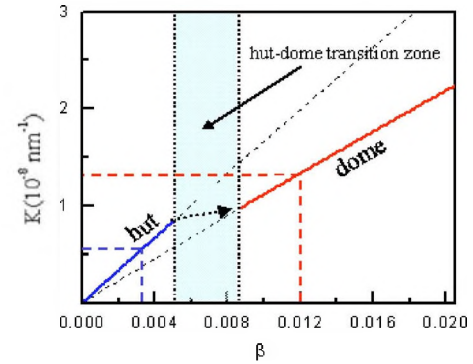

FIG. 8. (Color online) Average bending curvature induced by a quadratically relaxed hut and a linearly relaxed dome if they grew on a 1 μm thick substrate. Notations are the same as in Fig. 2. A dome generally induces larger bending than a hut.

larger bending than the hut on a thick substrate. Using Fig. 8 and the island heights in Fig. 5, the bending induced by a dome of the experimentally observed size ($\kappa \sim 1.3 \times 10^{-8}$ nm^{-1}) would be more than two times larger than that induced by a hut of the observed size ($\kappa \sim 0.55 \times 10^{-8}$ nm^{-1}), if they had grown on a 1 μm thick Si substrate.

In conclusion, we demonstrate a bending behavior unique to nanometer-thin stressed substrates, where a thicker film (a larger island) may induce a much smaller bending than a thinner film (a smaller island). We explain this behavior by the maximum achievable strain sharing. When a thin film (island) grows toward a thickness comparable to the substrate thickness, the increasing amount of strain sharing at the interface leads to a complex dependence of bending curvature on film thickness. The bending first increases at low coverage, then slows down to reach a maximum, and thereafter decreases with increasing film thickness. The reason this behavior is not observable for thick substrates is that for thick substrates β is always small. Growth of a thicker film (to increase β) leads to the activation of a strain relaxation mechanism, dislocation formation, and loss of coherency as the critical thickness is exceeded. We are able to observe it even though our substrate, the Si template of SOI(001), is not free-standing because the shear stress induced by huts and domes allows the oxide to lower its viscosity and flow.[20]

Our analysis not only provides insights to the understanding of bending of nanoscale thin films but also has important implications in thin-film nanotechnology, such as in fabrication of nanostructures and in growth of compliant thin films. The bending behavior we describe should be apparent in any "coherent" film system in which the substrate is very thin, e.g., high-aspect-ratio structures in advanced semiconductor patterning or NEMS (nanoelectromechanical system) devices. It may therefore have important implications in such devices, as the mechanical deformation in turn changes electronic and electrical properties.

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