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Critical Stresses for Si_xGe_{1-x} Strained-Layer Plasticity

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We have measured the temperature-dependent onset of strain relief in metastable Si_xGe_{1-x} strained layers grown on Ge substrates. On the basis of these measurements, and physical arguments, we propose that strained-layer breakdown is most directly determined not by thickness and lattice mismatch, but rather by (1) an "excess" stress (the difference between that due to misfit strain and that due to dislocation line tension) and (2) temperature. With use of these parameters, observed regimes of stability and metastability are shown to be described within a simple, unified framework.

Strained epitaxial films, first studied theoretically nearly four decades ago,[1] have attracted much interest recently. Partly, this interest stems from observations of structural metastability in films grown by state-of-the-art techniques. In this regard, an outstanding question has been how to correlate growth conditions with subsequent structural perfection of the film. The original equilibrium theories of Ball and van der Merwe,[2] Matthews and Blakeslee,[3] and co-workers predicted that, below a critical thickness, lattice mismatch between substrate and film would be accommodated entirely by film strain. Above this thickness, film strain would be partly relieved by misfit dislocations.

The pioneering work of Kasper,[4] Bean,[5] and co-workers in the SiGe system showed, however, that under some growth conditions strain in films above the critical thickness is *not* measurably relieved. Only above a second critical thickness does measurable relief occur, and even then, the amount of relief is not in accord with equilibrium theory. Most recently, the work of Fritz[6] and of Dodson and Tsao[7] suggests that the observed metastability can be explained by sluggish plastic deformation rates accompanied by a finite experimental resolution. The second critical thickness is that for which strain relief is just sufficient to be observable.

A full treatment of the kinetics of plastic deformation of thin epitaxial films, however, is nontrivial. Even deformation of bulk materials occurs by a number of complex mechanisms, and little is known about whether deformation in thin films occurs by the same mechanisms. Nevertheless, it is clear that *any* mechanism must be governed principally by the two parameters shear stress (the driving force for deformation) and temperature. Indeed, for bulk materials, deformation rates can be elegantly expressed with the stress-temperature diagrams (or "deformation mechanism maps") introduced by Frost and Ashby.[8]

In this Letter, we argue that the stability and metastability of thin strained layers is determined mainly by the kinetics of plastic deformation and hence is governed by stress and temperature. However, we propose that the stress which actually drives dislocation motion is the difference between the usual stress due to misfit strain and an "effective" stress due to dislocation-line tension. Observable strain relief occurs only if this "excess" stress exceeds a critical value which depends on temperature. Furthermore, we report the first measurements of the temperature dependence of such critical excess stresses, in the Si_xGe_{1-x}/Ge system. Finally, we show how the concept of excess stress can be used to construct a stress-temperature stability diagram which unifies present and previous results on Si_xGe_{1-x} strained-layer stability and metastability.

Our Si_xGe_{1-x} alloy layers were grown by molecular-beam epitaxy over a range of temperatures for various alloy fractions and thicknesses. Unlike most previous investigations, the substrates were (001) Ge, rather than (001) Si, and the alloys were Ge, rather than Si, rich. We find that these choices maximize the temperature window for epitaxial growth: Film quality could be maintained to lower growth temperatures in these Ge-rich films without affecting measurably the onset of islanding[9] at higher growth temperatures. Indeed, the measurements reported here span a wider temperature range than any to date.

Our growth chamber has a base pressure of 3×10^{-10} Torr, rising typically to 3×10^{-9} Torr during deposition. Substrates were prepared by sequential pad polishes in Br: methanol and methanol.[10] Final *in situ* cleaning consisted of a 20-min 750°C anneal, followed by the deposition at 750°C of 10 Å of Ge at very low (0.03 Å/s) rates. Growth of the alloy layers was preceded by growth at 550–575°C of approximately 1000-Å-thick Ge buffer layers, and generally capped by 500–1000 Å of Ge as well. Typical growth rates were 0.3–0.4 Å/s.

Film quality was measured by axial [001] and [011] ion channeling with use of 2-MeV He^+. Typical [001] minimum yields (X_{min}'s) were ≈ 0.04—indistinguishable from those of the starting substrates. Thicknesses and misfits[11] for a series of alloy films grown at 494°C are shown in Fig. 1. The filled circles correspond to films in which [001] dechanneling is indistinguishable from that
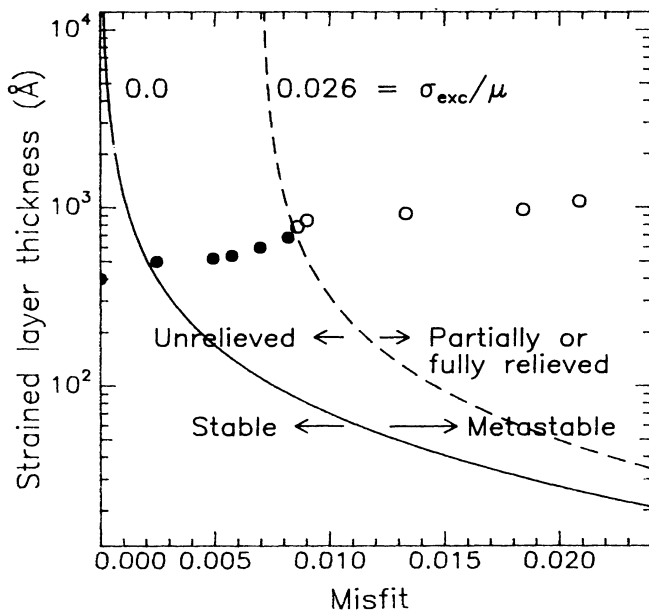
FIG. 1. Thicknesses and misfits for a series of Si_xGe_{1-x} films grown at 494 °C. Fully strained (filled circles) and partially strain-relieved (open circles) films are separated by the $\sigma_{exc}/\mu = 0.024$ isobar. The $\sigma_{exc}/\mu = 0$ isobar separates absolutely stable from metastable films.

in the starting substrate, implying strain relief less than $\approx 1 \times 10^{-3}$.[12] These films are essentially fully strained. The open circles correspond to films in which [001] dechanneling is noticeably poorer, implying strain relief greater than $\approx 1 \times 10^{-3}$. It can be seen that pseudomorphic films may be grown to greater thickness-misfit combinations than predicted by equilibrium theory, in agreement with previous measurements on Ge_xSi_{1-x} films grown at 550 °C.[13] Indeed, it has been shown that, for the Ge_xSi_{1-x}/Si system, the experimental thickness versus mismatch boundary separating partially strain-relieved from fully strained films may be described exceedingly well by modification of the mathematical form of conventional equilibrium theory so as to offset the curve toward higher misfits.[14]

Here, we suggest that the *physical* basis for such an offset is that strained-layer breakdown requires a fixed, *but nonzero,* excess film stress. To see this, consider the action of the stress field due to biaxial film strain on a 60° dislocation threading through a diamond-cubic strained layer. The glide component of the Peach-Kohler "misfit-strain" force[15] parallel to the (001) film plane, integrated over the length of such a dislocation, is

$$F_{M,\mathrm{par}} = \tfrac{1}{2} hb\sigma_M [1/\sqrt{2}, -1/\sqrt{2}, 0], \qquad (1)$$

where h is the film thickness, $\mathbf{b} = (a_0/\sqrt{2})[1/\sqrt{2}, 0, 1/\sqrt{2}]$ is the Burgers vector, a_0 is the alloy lattice constant, the dislocation lies along the unit vector $\hat{\xi} = [0, 1/\sqrt{2}, 1/\sqrt{2}]$,

$\sigma_M \approx 2e\mu(1+v)/(1-v)$ and e are the principal in-plane stresses and strains, respectively, μ is the shear modulus,[16] and $v \approx 0.3$ is Poisson's ratio.

Because there is no stress field acting on the dislocation in the (nearly) unstrained substrate, the dislocation must elongate along the substrate-film boundary in order to move laterally in the film above. As originally noted by Matthews, this elongation generates an opposing force due to the "image" (or self) energy[17] of the dislocation,

$$F_{D,\mathrm{par}} = \tfrac{1}{2} hb\sigma_D, \qquad (2)$$

where we have introduced an "effective" stress

$$\sigma_D \approx \frac{\mu}{2\pi} \left[\frac{1 - v\cos^2\beta}{1-v} \right] \frac{\ln(4h/b)}{h/b}, \qquad (3)$$

with $\beta = 60°$. Note that this is a fictitious stress, which does not arise from an actual stress field acting on the dislocation.

The difference between these two forces is the net, or excess, force on the dislocation, $F_{exc} = hb\sigma_{exc}/2$, where we have introduced an excess stress

$$\sigma_{exc} = \sigma_M - \sigma_D$$

$$= 2e\mu \frac{1+v}{1-v} - \frac{\mu}{2\pi} \left[\frac{1 - v\cos^2\beta}{1-v} \right] \frac{\ln(4h/b)}{h/b}. \qquad (4)$$

This excess stress is a measure of the driving force for strain relief,[18] *and hence both for the deviation from equilibrium and for the degree of metastability.* Two contours of constant excess stress ("isobars") are shown in Fig. 1, corresponding to thickness-mismatch combinations which result in the same excess stress, and hence the same deviation from equilibrium. If this stress is less than or equal to zero, then there is no net force driving dislocation motion; this is the original Matthews-Blakeslee criterion for absolute film stability, shown by the $\sigma_{exc}/\mu = 0$ isobar in Fig. 1. If this stress is greater than zero, then there is a net force driving dislocation motion.

Even for a positive net excess stress, however, dislocations do not necessarily move freely. On a given experimental time scale, there must be enough excess stress to lead to *observable* plastic deformation. For the films shown in Fig. 1, the excess stress at which partial strain relief becomes observable can be seen from the second isobar to be $\sigma_{exc}/\mu = 0.024$. We therefore identify this excess stress as *the critical excess stress for strained-layer breakdown* at 494 °C. Note that the effect of requiring such a nonzero excess stress for observable strain relief is equivalent, as mentioned above, to offsetting linearly the equilibrium curve toward higher misfits, and can be shown to fit reasonably well the thickness versus mismatch boundary measured by Bean and co-workers.

Using dechanneling measurements similar to those described above, we have deduced critical excess stresses
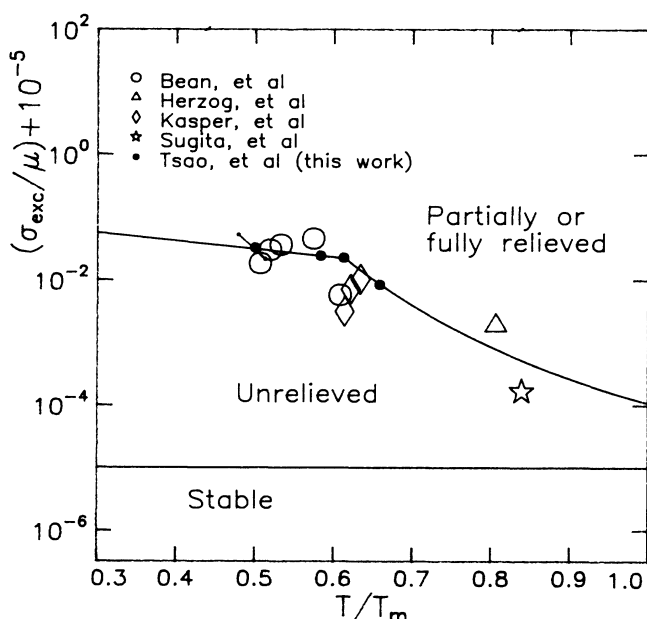
FIG. 2. Excess stress (scaled by the shear modulus, μ) vs temperature (scaled by the melting temperature, T_m) diagram demarcating the observed stability and metastability regimes for Si_xGe_{1-x} epitaxial strained layers. To allow the absolutely stable ($\sigma_{exc}/\mu \leq 0$) regime to be shown on a logarithmic scale, an offset of 10^{-5} has been added to the scaled excess stress. The data points (filled circles) are taken from this work; except as indicated for the lowest-temperature data point, error bars are less than the size of the points. Also plotted are data from other sources, as noted in the text. The curve separating the unrelieved from the partially or fully relieved regions is a fit to our data; the different behaviors observed at low (low-temperature plasticity) and high (power-law creep) temperatures are consistent with those observed for bulk deformation.

over a range of temperatures, as summarized in Fig. 2. To take into account in an approximate way differences between plastic deformation rates in films of different compositions, the growth temperatures have been normalized to the melting temperatures[19] of the films. Films grown at temperatures of 395 and 534 °C show critical excess stresses comparable to that for the 494 °C data, with a definite (though weak) trend towards higher critical excess stresses at lower temperatures. This trend is consistent with our expectation that dislocations are less mobile at lower temperatures and hence require a greater driving force in order to move at comparable rates.

At a growth temperature of 568 °C, however, films showed a significantly lower ($\sigma_{exc}/\mu \approx 0.0085$) critical excess stress. This result suggests a change to a different mechanism for plastic deformation at high temperature, e.g., from low-temperature plasticity to high-temperature power-law creep. Indeed, it is not possible to fit our data by a smooth, simply activated power-law curve,

as would be expected for a single deformation mechanism. Rather, at least two mechanisms are required,[20] as illustrated by the kinked solid curve drawn in Fig. 2.

Also shown in Fig. 2 are data taken from previous measurements in the SiGe system by Bean et al.,[21] Kasper and Herzog,[22] Sugita, Tamura, and Sugawara,[23] and Herzog, Csepregi, and Seidel.[24] Although the growth conditions, film thicknesses, and diagnostics techniques use in these previous measurements vary considerably, *the data are remarkably consistent with ours when plotted in terms of temperature-dependent critical stresses rather than in terms of misfit-dependent critical thicknesses.* We believe this consistency to be a strong argument for the idea that, to first order, excess stress and temperature govern the metastability of strained SiGe films.

Figure 2 is remarkably reminiscent of the deformation mechanism maps[25] referred to earlier, which plot isostrain-rate contours on a stress-temperature diagram. On such a map, the boundary between the unrelieved and partially relieved films would be, to first order, the isostrain-rate contour corresponding to just observable strain relief on ordinary growth time scales. Indeed, our data, when transposed to deformation-mechanism maps for bulk Si or Ge, are fitted reasonably well by the isostrain-rate contours (in the range $10^{-5}-10^{-7}$ s^{-1}) required by our experimental growth time scale. Moreover, our observation of qualitatively different behavior above and below $T/T_m \approx 0.6$ is also reproduced by bulk deformation-mechanism maps for Si and Ge. These similarities suggest a close analogy between Fig. 2 and bulk deformation-mechanism maps, and that the microscopic mechanisms for bulk and thin-film plastic deformation may not be very different.

Finally, it is also possible to use Fig. 2 as a "stability diagram" in which the various regimes of stability and metastability are demarcated.[26] Films with excess stress less than zero (the van der Merwe-Matthews boundary) are absolutely stable. Films with excess stress greater than zero, but less than that required for plastic deformation on the experimental time scale (the Kasper-Bean boundary),[27] are not observably strain relieved (although they may contain defects), and hence are metastable. Films with yet greater excess strains are observably strain relieved, although they may still be metastable.[28]

In conclusion, we have proposed that excess stress and temperature are the key parameters governing structural stability and metastability in strained epitaxial films. The idea is the same as that which has proven so fruitful in describing plastic deformation in bulk materials, except that the concept of stress must be extended to include both that required to accommodate misfit strain, and that associated with dislocation self-energy. Our measurements over a wide temperature range and under carefully controlled growth conditions, together with previous measurements under very different growth condi-

tions, support this idea. Finally, we have introduced stress-temperature stability diagrams, which provide a simple, unified description of the stability and metastability regimes of strained layers.

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[15]$d\mathbf{F}_M = (\mathbf{b} \cdot \mathbf{\Sigma}) \times \hat{\xi}$, where

$$\mathbf{\Sigma} = \begin{bmatrix} \sigma_M & 0 & 0 \\ 0 & \sigma_M & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

is the stress tensor.

[16]Values for the shear moduli for Si and Ge may be found in Frost and Ashby, Ref. 8.

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