Commensurate and incommensurate structures in molecular beam epitaxially grown Ge_xSi_{1-x} films on Si(100)

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The transition between commensurate and incommensurate growth of Ge_xSi_{1-x} alloys on Si is observed directly by means of ion channeling and x-ray diffraction measurements. Molecular beam epitaxial films of thickness h up to 2500 Å thick show commensurate epitaxy for $x \lesssim 50\%$ and $h \lesssim h_c$, a critical thickness dependent upon x. The observed values of h_c are discussed in terms of a model invoking the maximum theoretical interfacial shear strength and a barrier to misfit dislocation formation.

Epitaxial thin films of Ge_xSi_{1-x} alloys on silicon substrates form heterostructures with a continuously variable lattice-parameter misfit up to 4.2%. We present a structural analysis of samples grown by molecular beam epitaxy (MBE) for which the growth morphologies and optimal growth temperatures are reported in Ref. 1. Such films are of interest as the first layer in the preparation of strained-layer superlattices.[2,3] Pioneering work by Kasper *et al.*[4] in the region $x \lesssim 15\%$ found critical thicknesses for pseudomorphic or coherent growth, h_c, which were a factor of 4 above the prediction of Frank and van der Merwe's misfit dislocation model.[5] Our results show that coherent films are stable for x up to 50%, which is in a range of technological interest. The observed h_c is at least an order of magnitude thicker than the theory and, moreover, the dependence on lattice misfit is significantly more abrupt. We show that a relevant critical quantity in the relaxation of the misfit strain is the interfacial shear strength. These results are based on measurements of coherency strains in films of Ge_xSi_{1-x} alloy obtained by the independent techniques of ion channeling and x-ray diffraction.

The alloys were grown in the UHV silicon MBE apparatus described previously.[6] Ge and Si from two e-beam sources were deposited in 3-in. (100) Si wafers at rates totaling 5 Å s^{-1}, immediately following a 0.1-μm growth of pure epitaxial Si. Substrates were held at temperatures ranging from 400 to 750 °C and a series of film thicknesses were grown on a given wafer. Thicknesses ranged from 50 Å to 1 μm. The samples were exposed to air and cleaved into 1-cm^2 pieces for subsequent analysis.

Rutherford backscattering spectrometry (RBS) with 1.8-MeV $^4He^+$ ions was used for quantitative analysis. Stoichiometry of films thicker than 100 Å was determined from the relative backscattering yield from Ge and Si in the film. Channeling spectra were used to ascertain crystalline quality. Minimum channeling yields for backscattering from near the film surface, χ_{min}, was 0.02 at $x = 5\%$ increasing to 0.04 at $x = 20\%$, for normal incidence [100] channeling. The variation of dechanneling with depth[7] is similar to that of bulk Si crystals. Conversely, for $x \gtrsim 50\%$, χ_{min} depends on film thickness (0.1 for 500 Å, and 0.046 for 2500 Å) and the dechanneling yield from the film near the interface increases to about 25%. A conclusion we may draw from these observations on films thickner than 100 Å is that for $x \lesssim 20\%$ the interfacial region shows coherent epitaxy, whereas for $x \gtrsim 50\%$ there is a type of interfacial disorder detected by ion channeling. The 100-Å films were too thin to quantify accurately, owing to finite depth resolution.

Commensurability requires equality of the in-plane lattice spacings at the interface and a planar compressive strain in the film equal to the misfit, defined as $f = (b - a)/a$, where b and a are bulk lattice parameters of the alloy and Si, respectively. Ideally, such a film is free of dislocations and the misfit is taken up by a homogeneous tetragonal distortion. Ion channeling spectra along off-normal channeling directions showed evidence for the distortion as poor alignment in the substrate with the beam well aligned in the film. In the distorted crystal the off-normal ⟨110⟩ and ⟨111⟩ axes make angles θ with respect to the [100] normal which are smaller than in the case for cubic symmetry (no strain). The difference, denoted by $\Delta\theta$, is related to the tetragonal distortion ϵ_T by

$$\epsilon_T = \frac{b_\perp - b_\parallel}{b} = \frac{-\Delta\theta}{\sin\theta\cos\theta}, \tag{1}$$

where $b_\perp$ and $b_\parallel$ are the respective normal and in-plane lattice constants. A typical result is shown in Fig. 1 for $x = 50\%$ for a scan in a (110) plane passing through a ⟨111⟩ direction. Plotted are the backscattering yields from Ge in the film and from Si in the substrate. The displacement between the curves gives $\Delta\theta = -0.91°$ and $\epsilon_T = 3.3\%$. We have not applied this method to films thicker than 100 Å because, as we have found, channeling in the epilayer steers the beam before it enters the substrate. Instead we used the difference in the orientations of pairs of crystallographically equivalent axes, separated by 2θ.

We show our results in Fig. 2, where ϵ_T is plotted against Ge concentration for various film thicknesses. No systematic difference in ϵ_T was found for measurements of the ⟨110⟩ and ⟨111⟩ channels, so the results were averaged. Except for an anomalous point (100 Å, 20%), there is no dependence of ϵ_T on growth temperature or on film thickness up to 2500 Å in the region $x \lesssim 20\%$. The solid curve is a theoretical dependence based on the assumption of exact in-plane registry in the epilayer, i.e., $b_\parallel = a$, which from elasticity theory gives

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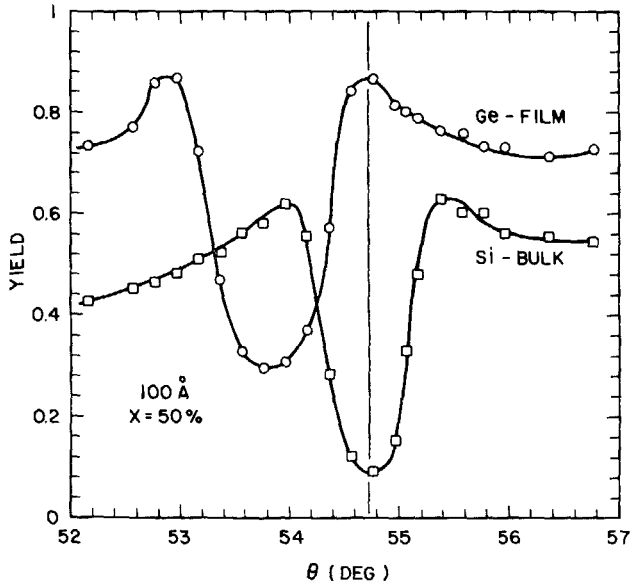

FIG. 1. Ion backscattering yields as function of tilt angle θ, measured with respect to [100] surface normal, scanned in a (110) plane across a ⟨111⟩ Si-substrate axis, for which $\theta = 54.74°$. Ge: yield from film; Si: yield from substrate.

$$\epsilon_T = \left(\frac{1+\nu}{1-\nu}\right) f, \tag{2}$$

where ν is Poisson's ratio and $f = (b - a)/a$. We computed f by taking for b the measured values of the lattice parameters of bulk Ge_xSi_{1-x}[8,9] and for ν an interpolation between Ge ($\nu = 0.273$) and Si ($\nu = 0.280$). The points in Fig. 2 break away from the dependence given in Eq. (2) at increasing Ge concentrations. Thickness h_c obtained from the break points separate regions of commensurate (C) from incommensurate (I) behavior and are shown in Fig. 2, inset. The h_c vs x plot includes additional data for $x = 11\%$ and 70%, not shown in the main part of the figure.

X-ray measurements were made with a rotating anode source (Cu K_α) and conventional two-circle diffractometer.[9] We determined $b_\perp$ directly by means of the normal (400) reflection; the same radial scan shows distinct peaks for the alloy film and silicon substrate superimposed. We measured $b_\parallel$ similarly by using the in-plane (022) reflection in the glancing-incidence geometry. In the composition region $x \lesssim 20\%$, only a single peak in the in-plane scan of 2500-Å-thick films was seen, confirming that $b_\parallel = a$ and thus that the epitaxy is commensurate. The strain ϵ_T derived from $b_\perp$ was in complete agreement with the ion channeling results in Fig. 2.

The $x = 50\%$ series of samples exemplifies the trend in thickness from commensurate to incommensurate films. These results are presented in Table I and some typical scans are shown in Fig. 3. A large tetragonal strain of 3% is seen in the commensurate 100-Å film, but thicker films (above h_c) are less strained. The loss of strain above h_c is presumably due to the introduction of misfit dislocations. The values of $b_\perp$ and $b_\parallel$ approach the ideal unstrained value b from above and below, respectively; the tetragonal strains [Eq. (1)] are plotted along with the ion channeling values in Fig. 2. The incommensurate films also show a progressive loss of orientational order, as seen in the mosaic spread ($\Delta\omega$) of the alloy layer, Fig. 3, inset.

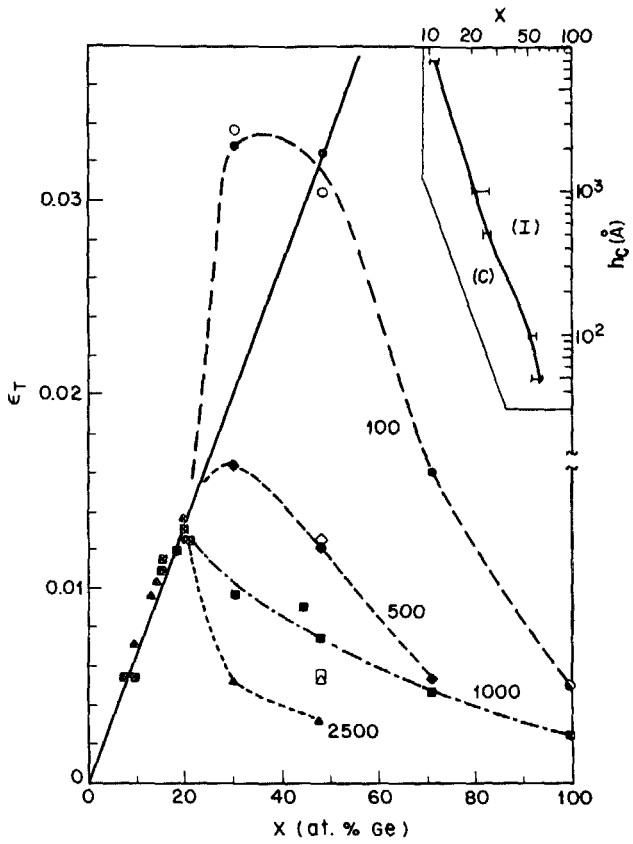

FIG. 2. Ge concentration dependence of the tetragonal distortion obtained by ion channeling (closed symbols) and x-ray differaction (open symbols) for film thicknesses indicated in Å units. Solid curve: theoretical strain for commensurate growth, Eq. (1). Inset: Concentration dependence of critical thickness in the form of a phase diagram separating commensurate (C) from incommensurate (I) regions.

Our observations do not support the Frank and van der Merwe misfit dislocation model,[5] since the critical thicknesses are too large and do not obey the theoretical dependence on misfit. Considering various specific assumptions about the misfit dislocation energy,[3,10,11] the model predicts mechanical stability for $h < h_c = \alpha a/f$, where α depends logarithmically on f and is typically between 0.1 and 0.2. The prediction for $x = 10\%$ is $h_c = 250$ Å, which is in disagreement with our observation of commensurate 2500-Å films for x up to 20%. Moreover, the approximate f^{-1} dependence of the model disagrees with the more rapid dependence shown in Fig. 2, inset.

We therefore suppose that there are barriers to the nucleation and transport of enough misft dislocations to significantly relax the tetragonal strain in the commensurate het-

TABLE I. X-ray structural parameters for $x = 50\%$ films.

h (Å)	$\frac{b_\parallel - a}{a}$ (%)	$\frac{b_\perp - a}{a}$ (%)	ϵ_T (%)	$\Delta\omega_{FWHM}$ (deg)
100	0	3.05	3.05	<0.1
500	1.25	2.50	1.25	0.28
1000	1.95	2.50	0.55	0.42
2500	2.00	2.50	0.50	0.40

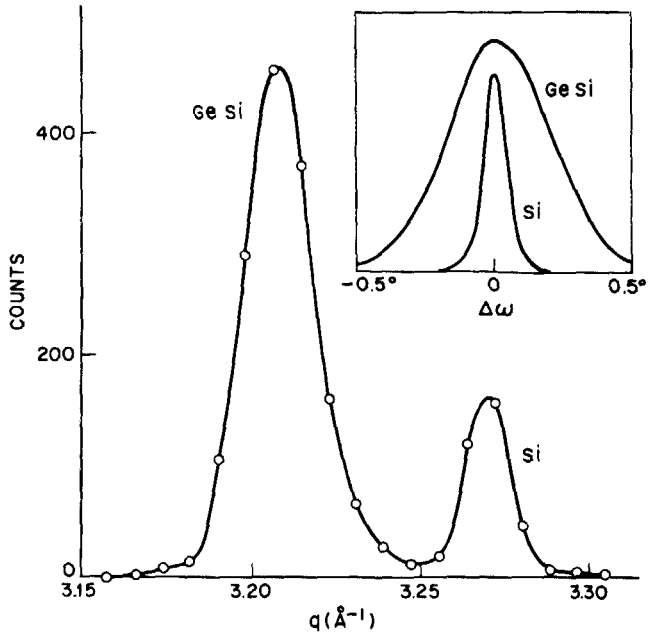

FIG. 3. X-ray diffractometer scan for (022) reflection; $\hbar q$ is the momentum transfer. Peaks are identified wth GeSi film ($x = 50\%$, 1000 Å) and Si substrate. Inset: Rocking curves (superimposed) at the two maxima in the q scan. GeSi curve shows mosaic spread; Si curve is resolution limited.

erostructures. The nucleation barriers may be calculated by considering shear displacements of the epilayer with respect to the substrate. The average interfacial energy areal density at the critical thickness may be estimated as $\mu c^2/4\pi^2 d$, where μ is the shear modulus, c the slip distance, and d the interplanar spacing.[5] Equating this to the coherency-strain energy areal density, $2(1+\nu)\mu f^2 h_c/(1-\nu)$, and taking $d = a/4$ and $c = a/\sqrt{2}$, gives the result $h_c \approx 0.014 a f^{-2}$. This estimate accounts for our observation in the vicinity of $x = 25\%$, where it predicts $h_c = 900$ Å, although the dependence in Fig. 2 is somewhat faster than f^{-2}. We find that ϵ_T relaxes with a $h^{-1/2}$ dependence on thickness for $h > h_c$, which also indicates relevance of the metastability energy. We conclude that the strong interfacial bond is important for a large h_c, but is not the entire explanation. The difference between the explanation given here and the Frank and van der Merwe treatment arises from the fact that these strained films are not in mechanical equilibrium.[4]

Our experimental resolution in measuring changes in ϵ_T and $\chi_{\min}$ corresponds to misfit dislocation spacings $\gtrsim 1$ μm at $h = h_c$, which gives a slight overestimate of h_c. We may explain the larger h_c compared to earlier work[3,4] by recent improvements in Si MBE, particularly substrate preparation techniques. This has allowed a 200 °C reduction in the epitaxy temperature, permitting the growth of smooth alloy films over the entire Ge concentration range. In summary, then, we have found that MBE Ge_xSi_{1-x} epitaxy on Si(100) is characterized by a strong heteroepitaxial bond, where the introduction of interfacial misfit dislocations is inhibited. Relatively thick, strained, commensurate epitaxial layers have been obtained at Ge concentrations at least up to 50%, with tetragonal distortions as large as 3%.

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