TETRAGONAL STRAIN IN MBE Ge_xSi_{1-x} FILMS GROWN ON (100) Si OBSERVED BY ION CHANNELING AND X-RAY DIFFRACTION

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ABSTRACT

Structure of Ge_xSi_{1-x} alloy films grown on (100) Si by molecular beam epitaxy is analyzed by MeV He^+ ion channeling and X-ray diffraction as functions of Ge concentration, film thickness and growth temperature. Critical thicknesses for pseudomorphic growth are determined for $x \leqslant 0.5$, where coherent tetragonally-strained layers are observed. The average strain decreases approximately as the square-root of thickness when the critical thickness is exceeded. At temperatures near the threshold for islanding growth, surface roughness appears as a precursor to degradation of strained-layer epitaxy. No effect on the amount of the tetragonal strain was found in a study of ion-beam damage.

INTRODUCTION

Preparation of nearly defect-free layers of Ge-Si alloys on Si is of interest for modified semiconductor materials in Si technology. We have recently reported that smooth Ge_xSi_{1-x} alloy layers can be grown on (100) Si in a state-of-the-art MBE apparatus over the entire $0 \leqslant x \leqslant 1$ Ge concentration range [1-3]. Using properly prepared substrates in a UHV environment, smooth epitaxial films will grow at a substrate temperature of 550 °C for all x. Three-dimensional islanding occurs at higher temperatures, e.g., 750 °C at $x=0.1$ decreasing to 600 °C for $x \geqslant 0.4$. The 4.2% lattice-parameter mismatch between Ge and Si gives rise to a misfit energy between Ge_xSi_{1-x} and the Si substrate which favors the islanding growth mode.

The smooth films grown at lower temperatures also grow pseudomorphically up to a critical thickness h_c which decreases as x increases. Predictions of h_c and strain by mechanical equilibrium theory [4] are much smaller than observed. Films thinner than h_c have coherent interfaces, i.e., they are strained in planar compression so that the parallel lattice spacing matches the Si substrate. Thus metastable dislocation-free alloy films can be grown at the relatively low temperature of 550 °C. Crystalline quality and the tetragonal strain associated with lattice mismatch are determined by ion channeling and X-ray diffraction measurements. For the analysis we use the following definitions: a is the lattice parameter of Si, $b_\parallel$ and $b_\perp$ the parallel and perpendicular lattice spacings in the alloy film, b the bulk alloy lattice parameter, and $f = (b-a)/a$ the misfit parameter, which is approximately given as $f = 0.042x$.

ION SCATTERING

Rutherford backscattering of 1.8 MeV He^+ ions was used to ascertain the stoichiometries and thicknesses of the Ge_xSi_{1-x} films. For these purposes "random" backscattering spectra were taken with the ion beam oriented away from a channeling axis while the crystal was rotated about the azimuth. Channeling spectra were obtained with the beam aligned with the [100] normal and low-index off-normal directions, [110] and [111]. The ratio of the channeling yield, taken at an energy just below the surface peak in the channeled spectra, to the random yield is denoted χ_{min}. For pure bulk Si and Ge (100) $\chi_{min} \approx 3.5\%$. A low value of χ_{min} is generally taken as an indication of a low density of structural defects; a perfect crystal has a non-zero χ_{min} because of thermal motion of the atoms.

For alloys grown at $x \leqslant 0.2$ we find the χ_{min} obtained from the Ge backscattering is somewhat less than the pure Si or Ge values. For example, $\chi_{min} = 2.9\%$ at $x \approx 0.15$ in a 1000-Å thick film. Smaller apparent χ_{min} may arise from the statistics of channeling in a thin layer. But there could also be a reduction in the atomic vibration amplitudes in the transverse (100) plane of the strained layer.

Larger χ_{min} values are found for 1000-Å films at $x > 0.2$ because the interface coherency breaks down for this thickness. The χ_{min} increases to a maximum of about 13% near $x=0.5$ and then decreases to about 7% for pure Ge films. Channeling spectra taken with a 100° scattering angle (a technique to increase depth resolution) show larger dechanneling rates and interfacial peaks for the $x > 0.2$ films. Analysis by electron microscopy (TEM) shows that dislocations are the source of the disorder in the 1000-Å films for $x > 0.2$ and that an array of misfit dislocations is formed at the film/substrate interface [3].

Ion channeling along off-normal <111> and <110> directions was used to measure the tetragonal distortion in the films. We express the tetragonal distortion ϵ_T in terms of the angular difference $\Delta\theta$ between the symmetry direction in the distorted lattice and that of corresponding cubic symmetry: $\epsilon_T = -\Delta\theta/\sin\theta\cos\theta$. In this expression θ is the angle between the measured symmetry direction and the surface normal. The channeling axis is determined from the minimum in the backscattering yield from the Ge in the film as a function of θ and the azimuthal angle. The quantity $\Delta\theta$ was obtained from measurements of the angles between the [110] and [1$\bar{1}$0] axes and between [111] and [1$\bar{1}\bar{1}$]. These two separate measurements yielded the same value for ϵ_T.

An example of a channeling angular scan in a (110) plane is shown in Fig. 1 for a 100-Å thick 30% Ge film. The curves are normalized to the yield for a random (non-channeling) direction in the crystal. The lower set of curves is the yield from Si in the substrate and the upper set is the yield from Ge in the film. The minimum in each curve occurs where the beam is aligned with respect to the <111> directions, which are displaced in orientation by the amount $\Delta\theta$. A larger normalized yield is expected for Ge because of scattering from the surface of the film. Analogous angular scans were also made for 1000-Å thick films, which do not show the displaced effect. In this case dechanneling and guiding of the beam is sufficient to mask the separate channeling dip for scattering by the substrate with the result that the minima occur at the channeling axis of the film.

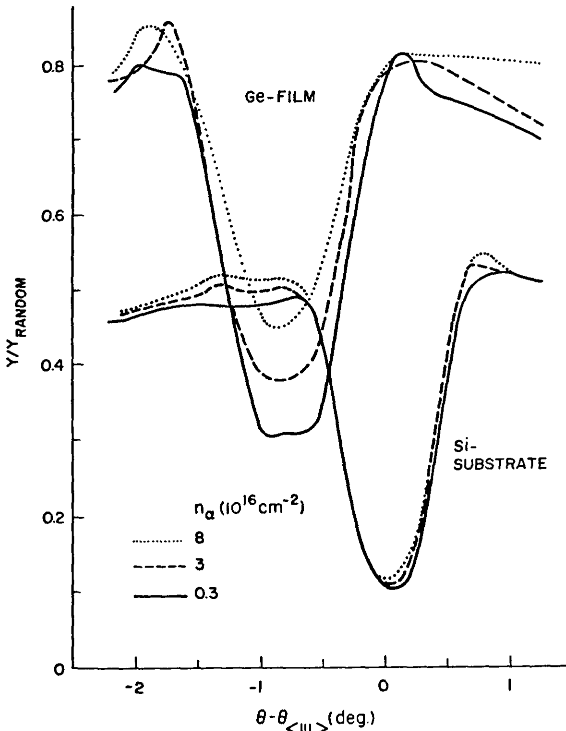

Fig. 1. Three 1.8 MeV He^+ channeling angular scans in a (110) plane through a <111> off-normal axis of the backscattering yield from a 100-Å $Ge_{0.3}Si_{0.7}$ film on Si. Y_{random} is the non-channeling yield, $\theta_{<111>} = 54.74°$, and n_α the irradiation exposure prior to each scan.

The effect of ion-beam damage is also shown in Fig. 1. The three sets of curves were taken after irradiating the sample in a planar channeling direction to the 1.8 MeV He^+ doses shown. The fraction of displaced atoms is proportional to the increase in the minimum yield at the channeling dip. A greater effect is observed in the Ge film, mainly because the collision cross-section is proportional to the backscattering yield, which varies as the square of the atomic number of the scattering nucleus. Taking this into account, the resulting damage cross-section for the film is about twice that of the Si substrate. It is noteworthy that a substantial increase in disorder produces no shift in the position of the channeling dip in the film, hence, no relaxation of the strain. Evidently, the type of point defect damage produced by irradiation does not upset the metastability of the film.

X-RAY DIFFRACTION

X-ray diffraction affords a direct measurement of the separate lattice spacings in the films. We used Cu K_α radiation from a high-power rotating anode and a conventional 4-circle diffractometer to measure the parallel and perpendicular lattice spacings of films with composition $x = 0.5$. Figure 2 shows the grazing incidence geometry used to measure the (022) Bragg reflection, giving $b_{\parallel}$. Although strictly lying in the surface plane, at least one of the symmetry equivalents of this reflection was found to lie above the plane because of crystal miscut. The normalization of all scans shown is therefore somewhat arbitrary because of the unknown penetration depth, which is a very sensitive function of incidence angle. The 100-Å thick film seen in Fig. 2 is commensurate with the Si substrate, as only one diffraction peak located at the (022) position of bulk Si is observed. For the 500- and 1000-Å thick films, two peaks are observed. Here, the larger peak on the left is attributed to diffraction from the film, while the smaller peak comes from the underlying Si substrate. The larger in-plane $b_{\parallel}$ parameter of the thicker films demonstrates the relief of the strain and the breakdown of pseudomorphic growth.

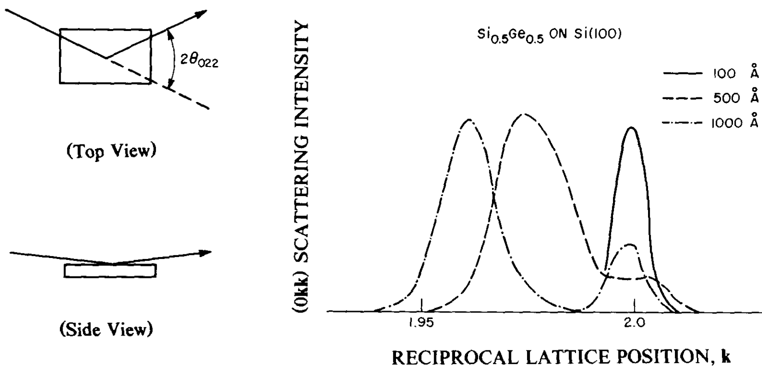

Fig. 2. Grazing incidence X-ray scans along the in-plane (0kk) direction as a function of the reciprocal lattice coordinate, **k**. Data are shown for three Ge-Si alloy film thicknesses. The (022) position for bulk Si is at **k** = 2.0 and for bulk Ge-Si it is near the peak for 1000-Å film. Two views of the scattering geometry are shown.

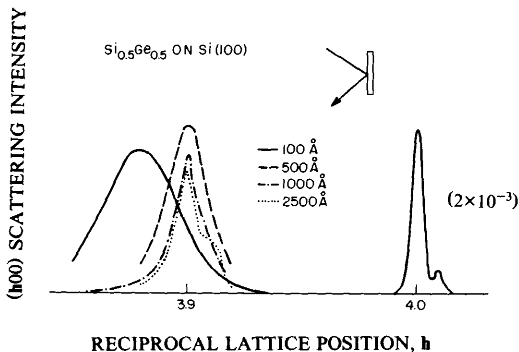

Fig. 3. X-ray diffraction intensities along (h00) scanned as a function of reciprocal lattice coordinate, **h**. The bulk Si (400) peak position is at **h** = 4.0. Vertical scale is arbitrary; bulk Si signal is 10^3 larger than that of thickest film.

Figure 3 shows X-ray scans of the perpendicular (400) Bragg reflection for films of several thicknesses and $x = 0.5$. The inset shows the corresponding scattering geometry. The peak on the right at $\mathbf{h} = 4.0$, shown for the 100-Å thick film, corresponds to the Si substrate. Similar peaks were also observed for the other films, but were omitted for clarity. The doublet structure is due to the two K_α lines of the characteristic X-ray source. The peaks on the left arise from the alloy films, which have a larger lattice spacing $b_\perp$. The peak associated with the 100-Å film is shifted with respect to the others because of the dilation of the perpedicular lattice spacing in the case of pseudomorphic growth. The penetration depth is large compare with the film thickness in this geometry. Spatial correlation lengths, $\Gamma_\perp$, were derived from the linewidths by deconvolution of the instrumental width. These are measures of the coherently scattering domain sizes in the films. Dependence of $\Gamma_\perp$ on film thickness h is shown in Fig. 4. The line $\Gamma_\perp = h$ denotes the width intrinsic to finite film thickness. The strain-relaxed films fall below the line, implicating the disruptive role of defects in the thicker films.

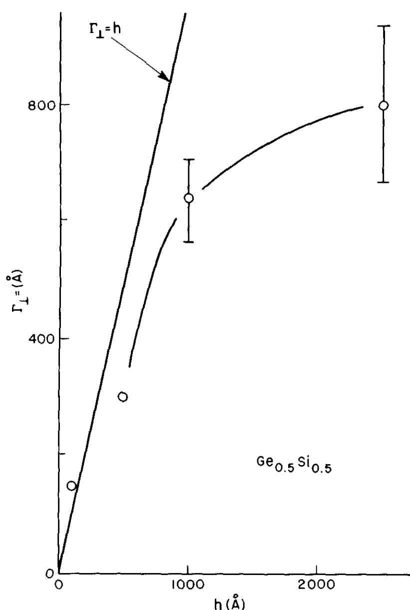

Fig. 4. Perpendicular correlation lengths $\Gamma_\perp$ obtained from data of Fig. 3 and plotted as a function of film thickness h.

These results at $x = 0.5$ show that the initial 100 Å growth of film is strained coherently. Then at a thickness between 100 and 500 Å the built-in strain is relieved and, as TEM and ion scattering show, dislocations enter the film. Dislocations may glide into the film along the inclined {111} slip planes. Edge dislocations which glide along {111} have an edge component of their Burgers vector in the (100) plane and can accommodate the misfit. The screw component gives rise to a crystal rotation about the [100] direction by an angle approximately equal to f. We have observed this effect as a mosaic spread in X-ray rocking curves of the film (022) diffraction peaks [2]. Full-width half-maximum mosaic spreads of 0.4° were found for the 1000- and 2500-Å films, in good agreement with the expectation for this mechanism.

STRAIN RESULTS

A summary of the strain measurements is plotted as a function of Ge fraction for various film thicknesses in Fig. 5. The X-ray results were used to compute strain according to the relation $\epsilon_T = (b_\perp - b_\parallel)/b$ and are shown as open symbols in the figure. Several samples for $x \leqslant 0.2$ were examined by both ion channeling and X-rays and produced identical results. The solid curve is the theoretical

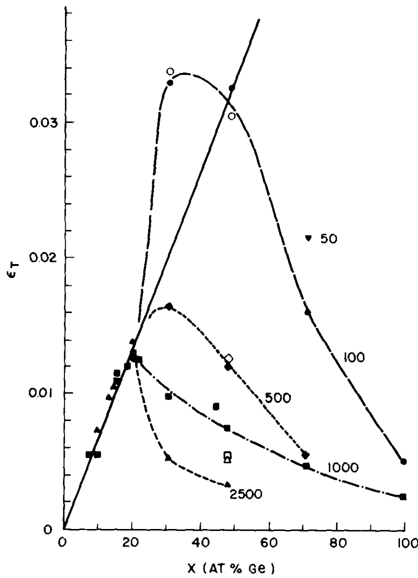

Fig. 5. Tetragonal strain measurements plotted as a function of Ge concentration. Filled symbols: ion channeling, open symbols: X-ray diffraction. Broken curves connect points of constant film thickness, given in Å units. Solid curve is the theoretical maximum coherency strain.

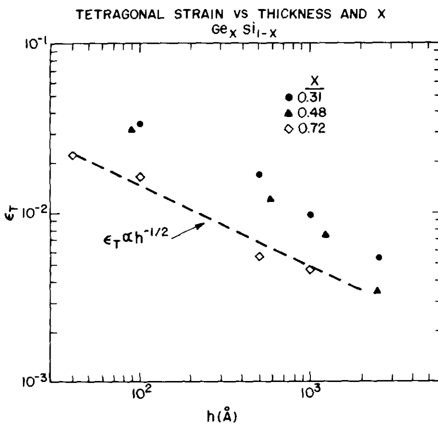

Fig. 6. Tetragonal strain as a function of film thickness h for three values of Ge concentration x. Dashed curve assumes constant strain energy areal density.

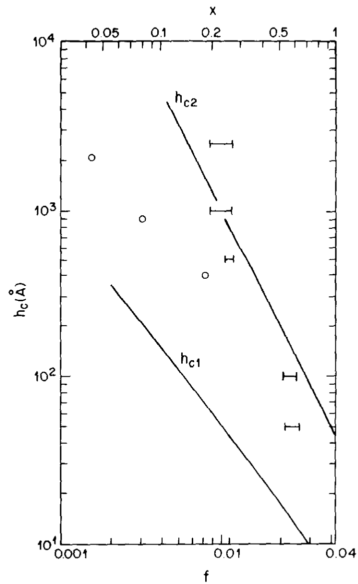

Fig. 7. Critical thickness for coherent epitaxy in $Ge_x Si_{1-x}$ films on (100) Si as a function of x (upper scale) or misfit parameter f (lower scale). Bars denote present work, open symbols from Ref. 5. Lower h_{c1} curve is derived from Frank and van der Merwe theory, Refs. 4 and 5. Upper h_{c2} curve is derived from a constant critical strain energy areal density.

expression for the coherency tetragonal strain, $\epsilon_T = (1+\nu)f/(1-\nu)$, where ν is Poisson's ratio. We observe that the strainsfor all samples fall on the theoretical curve for $x \leqslant 0.2$. For larger x, the broken curves for the various film thicknesses break away from the theoretical curve at points which define the critical thicknesses for coherent epitaxy.

The variation of the strain with film thickness is replotted in Fig. 6. In the case of the highest concentration shown, all thicknesses are above the critical value. We observe a comparatively gradual $h^{-1/2}$ thickness dependence, illustrated by the dashed curve, in contrast to the h^{-1} dependence predicted for an array of misfit dislocations in equilibrium at the interface [4]. Moreover, since the magnitudes of the strain exceed theoretical equilibrium values, we conclude that there is an energy barrier for dislocation formation (or transport) which scales with the stored elastic energy areal density, $E = 2(1+\nu)\mu f^2 h/(1-\nu)$, where μ is the shear modulus.

Critical thicknesses were estimated directly from Fig. 5 and are plotted against misfit f and Ge concentration x in Fig. 7. This graph is effectively a phase diagram, where the stability boundary of the region of coherency (lower left) is mapped out by the critical thickness data points. The error bars reflect the uncertainty associated with our finite number of measurements. For comparison we show as open symbols the critical thicknesses deduced by Kasper and Herzog [5] in earlier work on the region $x < 0.2$. Also shown as the curve labeled h_{c1} is the prediction of the equilibrium theory of Frank and van der Merwe [4], which significantly underestimates h_c and which does not predict the more rapid observed dependence of h_c on f. The curve labeled h_{c2} in Fig. 7 illustrates the behavior possible if the critical thickness is associated with an energy barrier proportional to the coherency-strain energy. Without specifying an explicit model of dislocation nucleation, this curve was produced by equating the coherency strain energy at $h = h_c$ to the maximum misfit interfacial energy, estimated as $\mu c^2/4\pi^2 d$, with $c = 2^{-1/2}a$ as the slip distance and $d = a/4$ as the interplanar spacing. The result is $h_c = 0.014af^{-2}$, which is plotted as h_{c2} and is in fair agreement with the data. A proper model would of course include the geometry of the nucleation process and an activation factor. Guided by the results of Fig. 7, we have recently reported [3] growth of a dislocation-free strained-layer superlattice of alternating $Ge_{0.3}Si_{0.7}$ and Si layers.

That all films up to 0.25μm thick are coherent for $f < 0.01$ can also point to the existence of a critical strain for thick films. Models of dislocation nucleation predict a critical strain in terms of thermally-activated processes. All of the films examined at $x \leqslant 0.2$ showed the theoretical coherency strain, however, even though the growth temperatures varied between 400 °C and 750 °C. The region of higher growth temperature near the threshold for islanding growth was also examined. At 650 °C and $x \approx 0.2$ Nomarsky microscopy showed that the precursor to islanding is a surface texture of <110>-oriented steps. Ion channeling showed no increase in χ_{min} and the theoretical ϵ_T in this case. In contrast films for larger x or high temperature, irregularly textured islands were observed and χ_{min} increased to about 10% while ϵ_T decreased. Hence, we conclude that islanding is the equilibrium state and the increase in surface energy is less than the decrease in misfit strain energy.

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