Ge_xSi_{1-x}/Si strained-layer superlattice grown by molecular beam epitaxy

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Ge_xSi_{1-x} films are grown on Si by molecular beam epitaxy and analyzed by Nomarski optical interference microscopy, Rutherford ion backscattering and channeling, x-ray diffraction, and transmission electron microscopy. The full range of alloy compositions will grow smoothly on silicon. Ge_xSi_{1-x} films with $x \leqslant 0.5$ can be grown free of dislocations by means of strained-layer epitaxy where lattice mismatch is accommodated by tetragonal strain. Critical thickness and composition values are tabulated for strained-layer growth. Multiple strained layers are combined to form a Ge_xSi_{1-x}/Si strained-layer superlattice.

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I. INTRODUCTION

It is known that lattice mismatched layers may be grown without misfit dislocations if sufficiently small layer thicknesses are employed.[1] The layers are strained such that the lattice parameters parallel to the interface are equal and the growth is described as pseudomorphic or commensurate. Pseudomorphic growth has been demonstrated for dilute Ge_xSi_{1-x} alloys on Si,[2,3] for $GaAs_xP_{1-x}$ on GaAs,[4–7] and for $In_xGa_{1-x}As$ on GaAs.[7] Multilayer pseudomorphic structures are referred to as "strained-layer superlattices."[6–9] In this paper we describe the pseudomorphic growth of Ge_xSi_{1-x} on Si. Our results differ from earlier Ge_xSi_{1-x} work in that: (1) A wider range of growth parameters is employed. (2) Pseudomorphic growth is demonstrated on Si for alloys with up to 50% Ge. (3) Pseudomorphic growth is maintained for alloy layer thicknesses up to 1/4 μm. (Such thicknesses are approximately an order of magnitude larger than those observed in earlier experimental work[2–7] or predicted by equilibrium theory.[1]) (4) The growth of a Ge_xSi_{1-x}/Si strained-layer superlattice is reported. Preliminary results of this work were given elsewhere.[10,11]

Ge_xSi_{1-x} alloys were grown on (100)Si substrates by codeposition from two electron beam evaporation sources. The fluxes from the Si and Ge e-beam sources were separately sensed and controlled to yield a total deposition rate of 5 Å/s. Prior to deposition substrates were chemically cleaned, then argon sputtered *in situ* and annealed at 800–850 °C. Above 500 °C, annealing and growth temperatures were measured directly by infrared pyrometry; below 500 °C temperatures were estimated by extrapolation of heater power versus temperature curves. The apparatus, cleaning, and deposition procedures are detailed in earlier publications.[10–13]

II. MORPHOLOGY

In lattice mismatched systems where high interfacial energies are expected there is a general tendency for deposited material to grow in three-dimensional islands rather than in smooth layers. Islanding may persist for thicknesses of hundreds or even thousands of angstroms. This behavior has been observed for molecular beam epitaxial (MBE) growth of Ge,[14] Ge_xSi_{1-x},[2,3] $NiSi_2$,[15] and $CoSi_2$[15] on Si, and for growth of Si on Al_2O_3,[16] $MgAl_2O_4$,[16] and ZrO_2.[17] To establish whether thin Ge_xSi_{1-x} films could be grown on Si without islanding, we grew films at temperatures of 400–750 °C, with alloy compositions of $x = 0$–1, to thicknesses of 100, 500, 1000, and 2500 Å. Film morphology was then evaluated by Nomarski optical interference contrast microscopy.

We found that at a given epitaxial growth temperature films grew on Si with optically flat surfaces up to a critical alloy composition x_c. Above x_c islands formed and often persisted in films of 1000 or 2500 Å thickness. In certain alloy films, sources and substrates were positioned deliberately to yield a steady variation in alloy composition across a wafer. In these samples the transition from two-dimensional to three-dimensional growth could be seen as an abrupt change from a specularly reflecting to a hazy surface as one scanned towards the Ge rich side of the wafer.

Rutherford backscattering was used to measure the alloy composition at specific points and to determine the value of

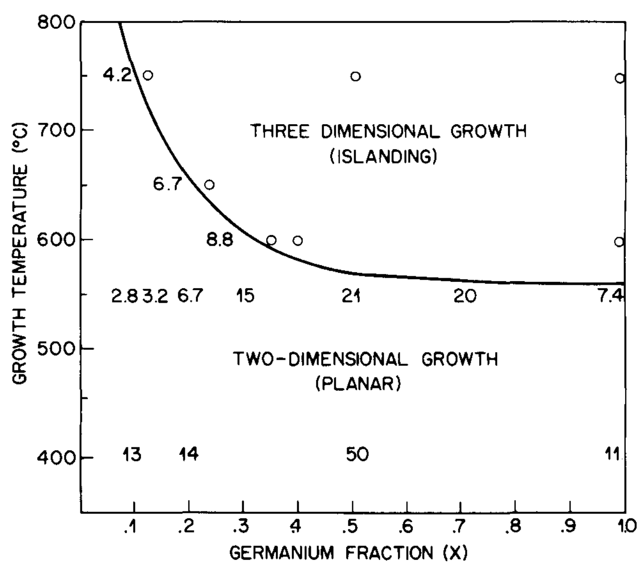

FIG. 1. Plot of Ge_xSi_{1-x} film morphology and y_{chan}/y_{rand} vs growth temperature and film composition for 1000 Å thick alloy films. Rough film produced at points indicated by (O). Smooth films produced at points indicated by numerical values. Numerical values are the ratio of ⟨100⟩ channeling to random backscattering yields (y_{chan}/y_{rand}) in percent. Values of 2%–3% and 100% would indicate perfectly crystalline and completely disordered films, respectively.

x_c in films grown at different temperatures. These data are shown in Fig. 1 where layer growth temperature and composition are plotted as circles for conditions yielding rough growth or y_{chan}/y_{rand} values (discussed below) for conditions yielding smooth growth. It is seen that the value of x_c is highest at lower growth temperatures and that below ~575 °C alloys of all compositions up to pure Ge will grow smoothly on Si.

III. CRYSTALLINITY

The observation that lower temperatures are required to avoid island growth in concentrated alloys suggests that islanding is being "frozen out." That is, as alloy composition increases, the Ge_xSi_{1-x}/Si interfacial energy increases relative to the free-surface energy to the point where islanding becomes energetically favorable. Then islanding can be avoided only by so limiting the surface atomic migration lengths that macroscopic islands cannot form (i.e., there is a kinetic barrier). If this were the case one could also reasonably ask whether surface migration lengths are so limited as to achieve smooth growth only at the cost of reduced crystallinity.

To test this possibility, Ge_xSi_{1-x} films were examined by Rutherford backscattering and channeling measurements. A typical set of backscattering spectra are shown in Fig. 2 for a smooth 1000 Å thick $Ge_{0.2}Si_{0.8}$ film on Si. The spectrum taken with an unaligned helium beam gives an indication of composition versus depth in that particles scattered at greater depths reach the detector at lower energies. The yield below 1.2 MeV corresponds to Si in the substrate, the yield from 1.2–1.3 MeV to Si in the alloy, the yield from 1.4–1.6 MeV to the Ge in the alloy (the Ge peak is displaced to the right because the higher mass Ge scatters He back at a higher energy). Crystallinity is measured by comparing channeled to random yields. As crystallinity improves the channeling yield decreases. For conditions yielding smooth growth, we have tabulated the ratio of channeled to random yield (y_{chan}/y_{rand}), integrated over the entire Ge peak, and values are inset in Fig. 1. An ideal crystal should give a y_{chan}/y_{rand} value of ~3.0%.

Values of y_{chan}/y_{rand} are indeed low for a range of growth conditions indicating good epitaxial layer quality. In particular, at 550 °C it is evident that smooth growth is achieved for all alloy compositions without a major sacrifice of crystalline quality. It is significant, however, that at both 400 and 550 °C growth temperatures y_{chan}/y_{rand} values increase sharply between $x = 0.2$ and $x = 0.5$ alloy compositions. This indicates a change in growth mode which can be observed directly in either high depth resolution (⟨100⟩ incidence/grazing exit) or by cross-sectional transmission electron microscopy (TEM) as shown in Figs. 3 and 4, respectively. Figure 3 shows an enlargement of the channeled backscattering spectra from the Ge in 1000 Å thick Ge_xSi_{1-x} films of composition $x = 0.1, 0.2, 0.5$, and 1.0. In the dilute alloys the backscattering yield is low behind the 1.58 MeV surface peak. In the concentrated alloys the yield increases steadily with depth and an additional disorder peak is evident at the Ge_xSi_{1-x}/Si interface (1.43 MeV). Figure 4 shows bright field TEM cross sections for the same four films. In each cross section the alloy is the top band, the

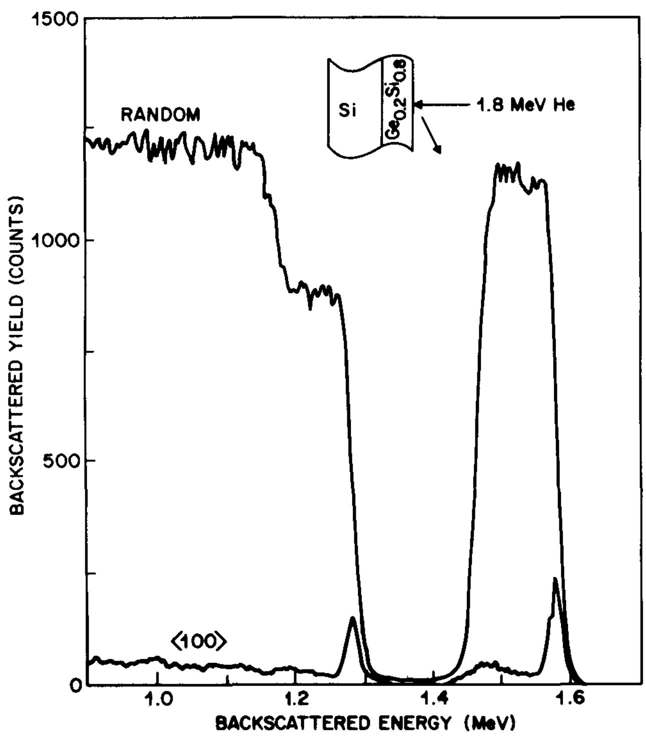

FIG. 2. Rutherford backscattering spectra of 1000 Å $Ge_{0.2}Si_{0.8}$ epitaxial layer on Si. Spectra taken with near normal incidence and grazing exit angle to enhance depth resolution. Upper spectrum taken with random incidence, lower spectrum with ⟨100⟩ channeled incidence.

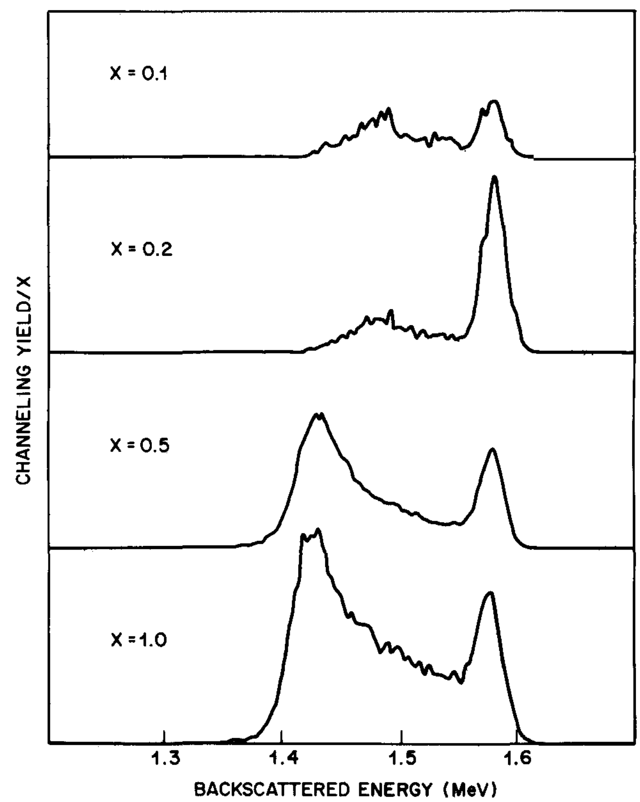

FIG. 3. Enlargement of channeled Ge spectra from 1000 Å Ge_xSi_{1-x} films with $x = 0.1, 0.2, 0.5$, and 1.0. Spectra show transition from commensurate (dislocation free) growth in dilute alloys to incommensurate growth in concentrated alloys. Films grown at 550°C y_{chan}/y_{rand} values given in Fig. 1.

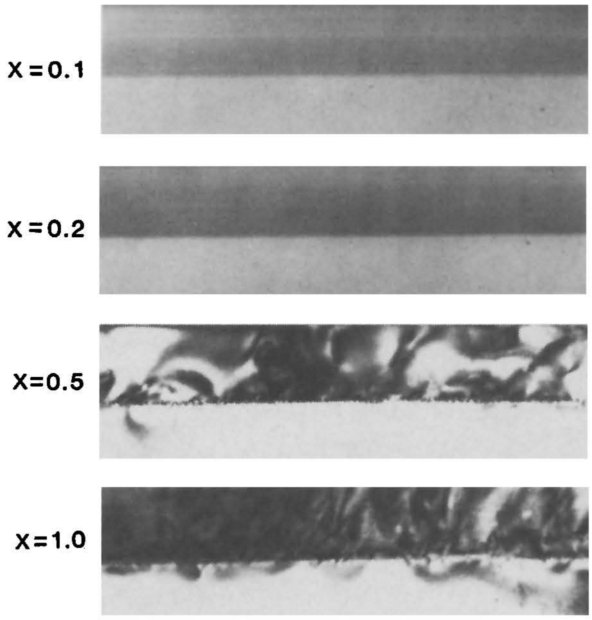

FIG. 4. TEM cross section of 1000 Å thick Ge_xSi_{1-x} films shown in backscattering spectra of Fig. 3. Again, transition from commensurate to incommensurate growth is clearly evident.

substrate the bottom band. It is clear that the $x = 0.1$ and 0.2 alloys contain no observable dislocations even though bulk alloys of these compositions would be mismatched from the Si substrate by ~0.4% and 0.8% respectively.[18] In the $x = 0.5$ and 1.0 films, dislocations and inhomogeneous strain are evident correlating well with the backscattering spectra of Fig. 3.

IV. STRAIN

The data above strongly suggest a transition from commensurate (or pseudomorphic) growth to incommensurate growth as defined by the schematics of Fig. 5. To confirm the transition directly we have used x-ray and backscattering analyses to measure alloy and substrate lattice parameters. X-ray measurements were made with a rotating anode source (Cu K_α) and a conventional two-circle diffractometer. Two geometries were employed: (1) incident and diffracted beams symmetric about the surface normal to reach the (400) reflection, and (2) incident and diffracted beams grazing the crystal surface to reach the in-plane (022) reflection. The alloy lattice constants were thus measured parallel and perpendicular to the interface and are plotted in Fig. 6 for $Ge_{0.5}Si_{0.5}$ as a function of film thickness. (In the figure, a is the silicon lattice constant, $b_\perp$ and $b_\parallel$ are the alloy lattice constants perpendicular and parallel to the alloy/silicon interface, respectively.) The 100 Å alloy film is the most strained of the series, the lattice constants being equal to the substrate in plane and 3% larger in the normal direction. As the thickness increases, the two lattice constants converge progressively towards an intermediate value which is 2% larger than that of the silicon substrate as would be expected in a relaxed $Ge_{0.5}Si_{0.5}$ alloy. Because the x-ray penetration depth was not well defined for the in-plane measurements, it is not possible to say how much gradation of lattice constant exists across the films. The measured values are, therefore,

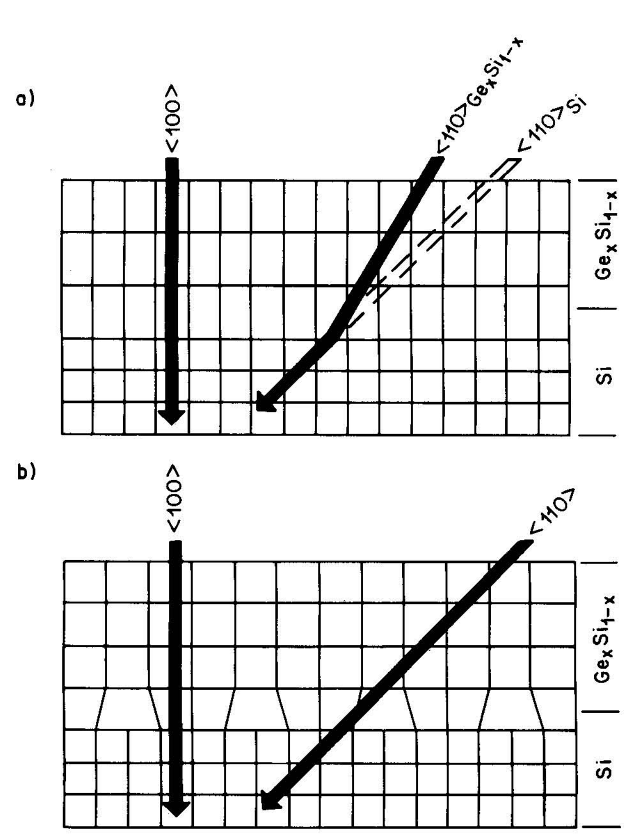

FIG. 5. Schematic illustrations of commensurate (a) and incommensurate (b) epitaxial growth. Lattice mismatch exaggerated for clarity. Note shift of off-normal crystalline channels in commensurate films.

weighted averages with the major contribution arising from the topmost 200 Å.

Backscattering measurements also provide a measure of alloy lattice distortion and were used to document the com-

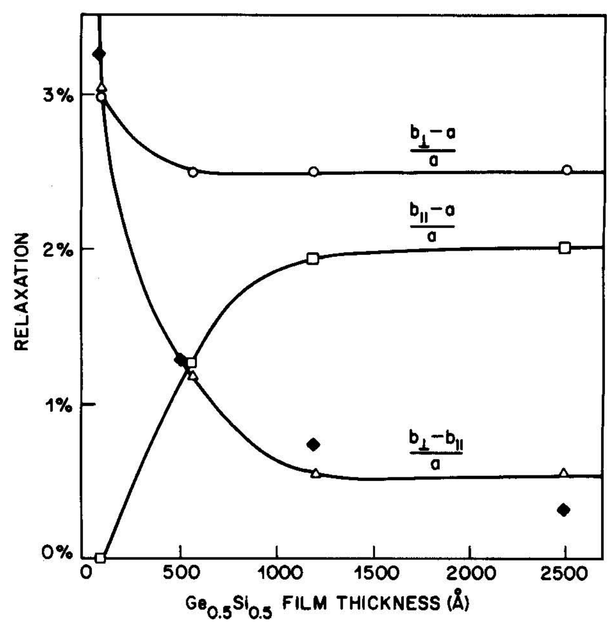

FIG. 6. Results of x-ray measurements of lattice relaxation in $Ge_{0.5}Si_{0.5}$ films of 100, 500, 1000, and 2500 Å thicknesses on Si. Commensurate/incommensurate transition occurs between 100 and 500 Å thickness where $b_\parallel$ ceases to equal a. X-ray data shown by open symbols, comparative backscattering data by filled symbol.

mensurate/incommensurate transition in a variety of samples of differing thickness and composition. As indicated at the top of Fig. 5, for commensurate growth the ⟨110⟩ alloy channel is misaligned with respect to the ⟨110⟩ substrate channel. Backscattering measurements of this angular displacement permit calculation of the tetragonal distortion in the alloy according to the relationship

$$\epsilon_T = (b_\perp - b_\parallel)/b = -\Delta\theta/\sin\theta\cos\theta,$$

where θ is the angle between the ⟨110⟩ and normal ⟨100⟩ channels and b is the relaxed alloy lattice constant. These values are plotted in Fig. 7 as a function of alloy composition with film thickness (100, 500, 1000, 2500 Å) shown as a parameter. Also plotted in the figure are x-ray data on $(b_\perp - b_\parallel)/a$, which is effectively equal to ϵ_T given that $b \sim a$.

For each film thickness, strain increases roughly linearly with alloy composition then falls towards zero. If simple elasticity theory applies, the strain in the commensurate (dislocation free) films should vary as

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

where ν is Poisson's ratio and f is the lattice mismatch between silicon and a bulk alloy of composition x,[19] i.e.,

$$f = (b - a)/a.$$

This relationship is given approximately by $f = 0.042x$, where 0.042 is the mismatch between pure Si and Ge. This calculated distortion is indicated as the straight line in Fig. 7 and it is evident that thin or dilute alloy layers behave in near ideal manner. The point at which data fall from the straight line marks the onset of incommensurate growth or, equivalently, the onset of misfit dislocation formation. These points are tabulated in the inset of Fig. 7, which plots the critical thickness for alloys of different composition. The inset shows that whereas a $Ge_{0.2}Si_{0.8}$ film will grow commensurately on Si to 2500 Å, a $Ge_{0.5}Si_{0.5}$ film will grow commensurately only to 100 Å. Critical thicknesses have been verified directly by TEM cross sectioning of alloy layers.[10] The magnitude of the critical thicknesses is significantly larger than predicted by equilibrium theory[1] and strongly suggests that with our low temperature growth technique there is a kinetic barrier to dislocation formation and/or migration.

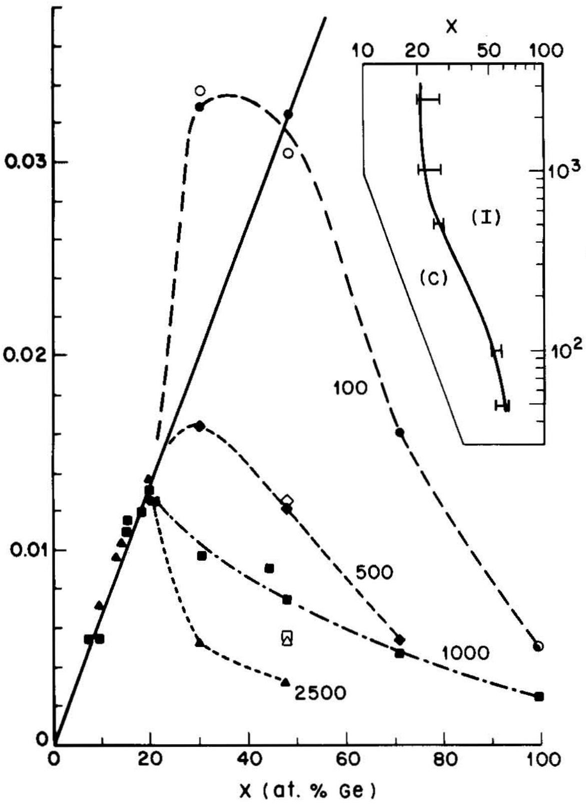

FIG. 7. Tabulation of tetragonal strain $(b_\perp - b_\parallel)/b$ in alloy layers of varying thicknesses and composition. Straight line gives calculated strain for ideal commensurate growth. Fall from straight line marks incommensurate transition for a given film thickness. Critical thicknesses plotted in inset. Solid symbols represent backscattering data, open symbols x-ray data.

V. STRAINED-LAYER SUPERLATTICE

The values of critical thickness given in Fig. 7 were used to design a Ge_xSi_{1-x}/Si strained-layer superlattice. In contrast to earlier work,[2–7] the aim was to make al layers commensurate with the substrate (i.e., we did not initially grade to a lattice constant intermediate to the Si and alloy values). To maintain this condition silicon layers were grown significantly thicker than intermediate alloy layers. The chosen sequence was (from the substrate upwards):

125 Å $Ge_{0.3}Si_{0.7}$/500 Å Si/125 Å $Ge_{0.3}Si_{0.7}$/
500 Å Si/125 Å $Ge_{0.3}Si_{0.7}$/100 Å Si.

Backscattering analysis confirmed that all layers were epitaxial with no detectable disorder. This is shown directly in the TEM cross section of Fig. 8 which was taken under bright field $g = (400)$ diffraction conditions to emphasize changes in lattice constant perpendicular to the (100) crystal surface. The Ge_xSi_{1-x} and Si layers are clearly distinguishable as dark and light bands. TEM images taken under $g = (022)$ diffraction conditions showed only minor contrast between layers confirming the commensurate nature of growth. Under both diffraction conditions, no dislocations were evident anywhere in the TEM sample.

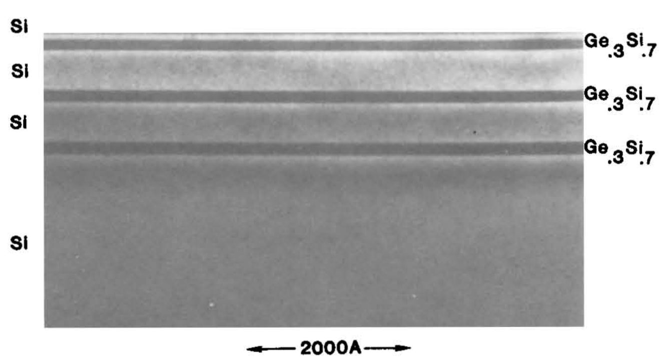

FIG. 8. TEM cross section of Ge_xSi_{1-x}/Si strained-layer superlattice. Layer sequence from bottom: Si substrate/125 Å $Ge_{0.3}Si_{0.7}$/500 Å Si/125 Å $Ge_{0.3}Si_{0.7}$/ 500 Å Si/125 Å $Ge_{0.3}Si_{0.7}$/ 100 Å Si. Note absence of dislocations.

VI. CONCLUSION

We have described conditions for MBE growth of smooth, commensurate, Ge_xSi_{1-x} films on Si. The commensurate nature of growth was confirmed by electron microscopy, Rutherford backscattering, and x-ray analysis. A commensurate/incommensurate transition was observed and used to tabulate values of critical thickness. These thicknesses are significantly larger than observed in earlier studies or predicted by theory. Finally, commensurate layers were combined to form the first Ge_xSi_{1-x}/Si strained-layer superlattice.

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