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Low-temperature homoepitaxial growth on Si(111) through a Pb monolayer

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A monolayer of Pb mediates high-quality homoepitaxial growth on Si (111) surfaces at temperatures where growth with other overlayer elements or on bare surfaces leads to amorphous or highly defective crystalline films. Nearly defect-free epitaxy proceeds for film thicknesses up to 1000 Å with no sign that this is an upper limit. The minimum temperature for high-quality epitaxy depends on the substrate miscut. For a 0.2° miscut, the minimum temperature is 340 °C. Films grown on substrates miscut 2.3° towards [$11\bar{2}$] show good crystalline quality down to 310 °C. © *1998 American Institute of Physics.* [S0003-6951(98)01747-1]

Since the discovery that thin metal overlayers on Si surfaces can change the growth mode of deposited semiconductor films,[1] much work has been devoted to describing and exploiting the processes by which growth occurs.[2,3] Characteristic features of the growth of semiconductor films through metal layers include the promotion of epitaxial growth at low temperatures and the segregation of the overlayer impurity to the surface of the growing film. Elements from groups III and V,[4] as well as Au[5,6] and Sn,[7] have been shown to promote Si (111) homoepitaxy at temperatures lower than those necessary on bare surfaces. Reducing the growth temperature mitigates the effects of dopant diffusion, reaction and evaporation of surface metallizations, and thermal stress. With the exception of Au, overlayer elements have been used to produce only limited film thicknesses, between 10 and 200 Å. All of the previously studied elements except Sn are electrically active impurities in Si and Ge, further limiting their practical use. The promising results using Sn have not been followed up with a study of thicker films and were achieved only when the Sn overlayers were deposited or annealed with the substrate at 500 °C.

In this letter we report homoepitaxial growth on Si (111) at temperatures of 340 °C on low-miscut substrates and 310 °C on vicinal substrates using a 1 ML (1 ML $=7.83\times10^{14}$ atoms/cm^2) Pb overlayer. We have grown films up to 1000 Å thick with no sign of decreasing quality with increasing thickness. As with other overlayer elements, the Pb layer segregates from the growing crystal to stay at the surface. The 310 °C growth temperature represents an improvement of 150 °C in the temperature at which thick, high-quality homoepitaxial Si (111) films may be grown using Au overlayers and an improvement of approximately 350 °C compared to growth on bare surfaces.[8] Without Pb, epitaxial growth at these temperatures breaks down within a few nanometers of the substrate–film interface, resulting in an amorphous film.

Previous studies of the behavior of Pb on Si (111) surfaces have shown that Pb allows near-surface Si atoms to move at low temperatures. For example, with 1 ML Pb the 7×7 reconstruction of the clean surface is changed to an incommensurate arrangement after annealing to temperatures below 300 °C.[9] The change in reconstruction indicates that the Pb enables the removal of the stacking fault present in half of the 7×7 unit cell. In addition, atomic scale Pb diffusion on the Si (111) 7×7 surface has been observed at temperatures as low as 25 °C.[10] As in the case of Sn, there are no known electronic deep levels associated with Pb impurities in Si.[11] Recent work using Pb layers to promote the growth of Si[12] and Ge[13] films has been limited to films a few atomic layers thick, deposited at or near room temperature.

The 500-μm-thick Si (111) wafers used in this study were cleaved into 1 cm×2 cm samples, chemically oxidized by the Shiraki process,[14] and loaded into an ultrahigh vacuum growth chamber with a base pressure of 1×10^{-10} Torr. To investigate the effect of miscut on growth quality, we used two types of wafers, one miscut 0.2±0.1° (0.1–0.4 Ω cm, P doped) and the other miscut 2.3±0.1° towards [$11\bar{2}$] (>1000 Ω cm, P doped). Using direct current resistive heating, each sample was degassed at 500 °C for one hour, heated to 850 °C to remove the oxide, cooled at a maximum rate of 1 °C/s to 500 °C and then slowly cooled to the growth temperature. Because the thermal time constants of the sample holder system were long, we left each sample at the growth temperature for up to 24 h prior to Pb and Si depositions to achieve steady-state heat flow conditions. Sample temperatures were measured with an optical pyrometer, calibrated to the resistance of a sample that displayed intrinsic variation of the carrier concentration with temperature above 150 °C. The resistance of the intrinsic sample was a sensitive function of temperature in the range of interest. All temperatures are accurate to within ±10 °C.

Depositions of Pb and Si were done using an effusion cell and an electron beam heated rod, respectively. Lead coverages were determined *in situ* to within 5% using Rutherford backscattering spectrometry (RBS). The Si deposition rates were 0.12±0.02 Å/s at a system pressure of approximately 2×10^{-9} Torr. The evaporation sources were masked to produce partially overlapping regions in which Pb and Si were deposited, allowing simultaneous growth of films with and without 1 ML Pb. Imperfections in the masking process yielded a low Pb flux, below 1% of that in the direct Pb beam, to the area where nominally only Si was to be deposited. At all growth temperatures studied, 295–340 °C, with 1 ML Pb coverage the sample displayed a 1×1 low energy

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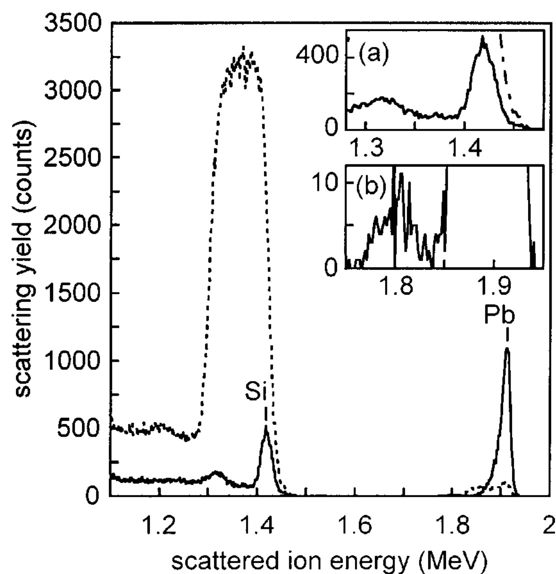

FIG. 1. Ion channeling spectra for 1000 Å Si films grown on Si (111) at 340 °C with (solid line) and without (dashed line) codeposition to achieve 1 ML steady-state Pb coverage. The improvement in crystal quality with 1 ML Pb present during growth dramatically reduces the scattering yield from the film. Inset are enlargements of the vicinities of (a) the Si surface peak, and (b) the Pb peak at 1.8 MeV due to 0.01 ML Pb at the substrate–film interface.

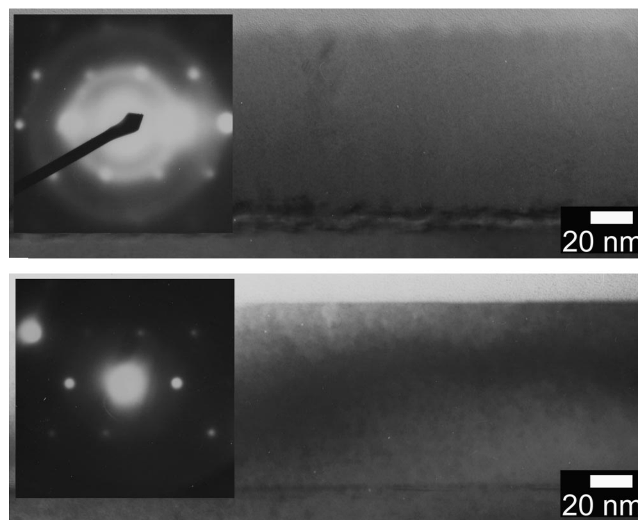

FIG. 2. Transmission electron micrographs with the incident beam along a ⟨110⟩ direction from Si films grown at 340 °C on Si (111) without Pb (top) and with codeposition to achieve a 1 ML Pb overlayer (bottom). Electron diffraction patterns (inset) reveal that the film grown without Pb is amorphous and that the film with Pb is an epitaxial extension of the crystal. The crystalline reflections in the top diffraction pattern are from the substrate.

electron diffraction (LEED) pattern, which is consistent with previous observations.[15] Upon cooling the samples to room temperature after growth, we observed the high-coverage rotated incommensurate (IC) LEED pattern in the areas of the sample where 1 ML Pb was present.[9] Regions on which neither Si nor Pb was deposited displayed bright, sharp 7×7 LEED patterns.

We have found that significant desorption of Pb occurs at temperatures above 280 °C. In order to keep the Pb coverage constant during growth of the Si film, Si and Pb were codeposited. Achieving a steady-state 1 ML coverage of Pb required a Pb flux that ranged from 0.12 ML/min at 295 °C to 0.18 ML/min at 340 °C. These fluxes correspond to desorption rates that are several times higher than one would expect due to evaporation from bulk Pb, according to vapor pressure measurements.[16] Similarly enhanced desorption rates have been observed at higher temperatures.[9] The rapid increase in desorption rate at coverages just higher than 1 ML, as observed in that study, simplifies the task of achieving 1 ML Pb through codeposition.

The samples were characterized after growth by *ex situ* 2 MeV $^4He^+$ ion channeling measurements with the incident beam along [111] and the detector aligned in a glancing exit geometry. Figure 1 shows two spectra from a film deposited at 340 °C on a low-miscut sample. The film grown on a region of the sample with only incidental Pb coverage (approximately 0.2 ML Pb distributed throughout the film) shows scattering consistent with growth of a 1000 Å amorphous layer. We observe no change in the growth of the amorphous film due to the small amount of Pb present; films with no Pb contamination are identical. In comparison, the film grown with codeposition to maintain 1 ML Pb during growth shows a dramatic reduction in scattering yield. The minimum yield, χ_{min}, which is the ratio of the ion-scattering yield behind the Si surface peak for the incident beam in a channeled direction to the yield for random incidence, is 2.4 ±0.3% for the grown sample, identical within measurement error to χ_{min} measured for a clean Si (111) sample on which 1 ML Pb had been deposited, 2.2±0.3%. The area under the peak at 1.9 MeV due to scattering from Pb corresponds to a Pb coverage of 1.07±0.05 ML. Small islands of Pb on the surface are responsible for the slight elongation of the Pb peak towards low energy. A lesser peak at 1.8 MeV corresponds to 0.01 ML Pb at the substrate–film interface. The silicon surface peak at 1.4 MeV is indistinguishable from that of Si (111) with 1 ML Pb on which no homoepitaxial film has been grown, indicating no detectable increase in disorder at the surface. The subsurface scattering peak at 1.3 MeV, a ''close encounter''[17] between channeled ions and Si atoms at lattice sites, also appears in spectra taken on bare surfaces. That the surfaces remain highly ordered after growth is confirmed by the observation of the IC LEED pattern.

Images and diffraction patterns obtained by cross-sectional transmission electron microscopy (TEM) with the incident beam along a ⟨110⟩ direction are shown in Fig. 2 for the films for which ion channeling spectra appear in Fig. 1. The film grown without Pb codeposited exhibits a diffraction pattern consisting of rings superimposed on the sharp spots due to diffraction from the substrate. The ring spacing indicates that the film is amorphous Si.[18] The amorphous film has a rough interface with the substrate indicating initial polycrystalline growth. In comparison, the film grown with 1 ML Pb shows high-quality epitaxy. We associate the line visible at the substrate–film interface with the 0.01 ML of trapped Pb. The film produces a sharp bright diffraction pattern identical to that of the substrate.

Films grown with codeposition rates giving Pb coverages less than 1 ML are defective. Examination of these films by TEM shows the defects to be stacking faults that nucleate at the substrate–film interface. At 340 °C, growth with 0.90±0.05 ML Pb consistently yields films with χ_{min}

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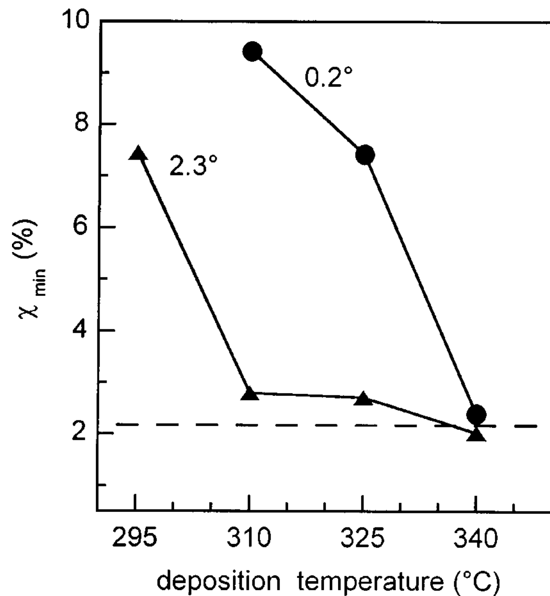

FIG. 3. The minimum yield χ_{min} measured for 500–1200 Å films grown on 0.2° (circles) and 2.3° (triangles) miscut Si (111) substrates. Growth of high-quality films is possible at lower temperatures on the 2.3° miscut substrates. The dashed line indicates χ_{min} measured for a wafer on which 1 ML Pb had been deposited.

$=5\%$, due to grown-in defects. With more than 1 ML Pb present on the surface, the excess Pb forms three-dimensional islands which can extend up to 1 μm in height, as measured by scanning electron microscopy. When the total Pb coverage is only slightly greater than the equivalent of 1 ML, ion channeling measurements indicate that the islands are surrounded by 1 ML Pb. Under these conditions, high quality epitaxial growth can still proceed. The formation of Pb islands at the lowest growth temperatures studied is consistent with the high mobility of Pb on the Si (111) surface.

For the samples produced from the low-miscut substrate, the quality of the grown film deteriorates for growth temperatures below 340 °C. The minimum yield χ_{min} increases rapidly with decreasing growth temperature, indicating a sharp decrease in the crystalline quality of the film. Films grown at 310 °C have $\chi_{min}=9\%$. As is the case at higher temperatures with homoepitaxy on bare Si (111), stacking faults nucleating at or near the substrate–film interface and propagating through the film are the dominant extended defect. We find, however, that the minimum temperature at which a high-quality film can be grown is a function of the miscut orientation of the substrate. Figure 3 compares the measured χ_{min} of 500–1200 Å films grown on 0.2° and 2.3° miscut substrates. Films of excellent crystalline quality can be grown on the 2.3° miscut samples for substrate temperatures as low as 310 °C. The lowering of the minimum temperature at which high-quality growth is observed from 340 °C for low-miscut substrates to 310 °C for stepped substrates is a clear indication that the step edges have a kinetic role in the film growth. Reflection electron microscopy experiments on Sn-mediated Si (111) homoepitaxy[7] and ion channeling experiments with Au overlayers[6] reached similar conclusions. We continue to investigate the mechanism responsible for the high-quality growth in the Pb-mediated case.

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