

# Effect of substrate miscut on low-temperature homoepitaxial growth on Si(111) mediated by overlayers of Au: Evidence of step flow

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Observations of homoepitaxial growth on low-angle miscut ( $\sim 0.1^\circ$ ) Si(111) substrates through an overlayer of Au, together with earlier results on highly miscut Si(111) surfaces, indicate that growth in this system occurs by step flow. The growth temperatures were between 375 and 500 °C. In the optimum range of Au coverage (0.6–1.0 ML), ion channeling measurements yield at best  $\chi_{\min} = 5.0\%$ , and cross-sectional transmission electron microscopy reveals stacking faults on (111) planes. Films produced under similar conditions on bare Si(111) substrates are much more defective. On the other hand, the defect density in the present films is higher than that in films grown on substrates with a higher miscut angle. The improvement in film quality resulting from the Au overlayers is attributed to an increase in the diffusion length of the Si adatoms, caused by Au passivation of the Si terraces. It is suggested that Au is more efficient than other overlayers in promoting step flow because Au passivates the Si(111) terraces *without* passivating the step edges.  
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Epitaxial growth of thin films using mediating overlayers<sup>1</sup> is of interest because this technique can produce high quality films at temperatures considerably below those required for growth on bare substrates. Another advantage of using mediating overlayers has been shown for homoepitaxial and heteroepitaxial growth where the overlayer can actually alter the growth mode of the process. In particular, heteroepitaxial growth of Ge on Si has been shown to occur by two-dimensional (2D) island growth rather than by the thermodynamically preferred three-dimensional island growth.<sup>2–8</sup> For the case of homoepitaxy, growth on Si(111) is inherently difficult at low temperatures. The substrate must typically be held above 700 °C<sup>9</sup> during deposition to obtain high quality films of unlimited thickness. At lower temperatures, Weir *et al.*<sup>10</sup> demonstrated homoepitaxial growth on bare Si(111) for limited thicknesses at growth temperatures of 380–450 °C. The films were about 350 Å thick, and the epitaxial quality deteriorated rapidly with distance from the initial interface. Buffer layers predeposited at 700 °C were necessary to obtain clean substrate surfaces.

Several studies have shown<sup>5,11–17</sup> that mediating overlayers can be used to improve homoepitaxial growth on Si(111). Most of these studies, however, have been on very thin epitaxial layers [1–50 ML of Si, where 1 ML =  $7.84 \times 10^{14}$  atoms/cm<sup>2</sup> on Si(111)], and therefore offer only limited insight into development of the microstructure. For Sn and Au overlayers on Si(111), Iwanari *et al.*<sup>11,12</sup> and Minoda *et al.*,<sup>16</sup> respectively, observed indications of step flow by monitoring growth of the first few monolayers with reflection high-energy electron diffraction and reflection electron microscopy. The development of the microstructure in thicker layers grown through Au on Si(111) was reported in our earlier letter,<sup>17</sup> where we showed that defect-free homoepitaxial films as thick as 5000 Å (with no indication that this was an upper limit) could be grown at temperatures as

low as 450 °C by using a 0.6–1 ML Au mediating layer.

The miscut angle of the substrates used in our earlier study was 2.63°. To investigate the importance of step flow, we performed a series of growth experiments on Au-covered Si(111) with low miscut angles, which have larger terrace widths. Samples were obtained from 3-in. *n*-type wafers, with  $\rho = 0.1$ – $0.4 \Omega \text{ cm}$  and a miscut angle less than  $0.1^\circ$  toward the  $\langle 11\bar{2} \rangle$  direction. The oxide desorption and film depositions were done in an ultrahigh vacuum (UHV) chamber, with a base pressure of  $7.0 \times 10^{-11}$  Torr. The experimental procedure used has been described in detail elsewhere.<sup>17</sup> Following epitaxial growth, the samples were analyzed by Rutherford backscattering spectrometry (RBS) using 2 MeV  $^4\text{He}^+$ , and ion channeling. After RBS, most of the samples were studied by cross-sectional transmission electron microscopy (TEM).

The substrate temperature and the Au coverage were varied to determine the effect of these parameters on the epitaxial quality of the film. In the previous study,<sup>17</sup> it was found that Si deposition rate had no observable effect on the film quality. Au coverages were in the range of 0.31–1.70 ML. Low-energy electron diffraction (LEED) and RBS showed the following surface reconstructions before and after Si deposition at 375–500 °C: mixed  $5 \times 1$  and  $7 \times 7$  for less than 0.4 ML of Au, mixed  $5 \times 1$  and  $(\sqrt{3} \times \sqrt{3})R30^\circ$  between 0.4 and 0.8 ML of Au; only  $(\sqrt{3} \times \sqrt{3})R30^\circ$  (or  $6 \times 6$ , above 1 ML) above 0.8 ML of Au. These observations are in agreement with earlier LEED and scanning tunneling microscopy (STM) studies.<sup>18</sup>

Figure 1 shows four sets of RBS data for deposition at 450 °C, at a deposition rate of about 1.0 Å/s. Figure 1(a) is for a control sample with no (0.0 ML) Au on the surface. The film is highly defective, and, as was seen for the analogous samples with higher miscut angles,<sup>17</sup> the film quality deteriorates rapidly starting at the interface. The present film, however, is much more defective, with  $\chi_{\min} = 60\%$ . This poor film quality is confirmed by TEM [Fig. 2(a)], which reveals a high stacking fault density throughout the film. Unlike in the films grown on highly miscut bare Si(111), the

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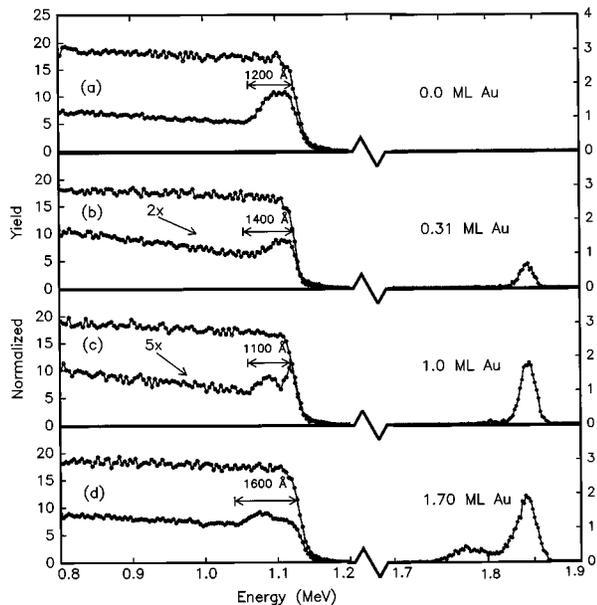


FIG. 1. Random and aligned Rutherford backscattering spectra of Si grown on  $0.1^\circ$  miscut Si(111) through overlayers of different thicknesses; all substrates were at  $450^\circ\text{C}$ . The normalized counts for the Au peaks at high energy are on the right axis. (a) 1200 Å film grown with no (0.0 ML) Au. (b) 1400 Å film grown with 0.31 ML of Au. (c) 1100 Å film grown with 1.0 ML of Au. The aligned yields in (b) and (c) have been multiplied by a factor of two and five, respectively, to show the detail in the spectra. (d) 1600 Å film grown through 1.70 ML Au with a large amount of Au trapped in the film.

stacking faults in the present film do not all begin at the interface; some stacking faults nucleate within the film. Further, several separate crystals can be seen within the film. Even though this sample has the poorest quality of all the

ones grown on low-miscut substrates, the film surface is still smooth after 1200 Å of growth.

Figure 1(b) is for a 1400 Å film grown with 0.31 ML Au, and resembles that for the film grown with no Au [Fig. 1(a)]. The film grown with Au, however, is of higher quality, with  $\chi_{\min}=21\%$ . The aligned spectrum has been multiplied by a factor of 2 to show its similarity to that of the film grown with no Au. The corresponding TEM image [Fig. 2(b)] shows that the film contains stacking faults on (111) planes. This film has a higher density of stacking faults than a film grown on a higher miscut angle substrate<sup>17</sup> deposited with a lower Au coverage (0.15 ML) (see Fig. 3).

Figure 1(c) shows the random and aligned spectra for a 1100 Å film grown with 1.0 ML Au. The Au coverage range 0.8–1.0 ML is optimal for both miscut angles. The aligned spectrum shows that the film is somewhat defective, with  $\chi_{\min}=9.2\%$ ; the defect density appears constant throughout the film thickness. The aligned spectrum has been multiplied by a factor of 5 to show detail. This quality is considerably lower than that of the corresponding film grown on the highly miscut substrate, which was nearly defect-free (see Fig. 3). The TEM image of Fig. 2(c) shows a few stacking faults that nucleated at the interface and extend into the film. The surface peak for Au at high energy in Figs. 2(b) and 2(c) shows that in both cases the Au remained at the surface during growth.

Figure 1(d) shows the spectra for a 1600 Å film grown with 1.70 ML Au. The high energy portion of the spectra shows that there is significant trapping of Au at the interface and throughout the film. The TEM image [Fig. 2(d)] shows that stacking faults are not present in the Si film, but that Au

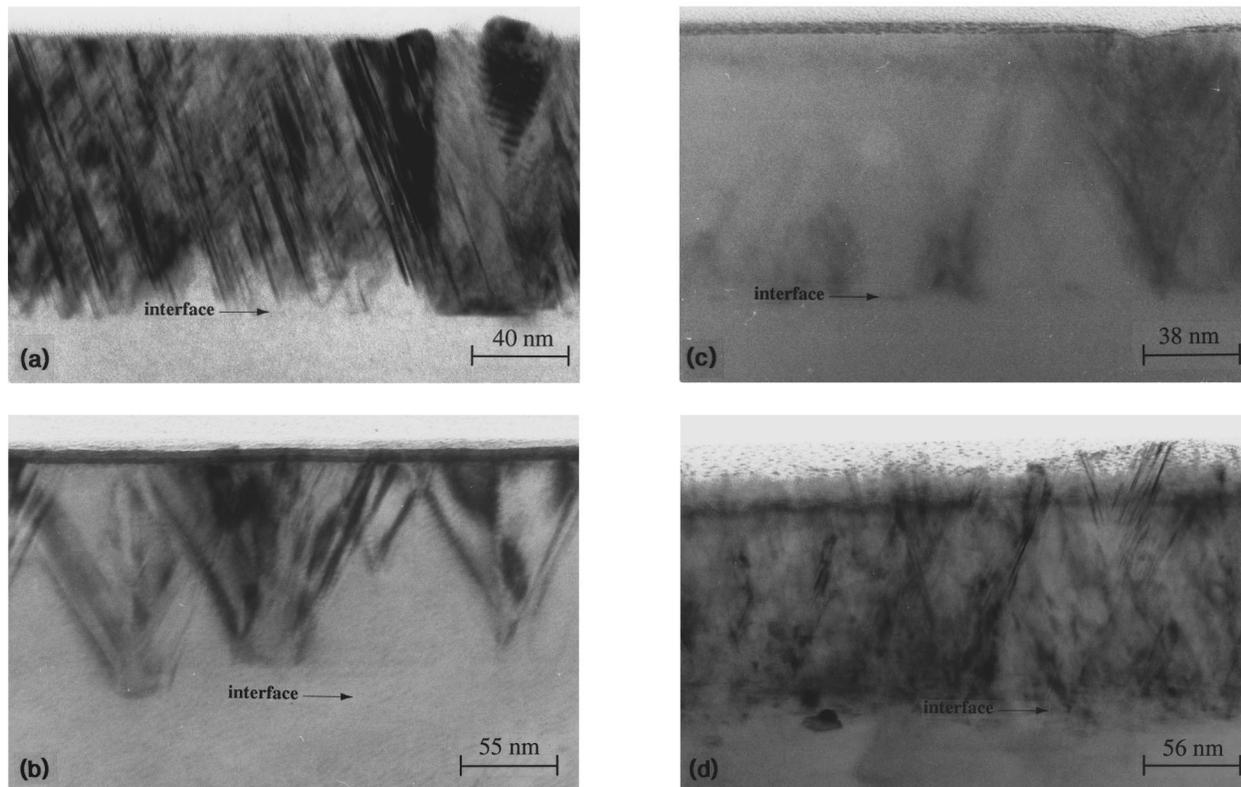


FIG. 2. Transmission electron micrographs of the samples in Fig. 1. Au overlayer coverages during growth: (a) 0.0; (b) 0.31; (c) 1.0; and (d) 1.70 ML.

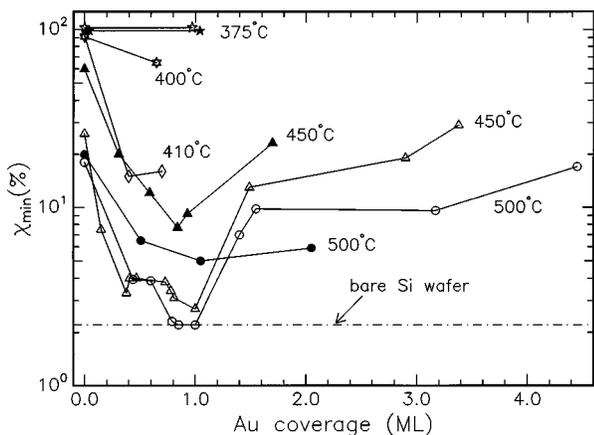


FIG. 3. Plot of  $\chi_{\min}$  vs Au coverage at several temperatures, for deposition rates of about 1 Å/s. Open symbols represent data points from films grown on highly miscut ( $2.63^\circ$ ) Si(111) (Ref. 17); filled symbols represent data points from films grown on low-miscut ( $0.1^\circ$ ) Si(111) in the present study. The data at  $375^\circ\text{C}$  for both substrates have  $\chi_{\min}=100\%$  even with 1 ML Au. These films are still fully crystalline, albeit highly defective.

inclusions cause the large amount of dechanneling in the spectrum of Fig. 1(d), with  $\chi_{\min}=23\%$ . Unlike films grown with 3.0 ML of Au on a more miscut substrate,<sup>17</sup> this film contains some stacking faults near the surface of the grown layer. This further supports the trend, illustrated by Fig. 3: the quality of films on low miscut substrates is much lower than that for the corresponding Au coverages on higher miscut substrates at the same temperature.

Figure 3 is a summary plot of  $\chi_{\min}$  as a function of Au coverage and substrate temperatures ranging from 375 to 500 °C, at deposition rates of 1.0–3.0 Å/s for films deposited on low- and high-miscut substrates. Open symbols represent data from our previous study<sup>17</sup> and filled symbols represent data from the low-angle miscut substrates used in the present study. The largest error in  $\chi_{\min}$  values was 0.2% for all samples measured. Note that the trend is the same for both types. In both cases, the minimum is found at about 1 ML Au. In this case,  $\chi_{\min}=5.0\%$  for low-miscut substrates, which is significantly worse than that for films grown on higher-miscut substrates, where  $\chi_{\min}=2.2\%$ . For both miscut angles at low Au coverages, the deposited films are defective because of a high stacking fault density. At Au coverages much above 1 ML, however, the films are poor because of a large amount of dechanneling from Au inclusions within the film. Although at  $375^\circ\text{C}$ ,  $\chi_{\min}=100\%$  for 1.0 ML Au on the low-miscut (and high miscut) substrate, TEM reveals that the films are still crystalline, albeit highly defective, and not amorphous.

The main difference between the low- and high-miscut substrates is the width of their terraces:  $\sim 1700$  and  $70$  Å, respectively, for the ones used in our two studies. The higher density of stacking faults found in films grown on low-miscut substrates under otherwise identical conditions can be explained by the higher probability of layer nucleation in the time it takes the Si adatoms to diffuse to a step edge for incorporation into the crystal. The difference in crystal quality between the two growth modes may therefore be considered as indirect evidence for step flow as a growth mechanism, since no such difference would be expected if the

growth were governed by two-dimensional island nucleation. Although others have shown step flow over larger terrace widths,<sup>11–13,16</sup> the films in our studies were grown at deposition rates of  $\geq 1$  Å/s, whereas the other studies used deposition rates of at most 1 ML/min [ $=0.025$  Å/s on Si(111)]. This difference in deposition rates accounts for the difference in observed terrace widths over which step flow occurs.

The most plausible interpretation of the role of the Au layer in the improvement of the crystal quality, for both miscut angles, over that grown on bare Si is that it increases the diffusion length of the Si adatoms by passivating the Si surface and thereby making the attachment of Si on the terrace less likely. Other overlayers that are known to passivate the Si(111) surface<sup>19</sup> such as As or Sb, however, have been observed to induce 2D island growth,<sup>5,15</sup> and are less effective than Au in improving the quality of the homoepitaxial crystal. This difference may be explained using the proposal by Kandel and Kaxiras<sup>20</sup> that all passivating overlayers increase the surface diffusion length, but that some overlayers such as As and Sb also passivate the step edges, whereas others such as Sn and Au do not. In support of this, Kandel and Kaxiras<sup>20</sup> have made specific arguments for the passivation of  $\langle 11\bar{2} \rangle$  steps by Sb atoms on the Si(111) surface, which makes it more likely for a Si adatom to exchange sites with an Sb overlayer atom on a terrace than with an Sb atom at a step, which would lead to 2D island growth despite an increased Si surface diffusion length.

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<sup>1</sup>The term “surfactant” has been widely used to describe a thin overlayer which mediates crystal growth. We prefer to reserve use of the term for situations where it is clearly demonstrated that the mechanism for crystal growth is governed by a decrease in the interfacial energy. This certainly does not apply in the case of homoepitaxy.

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