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SUBSTRATE TEMPERATURE EFFECTS ON ($\sqrt{3}\times\sqrt{3}$) R30° DOMAIN GROWTH OF Ag ON Si(111) SURFACE

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Abstract

High-resolution LEED angular profiles associated with $(\sqrt{3}\times\sqrt{3})$R30° domain growth of Ag on Si(111) have been analyzed following Ag deposition as a function of substrate temperature. We found that the $\sqrt{3}$ domain size distribution has the same Gamma distribution at different coverages and substrate temperatures. However, at higher substrate temperatures the $\sqrt{3}$ structure grows with coverage primarily by domain coalescence as indicated by the considerable increase in the mean and width of the size distribution. As a result, scaling, i.e. self-similar domain growth, is observed. At lower substrate temperatures the $\sqrt{3}$ structure grows with coverage by simply increasing the number of small, randomly nucleated domains. Hence, the domain size distribution at low temperatures is narrow and varies little with coverage.

1. Introduction

The $(\sqrt{3}\times\sqrt{3})R30°$ (hereafter denoted $\sqrt{3}$) structure of Ag on Si(111) has been extensively investigated by almost every surface analysis technique [1-10]. Most work has focused on the determination of the atomic geometry of this structure, with little attention paid to the $\sqrt{3}$ domain growth. Using a high resolution low energy electron diffraction (HRLEED) technique, we have studied the $\sqrt{3}$ domain growth as a function of Ag coverage and substrate temperature. As a matter of practical interest, the initial epitaxial structure frequently determines the subsequently grown film. From a fundamental viewpoint, the two-dimensional growth characteristics, such as the scaling and domain size distribution with, for example, time, temperature, coverage, or deposition rate, have recently attracted great interest [11-13].

In spite of great effort, there is still no consensus concerning the atomic geometry of the $\sqrt{3}$ structure and the corresponding saturation coverage θ_s. However, the honeycomb arrangement of atoms in the topmost layer was unequivocally confirmed by scanning tunneling microscopy (STM) images [2,5]. Since STM is not yet capable of elemental recognition, one group concluded that the honeycomb geometry in the STM image resulted from Si atoms atop Ag trimers (embedded trimer model) in which $\theta_s = 1$ ML (1 monolayer corresponds to 7.83×10^{14} atoms/cm^2, the area density of unreconstructed Si(111) surface). Another group proposed a honeycomb arrangement of Ag adatoms (honeycomb model) in which $\theta_s = 2/3$ ML. More recently, results from x-ray diffraction intensity measurements suggested a new model in which the Ag trimers are chained in a honeycomb arrangement (honeycomb chained triangle model). In this HCT model the saturation coverage is 1 ML. These three main models are each supported by various experiments [3,4,6,7,9,10].

In the present work we do not address the detailed $\sqrt{3}$ structure, but emphasize instead the effects of coverage and temperature on $\sqrt{3}$ domain growth. In particular, we address the evolution of the domain size distribution and scaling behavior.

2. Experimental

The experiments were performed in a UHV chamber with base pressure of $6.5–8.0 \times 10^{-11}$ Torr. The chamber is equipped with HRLEED, Auger Electron Spectroscopy (AES) using a double-pass cylindrical mirror analyzer, and a Ag evaporation source. The HRLEED (or Spot-Profile-Analysis LEED [14]) is interfaced with a personal computer. It is operated with a spatial resolution $\leq 6 \times 10^{-3}$ Å^{-1} in k-space. Also, a good signal to noise ratio is provided by using a channeltron detector. These features allow us to accurately and quickly record the angular profile of any diffraction beam, so that the evolution of the ordered domains can be determined.

The Si samples with size ~ 0.9×0.9 cm^2 were cut from a n-type Si(111) wafer. The misorientation from (111) is less than 0.2°. The sample, precoated by a very thin oxide layer using a chemical method [15], was mounted in a Mo housing in which the sample can be heated or cooled below room temperature. Temperature was measured by a W5%Re-W26%Re thermocouple in contact with a corner of the sample. The Si(111) sample was first annealed at ~ 1200 °C to remove the oxide layer and carbon impurities. Later, to desorb Ag overlayers from the substrate and restore the clean Si(111)7×7 surface, the sample usually was annealed at ~900–1000°C. After the annealing, no contamination was detectable with AES and the clean Si(111) surface exhibited a sharp 7×7 LEED pattern. The average substrate terrace was measured to be equal to or larger than the HRLEED transfer width, i.e. > 1000 Å. Ag atoms were deposited on the clean Si(111)7×7 surface at a fixed rate of ~0.2 ML/min by evaporation from a pure (5N) Ag foil heated by electron bombardment from the backside. We found that this evaporation method can produce a contamination free Ag deposition within our AES detection limit. An approximate calibration of Ag coverage was obtained by assigning the coverage at the break point in the plot of Auger intensity vs deposition time to be ~1 ML. Therefore, all coverages denoted hereafter are the nominal coverages. The absolute coverage for the $\sqrt{3}$ structure is not critical in the present study. For each LEED measurement, the deposition was interrupted and the sample cooled to near room temperature. The measurements have been repeated on two samples and all results were consistent.

3. Results and discussion

For submonolayer coverages of Ag deposited on Si(111) above ~200°C, a $\sqrt{3}$ superlattice forms which is detectable at less than 0.1 ML. By measuring the angular profile of a $\sqrt{3}$ superlattice diffraction beam, we are able to determine the $\sqrt{3}$ domain evolution as a function of Ag coverage (θ) and substrate temperature (T) since it depends solely on the domain size distribution. Figure 1 is a plot of the full width at half-maximum (FWHM) of a $\sqrt{3}$ superlattice diffraction beam vs θ at different temperatures ranging from 350 to 450 °C. The incident electron energy is chosen to be 84 eV for which the intensity is a maximum. From Fig.1 we can see that, for any θ the FWHM decreases with increasing temperature, and for any T, the FWHM behaves in the same manner with increasing coverage. Since the FWHM is inversely proportional to the average domain size, this indicates that the $\sqrt{3}$ domains grow with either increasing temperature or coverage. Also, the FWHM levels off at lower coverage with decreasing temperature. This implies that at lower temperature, the $\sqrt{3}$ structure is limited to smaller, randomly nucleated domains during deposition. This may be due to insufficient activation energy for Ag atoms to diffuse long distances or to eliminate the domain boundaries.

In order to quantitatively see the evolution of $\sqrt{3}$ domain size distribution as a function of coverage and temperature, we have employed an incoherent scattering model to fit the angular profiles. In this model, the $\sqrt{3}$ domains are assumed to be randomly distributed with no phase correlation. The intensity of any superlattice beam diffracted from this kind of domain structure is simply given by [16]

$$I(\underline{s},\theta) = \sum_{N} P(N,\theta) \frac{\sin^2 \frac{N}{2} (\underline{s}_{\|}\underline{a})}{\sin^2 \frac{1}{2} (\underline{s}_{\|}\underline{a})} , \tag{1}$$

where $P(N,\theta)$ is the domain size distribution function which describes the probability of finding a $\sqrt{3}$ domain with N inter-row spacings at a coverage θ, and $\underline{s}_{\|}$ and a are the momentum transfer parallel to the surface and the inter-row spacing (6.65 Å) of the $\sqrt{3}$ structure, respectively. We have tried several distributions including Guassian, Gamma and Raleigh distributions etc., and found that the Gamma distribution has the best fit. The Gamma distribution has the form:

$$P(N,\theta) = \frac{1}{\lambda^{\alpha}\Gamma(\alpha)} N^{\alpha-1} e^{-N/\lambda} , \tag{2}$$

with the mean $\bar{N} = \alpha\lambda$ and distribution width $\sigma = \sqrt{\overline{(N-\bar{N})^2}} = \lambda\sqrt{\alpha}$, where the α and λ were chosen as fitting parameters which are functions of θ and T. As α becomes large the Gamma distribution approaches a normal distribution. For $\alpha = 1$ it becomes an exponential distribution which is the continuum limit of the geometric distribution. Figure 2(a) shows the best fits, obtained utilizing the Gamma distribution in Eq. (1) convoluted with the instrument response function, to the angular profiles of a $\sqrt{3}$ superlattice beam at different coverages for T = 450°C. The best fits of the Gamma distribution for the different coverages all give $\alpha = 3.3 \pm 0.3$. However, λ increases drastically with a shift from 12.3 to 24.8 as the coverage ranges from 0.1 to 1.0 ML. The distribution width σ and the mean size $\bar{N}a$ are calculated to be 22.3 to 45.1, and 270 to 544 Å, accordingly. Figure 2(b) is a plot of the corresponding size distributions obtained from the fits in Fig. 2(a). As seen in Fig. 2(b), the increase of mean size and considerable broadening of the distribution width clearly imply that at higher T, domain coalescence is occurring with increasing Ag coverage to minimize boundary free energy. In this region one expects the existence of scaling because coalescence simply rescales length but leaves the basic morphology of domains unchanged [12,17]. Indeed, the Gamma distribution in Eq.(2) with fixed α can be written in a scaling form:

$$P(N,\theta) = \frac{1}{\alpha\lambda}\frac{\alpha^{\alpha}}{\Gamma(\alpha)} x^{\alpha-1}e^{-\alpha x} = \frac{1}{\bar{N}(\theta)} P'(x) , \tag{3}$$

where $x = N/\bar{N}$ and $P'(x) = \{\alpha^{\alpha}/\Gamma(\alpha)\}x^{\alpha-1}e^{-\alpha x}$ is a scaling function independent of coverage θ. Eq. (3) defines scaling, i.e., although the mean size varies with coverage θ, the functional form of size distribution does not. From Eq. (3), $P'(x) = \bar{N}P(N,\theta)$ is plotted in the inset of Fig. 2(b). As we can see, although $P(N,\theta)$ is broadening and its mean increases with θ, the $P'(x)$ curves superpose on each other, independent of coverage. 2D scaling was previously observed in the kinetics of 2D domain growth where the overlayer with a fixed coverage evolved with time from a disorder state to an order state [12]. To the best of our knowledge, this is the first observation of scaling with coverage in a 2D system.

In addition, we have also used a power Lorentzian,

$$I(\underline{s}) = \frac{A}{(s_{\parallel}^2 + \kappa^2)^m} \quad , \tag{4}$$

to fit the angular profiles of the $\sqrt{3}$ superlattice beam for different coverages with the inverse correlation length κ and exponent m as fitting parameters, where A is a constant. The best fits, plotted in Fig. 2(a) with dashed lines which are nearly identical to the Gamma distribution fits, all given $m = 1.5 \pm 0.1$ and κ varies with coverage. The occurrence of the same exponent m for different coverages implies that the angular profile $I(\underline{s})$ also contains a scaling form, $I'(x') = 1/(1 + x'^2)^m$ with $x' = s_{\parallel}/\kappa$, which is independent of θ. The angular profile depends solely on the domain size distribution. Therefore, it again proves the existence of scaling during the domain growth as a function of coverage. Additional information given by this line-shape is that, for large $s_{\parallel}$ this power Lorentzian will give Porod's law [18], $I(\underline{s}) \propto s_{\parallel}^{-3}$. Porod's law with exponent 3 is a well known consequence of scattering from a 2D random and compact domain structure in which domains are randomly distributed and the atom density is uniform within a domain. Thus, the $\sqrt{3}$ domains at higher temperature are quite compact even at low coverages. This power Lorentzian line shape with m ~1.5 was also obtained in the LEED measurements from the kinetics study of the Ge(111)($\sqrt{3}\times\sqrt{3}$)R30°-Ag system [13].

In contrast, we have employed the same model, Eq. (1), for the T = 350°C case. The Gamma distribution also gives the best fit among the trial distributions to the angular profiles of the $\sqrt{3}$ superlattice beam at different coverages. Best fit parameters are $\alpha = 4.5 \pm 0.3$ and $\lambda = 5.3$ to

7.3 for the coverage of 0.1 to 1.0 ML. Shown in Fig. 3 are the distributions at different coverages. The distribution width σ and the average domain size $\bar{N}a$ are calculated to be 11.0 to 15.5 and ~148 to 218 Å, accordingly. Compared with the higher temperature case, even though the distribution type is the same, the size distribution in this lower temperature case is much narrower and changes little with increasing coverage. It quantitatively indicates that at lower temperature the $\sqrt{3}$ superlattice grows with coverage primarily by increasing the number of small, randomly nucleated domains. Similarly, the power Lorentzian, Eq. (4) with $m = 1.9 \pm 0.1$, can also describe the angular profiles well. The larger m may result from the narrow size distribution of the randomly nucleated domains. The similar α in the Gamma distribution and m in the power Lorentzian for the different coverages also indicate that scaling exists in the lower temperature case. The scaling function $P'(x)$ for the T=350°C is plotted in the inset of Fig. 3, which is essentially independent of coverage.

4. Summary

By analyzing the HRLEED angular profiles of a $\sqrt{3}$ superlattice beam, we have studied $\sqrt{3}$ domain growth as a function of submonolayer coverage of Ag on Si(111) at different substrate temperatures. We found that the $\sqrt{3}$ domain size distribution has the same Gamma distribution at different coverages and substrate temperatures. But at higher temperatures the mean and width of the distribution increase considerably with coverage, indicating that $\sqrt{3}$ domains grow mainly by domain coalescence. At lower temperatures the size distribution is narrow and changes little with coverage, implying that the growth of $\sqrt{3}$ structure is limited to small, randomly nucleated domains. We also found that the angular profiles of the $\sqrt{3}$ superlattice beam for different coverages can be well described by a power Lorentzian with the same exponent. These results allow us to conclude that scaling in $\sqrt{3}$ domain growth with coverage does exist, which is observed here for the first time.

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Figure captions

Fig. 1. The FWHM of a $\sqrt{3}$ superlattice beam as a function of coverage at different substrate temperatures. The instrument response width has been removed.

Fig. 2. (a) The angular profiles of a $\sqrt{3}$ superlattice beam at different coverages for the T = 450°C case. The scan is along the $[\bar{2}11]$ direction. The solid lines are the best fits of Eq. (1) using the Gamma distribution. The dashed lines are the best fits of the power Lorentzian. These two kinds of fits are nearly identical and are almost indistinguishable in this figure. (b) Gamma domain size distributions with the best fit parameters obtained in (a) at different coverages. The inset is a plot of the corresponding scaling functions P'(x) vs x, which is independent of coverage.

Fig. 3 Gamma domain size distributions at different coverages obtained from the best fits of Eq. (1) to the angular profiles of a $\sqrt{3}$ superlattice beam for the T = 350°C case. The inset is a plot of the corresponding scaling function P'(x) vs x.