Facet Coexistence in the Roughening Transition of Ag(110)

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Abstract. We have studied the thermal behavior of the Ag(110) surface by synchrotron x-ray diffraction. In-plane diffraction data ($Q_z \approx 0$) agrees with earlier work and can be fit to a power law form. Out-of-plane data ($Q_{z,max}=0.7$), however, indicates the existence of two coexisting phases below the roughening transition: flat (110) oriented regions separated by inclined rough regions. The relative coverage of these two phases is found to depend on the temperature. Thus, the roughening process can be viewed as a continuous replacement of the flat faceted regions by the rough phase. Using the Wulff construction, we are able to describe the temperature dependence of the relative phase concentrations and extrapolate to an estimate of the Ag(110) roughening temperature.

Introduction. There is a great deal of interest in surface roughening, primarily because of its fundamental relationship to Equilibrium Crystal Shapes (ECS). The thermal evolution of the ECS of small Au and Pb crystals have been studied [1]. One of the results of those experiments was that close to the melting point only (100) and (111) facets persisted; all other orientations being rough by this temperature. Roughening of a flat surface has been predicted for several systems. Chui *et. al.*, showed that the Solid-on-Solid (SOS) model has a phase transition leading to a high temperature phase with a logarithmic height divergence [2],

$$\langle (h(r)-h(0))^2 \rangle \propto \ln(r), \tag{1}$$

where h(r) is the height of the surface a distance r from an arbitrary origin. Roughening transitions have been studied using x-ray diffraction [3,4], helium atom scattering [5,6] and LEED [7] for various metal surfaces. The expected diffraction lineshape has been calculated to have a power law form [4,6]

$$I(q_{\|},q_z) \propto \frac{1}{\sin^2(q_z/2)} \left| q_{\|}^2 \right|^{\eta(T,q_z)/2 - 1}, \tag{2}$$

where $q_{\|}$ and q_z are the parallel and perpendicular distances from the nearest Bragg point in reciprocal space and η is the roughness exponent ($\eta \propto q_z^2$). The first term in

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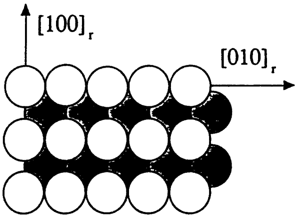

Fig. 1. Top view of the Ag(110) fcc surface. The rectangular coordinates system is shown (indicated by an index 'r'). Q is measured in units of [1.5377Å^{-1}, 2.1746Å^{-1}, 2.1746Å^{-1}]

eq. 2 is the crystal truncation rod (CTR) [8]. The second term is due to the roughness and broadens the CTR.

Held *et. al.* [3] studied Ag(110) in-plane lineshapes at $(1,0,Q_Z)_r$ for Q_Z close to zero (the index 'r' indicates rectangular coordinates, see Fig 1). They found satisfactory fits to eq.(2) over the whole temperature range. The Q_Z dependence, however, was not tested.

Experiments and Results. We looked at room temperature diffraction data at different points on the $(1,0)_r$ rod for perpendicular momentum transfer Q_Z=0.1, 0.4, 0.7. Parallel momentum scans were taken along the $[010]_r$ azimuth. Power-Law fits were only satisfactorily for Q_Z= 0.1, similar to Held *et. al.* [3]. At larger Q_Z, an obvious asymmetry develops that made the use of a two component lineshape necessary (see fig. 2). The χ^2 for the two component fits were smaller by at least a factor of 4.5.

Since both components increase in amplitude as Q_Z approaches the Bragg condition, local steps on a long range flat surface can be excluded. This is because steps give rise to a broad component that has a minimum at the Bragg point, where

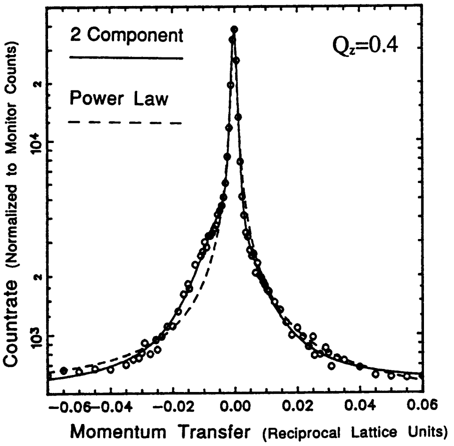

Fig. 2. Room temperature scans of the (10) beam in the $[010]_r$ direction at different Q_Z. The solid lines are the two component fits. Dashed lines are power law fits.

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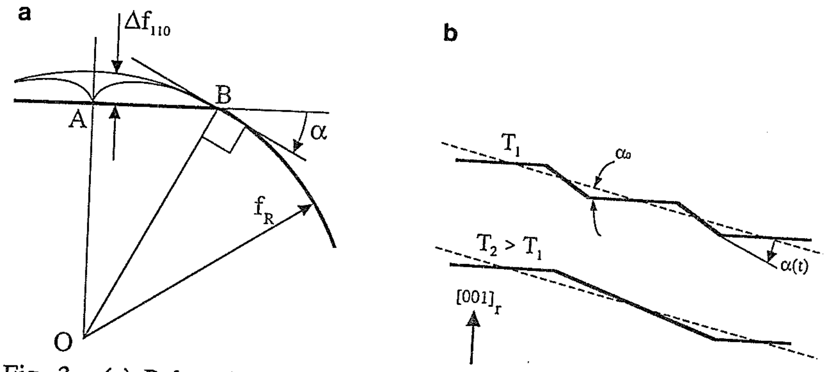

Fig. 3. (a) Polar plot of the free energy curve near the [110] direction. At temperatures above T_R the free energy surface is spherical. At lower temperatures a cusp develops in the [110] direction leading to the flat (110) phase. The thick line is the ECS. Orientations between A and B are forbidden. (b) Side view of the faceted surface at two temperatures. The rough inclined phase makes an angle α with the (110) plane that depends on the depth of the [110] cusp in the free energy curve.

all atoms scatter in phase [9]. The increase in intensity is instead consistent with two CTRs that have maxima at the Bragg point.

The theory of ECS can be used to understand this data [10,11]. As long as isolated steps can exist on the surface all crystal directions near (110) are present. If as in the case of these experiments on Ag(110), isolated steps congregate away from the facets, most crystal orientations close to (110) are forbidden. To see this let us assume that the free energy surface close to the melting point is spherical, and that a cusp develops in the [110] direction below the (110) roughening temperature (see fig. 3a) [10,12]. The ECS of such a system is the internal envelope of planes drawn perpendicular to the radius vectors at the intersection with the free energy plot [10]. Therefore, the cusp will lead to a flat region of (110) oriented surface. Note also, that all orientations in fig. 3a between A and B are forbidden since they do not contribute to the internal envelope. In this sense only points A and B are well defined. Any miscut leading to forbidden orientations must then phase separate into either the A (flat (110)) or the B (rough) phase ("hill-and-valley" structure), according to a lever rule [10,13]. Figure 3b shows a side view of the surface. In the diffraction profiles the sharp component arises from the flat (110) region while the broad component is due to an inclined region (see fig. 4). In this scenario the surface roughens by replacing the flat (110) terraces by the stepped facet. Note the symmetry at the in-plane position in fig. 4. This explains the symmetric lineshapes at Q_z=0.1. Further up the rod, the lineshape must be asymmetric and the intensity of the broad component increases, consistent with our data.

The angle between the flat (110) phase and the rough phase (tilt angle) can be evaluated from the separation of the two peak centers. To do this we took scans at $(1,0,0.5)_r$ as a function of $q_{||}$ in the $[010]_r$ direction, and fitted the data to a two component lineshape. The sharp component was fit by the square of a Lorentzian representing the convolution of the instrument function with the sample mosaic.

Fig. 4

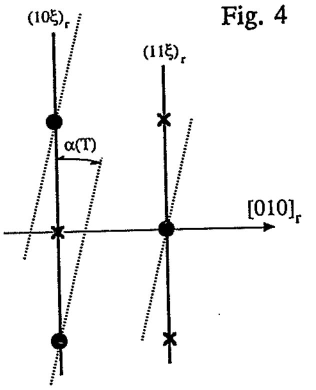

Fig. 5

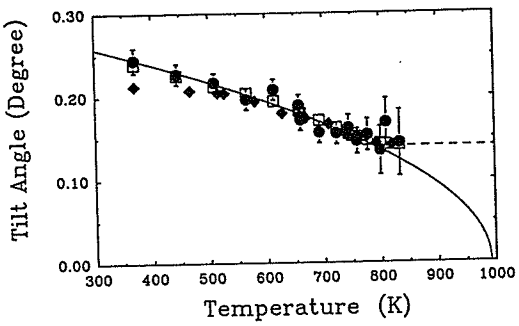

Fig. 4. Reciprocal space map showing the CTR's due to the two phases. The inclined dashed line corresponds to the stepped (rough) phase. The solid line is the CTR from the flat (110) phase. Circles and 'X' represent Bragg and Anti-Bragg points, respectively.

Fig. 5. Tilt angle α vs. temperature derived from the peak separation (filled circles) and by using the integrated intensities of the (1,0) beam (open squares) and (2,0) beam (filled diamonds). The solid line is a plot of eq. 3 with T_R=992 K.

The broad component is a single Lorentzian. Figure 5 shows a plot of the tilt angle versus temperature. At about 790K the tilt angle becomes a constant (α_o=0.14°), which agrees with the miscut of our sample determined by an optical measurement of the surface orientation.

From the geometry of fig. 3a it follows that for small α we have

$$\alpha(T) = \sqrt{\frac{2\,\Delta f_{110}(T)}{f_R}}. \tag{3}$$

If we assume a linear reduction of Δf110 (the depth of the cusp) with increasing temperature of the form,

$$\Delta f_{110} = |T_R - T|, \tag{4}$$

then the temperature dependence of α can be obtained after suitable adjustment of T_R and f_R (solid line in fig. 5). The roughening temperature of the (110) surface is found by extrapolating to the temperature where α(T)=0. If we have a surface that has been miscut by an angle α_o and Δf110 is such that α_o is an allowed orientation, then only the B phase will exist. At these temperatures all crystal directions close the miscut are possible; isolated steps can persist and the surface is rough.

The relative amount of the two phases is also reflected in the integrated intensities of the two CTRs. Because the scattered intensity goes as the square of the number of scatterers, it is proportional to the surface area. By conserving the miscut of the sample, we have a second independent measure of the tilt angle

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$$\alpha(T) = \alpha_0 \frac{(I_A+I_B)}{I_B}\ . \qquad (5)$$

These angles agree well with those derived from the peak separation, see fig. 5. The roughening transition is completed when the rough phase totally replaces the (110) flat phase. This temperature depends on the miscut of the sample.

Conclusions. The presented diffraction data can be satisfactorily fit to a two component lineshape. The results are interpreted as being due to two separate phases on the surface. A simple model has been developed that relates the tilt angle between the two phases with the roughening temperature T_R and the free energy f_R. The tilt angles calculated from the integrated intensities and the peak separations, respectively, were found to be in agreement with this model.

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