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"Patterning of surfaces during homoepitaxial growth and erosion"

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Steering-enhanced roughening during metal deposition at grazing incidence

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It is shown that steering may have an important influence on the morphology of growing films. Steering originates from long-range attractive forces between incoming atoms and substrate atoms and leads to preferential arrival of atoms on top of islands. This phenomenon is most pronounced for grazing incidence deposition and results in significantly increased roughness of the growing film. Steering, which is expected to be generally valid but has so far been disregarded in growth studies, is illustrated for the growth of Cu/Cu(001).

PACS numbers: 68.55.JK, 81.10.-h, 61.14.Hg, 68.35.Bs

The evolution of non-equilibrium surfaces is a complex phenomenon, even for homoepitaxial growth systems. The emerging growth front is the result of roughening, due to the incident flux, which is counteracted by multiple transport processes tending to smoothen the surface. An important parameter in the balance between roughening and smoothing is the Ehrlich-Schwoebel (ES) barrier, which hinders downward diffusion across step edges [1,2]. A finite ES-barrier results in a roughening of the growing surfaces [3–6], i.e., to the development of mounds.

Up to now, growth models have always regarded the flux of impinging atoms as being homogeneously distributed over the surface. Possible consequences of incident flux inhomogeneity as a result of surface roughness have been neglected completely. This shortcoming is remarkable since molecular dynamics calculations by Sanders et al. [7] show substantial deflection of non-normal deposited atoms towards the surface, starting at about one nm above the outermost layer. This phenomenon can simply be rationalised since the attractive well, as seen by the approaching metal atoms, is of the order of a few eV's, while their kinetic energy typically amounts to only a few tenths of an eV. Consequently, the atoms must undergo strong acceleration towards the surface. In this Letter we report an effect which we believe is the first evidence for the importance of *steering* for the evolving morphology of the growth front. It gives rise to a remarkable heterogeneity of the incident flux, i.e., redistribution of the incident atoms due to their morphology dependent trajectories. Because of steering, atoms arrive preferentially on protruding terraces resulting in an enhanced roughening of the growing surface.

The experiments reported here have been conducted in ultra-high vacuum (base pressure $<10^{-10}$ mbar) using spot profile analysis low energy electron diffraction (SPA-LEED). During growth, the temperature of the desulpherised Cu(001) substrate has been kept at 250 K, while during measurements the temperature has been maintained at 100 K. The deposited copper has been sublimated from an also desulpherised Cu-disk, heated from the rear by means of electron bombardment. For the data discussed here the copper atoms have been deposited at an grazing angle of incidence (80° from the normal) along the [110]-azimuth. The homoepitaxial films have been grown at a rate of about 0.25 monolayers per minute. This deposition rate is about equal to that used in normal incidence experiments reported earlier [8], to which we will frequently refer here.

The central piece of data in this Letter is shown in Fig. 1: a profile of the specular beam obtained after deposition of 40 ML of copper on Cu(001) at grazing incidence. The fourfold symmetry, expected for the Cu(001) surface, and indeed measured after normal incidence deposition at similar conditions [8–11], is obliterated. Besides the dominant central Bragg peak two well developed facet peaks have emerged. Both peaks are positioned in the plane of incidence of the copper beam, while no clear diffraction features can be detected out of this plane. The observed diffraction pattern can be interpreted straightforwardly as resulting from growth induced parallel ripples at the Cu(001) surface, which are oriented perpendicular to the plane of incidence of the copper beam. The absence of out of plane diffraction features and the small out of plane width of the diffraction peaks suggest that these ripples are quite well defined and have an average length of about 50 nm. The two clear in-plane facet peaks correspond to well established $\{1\bar{1}1\}$- and $\{1\bar{1}3\}$-facets on the illuminated and shadow sides of the ripples, respectively.

Compared to the facets which emerge after normal incidence deposition of 40 monolayers, the facets evolving at grazing incidence deposition are much better defined, i.e., their slope distribution is much narrower. In addition, the slopes of the illuminated and the shadow sides, corresponding to $\{1\bar{1}1\}$ and $\{1\bar{1}3\}$ respectively, are both substantially steeper than the average facet orientation of (115) [8,12,13] obtained after normal incidence deposition at 250 K. This illustrates the enhanced surface roughness after grazing incidence deposition. We attribute this enhanced roughness to the action of a basic phenomenon that until now has been discarded: redistribution of incident flux in favour of protruding surface areas due to a steering effect, which becomes substantial already at relatively large heights above the surface.

Steering is induced by long-range attractive forces between the substrate and the approaching slow copper atoms. Thermal copper atoms, approaching the surface at typical energies of 0.15 eV, experience a long ranged attractive well of several eV's. This gives rise to substantial acceleration towards the surface. For the flat substrate this phenomenon has initially no consequences: the incident flux remains homogeneously distributed. The copper atoms only arrive at an effectively smaller polar angle of incidence. However, as soon as aggregates start to build up, the redistribution of material becomes progressively more important. To substantiate this phenomenon for a macroscopic surface we have performed a number of trajectory calculations. Being primarily interested in a qualitative (or semi-quantitative) description, we have adopted a Lennard-Jones (12,6) potential. Its parameters have been chosen such that both the copper interatomic distance as well as its cohesive energy are described quantitatively [14]. Trajectory calculations show that 150 meV Cu-atoms, directed with an grazing angle of incidence of 80° towards the Cu(001) surface-plane do actually hit it at an angle of 17°. Clear deviations from the unaffected trajectories already occur at distances up to one nm above the outermost layer.

Figure 2(a) shows a cross-sectional view through the Cu(001) along the [110] azimuth. On this substrate we have constructed a monolayer high step in the direction perpendicular to the plane of incidence. This figure exhibits calculated equipotential energy contours (increment -0.1 eV) as well as two calculated atom trajectories for atoms deposited with a grazing angle of 80°. The equipotential energy contours show a pronounced step in the *attractive* potential, which extends laterally e.g. for the -0.1 eV contour by as much as about 15 Å. Trajectory B illustrates that, due to acceleration towards the surface, the incident atoms actually hit the surface a quite long distance (δ_B) before their target point. The target point is the intersection of the asymptotic long distance part of the trajectory with the surface. Far away from the step in the attractive potential δ is constant and irrespective of the level at which the atoms arrive. Consequently, a homogeneous flux is expected. Consider now the situation as illustrated by trajectory A: When the atom passes the island boundary it suddenly experiences the surface at a much smaller distance and impact is closer. Consequently, the value of δ_A is reduced giving rise to a local enhancement of the incident atom flux just behind the front edge of the island as illustrated in Fig. 2(b). In fact all arriving atoms whose trajectories pass through areas of substantial distortion of the attractive potential related to the ascending step contribute to enhanced flux. They are focused onto the near step region. We refer to this phenomenon as **steering**. For the one monolayer high island (see Fig. 2(b)) the enhancement factor amounts to about 1.6 at the front of the island and decreases to unity going further downstream on an extended island. Behind the island the density of the incident atoms is reduced, which is related to the shadowing by the island. The reduction of flux behind the island compensates the flux enhancement *on top* of the island as should be the case for particle conservation reasons. Note that the flux behind the island never becomes zero, which would be the case for classical shadowing without steering. For comparison, the classical situation is also drawn in Fig. 2(b). In this case no flux enhancement *on top* of the island is observed: the flux missing in the shadow zone is concentrated *in front* of the illuminated side of the island. So we clearly obtain a redistribution of incident flux, leading to steering of the atoms towards the top of the growing island. The effect being substantial already for monolayer high adatom islands strongly gains importance when the surface becomes rougher. Calculations of the normalised incident atom flux for a three monolayer high adstructure show enhancement factors well exceeding three! This phenomenon obviously leads to rougher growth fronts [15], which is enhanced by the presence of a finite ES-barrier. We note in addition that the range of the shadow region is much smaller for the classical shadowing case than in the presence of steering. This phenomenon contributes to improved lateral order. Indeed we observe a narrow separation distribution of the ripples in the plane of incidence [16].

Already in the sub-monolayer regime the growth behaviour is affected by the deposition geometry. This statement is illustrated nicely in Fig. 3 showing the specular beam profile obtained after grazing incidence deposition of about 0.5 ML. The narrow Bragg peak is surrounded by a ring, the position of which is given by the distance between the adatom islands. This quasi diffuse scattering ring has perfect rotational symmetry after normal incidence deposition [8]. In the present case of grazing incidence deposition, however, the ring intensity exhibits a clearly developed twofold symmetry. It possesses two pronounced minima, in the direction perpendicular to the plane of incidence. In fact, the development of a twofold symmetric rings is already visible after deposition of 0.3 monolayers copper. This remarkable beam profile reveals the presence of homogeneously distributed rectangular islands in contrast to the square ones growing at normal incidence. The step edges remain oriented along the close packed <110>-azimuths. Approaching the energetic preference for <110>-ledges is facilitated by sufficient ledge adatom diffusion at the considered substrate temperatures [17]. A comparison with calculations (kinetic approximation) indicates that the rectangular islands have an aspect ratio of about 1.05. They would have been hardly noticed with STM. The applied diffraction method is well able to distinguish this feature. The long sides of the islands are oriented perpendicular to the plane of incidence of the deposited Cu-atoms.

The development of rectangular islands at submonolayer coverages turns out to provide a welcome and independent check of the consistency of the model conclusions derived above. From the fact that homogeneously distributed square islands develop during normal deposition we learn that the diffusion of copper on Cu(001) is isotropic and ledge adatom diffusion is readily active at 250 K. This implies that the evolution of rectangular islands has to be related to peculiarities connected with the grazing incidence of the copper atoms. In particular, the total step edge advance rate perpendicular to the plane of incidence has to be slightly higher than the sum of the advance rates of the illuminated and the shadow edge of the adatom islands. Let us consider first the situation without steering. If the arriving atoms are instantaneously equilibrated the step edge advance rate on the shadow side is strongly reduced since its capture zone receives less atoms than in the case of normal incidence. On the basis of particle conservation this feature is exactly compensated by the larger impingement rate in the capture zone of the illuminated step edge. Consequently, the sums of the in-plane and of the out-of-plane step edge advance rates are exactly equal. Since in the considered case (there is no attractive potential and atoms stick where they hit the surface) there is no reason for an anisotropic descent of atoms from the top of the adatom island, square islands must develop. If, however, we switch on steering , the anisotropic shapes can be explained. Steering will lead to a redistribution of incident flux to the top of the adatom islands: flux that without steering would contribute to an upstream advance of the illuminated step edge, now lands on top of the island and is partly descending across the [110] oriented ledges. This gives rise to the evolution of rectangular islands (aspect ratio $\approx$1.05), which are elongated perpendicular to the plane of incidence. Estimates of the magnitude of this feature lead to values well consistent with the experimental data.

We finally note that steering should be anticipated in any growth situation. Its consequences may vary from insignificant for monolayer high islands at normal deposition to substantial at grazing incidence. Steering is expected to be stronger for metals than for semiconductors or isolators. In general, it will be strong when substrate and incident atom have relatively high electronic polarizabilities. The well established small but finite ES-barrier for Cu/Cu(001) [8,10,11] leads to significant effects. The consequences should be even more substantial for higher values of the ES-barrier, which are commonly related to close-packed (e.g. fcc(111)) metal surfaces.

Summarising, the presented grazing incidence growth experiments of Cu/Cu(001) provide unambiguous evidence for the importance of steering for the morphology of the growth front. Steering leads to a substantial redistribution of incident flux and in fact to focusing of the incident particles onto the uppermost levels of adatom aggregates. This so far unanticipated phenomenon in epitaxial growth leads to enhanced roughness of the growth front, in particular when the incident particles are deposited at grazing incidence. When comparing experiments mutually or to calculations the exact deposition geometry must be considered. In particular during the emergence of growth induced facets the above discussed steering effect may become of prime importance (Ref. [8], and references therein). Grazing incidence deposition of copper on Cu(001) leads to the emergence of highly ordered arrays of parallel asymmetric ripples oriented normal to the plane of incidence. This novel observation may well be used to structure substrates, which may subsequently act as templates for heteroepitaxial structures.

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FIG. 1. SPA-LEED spot profile of the specular spot acquired after deposition of 40 ML Cu at an angle of 80 from the surface normal with the substrate at 250 K.

FIG. 2. (a) Calculated equipotential energy contours and two atom trajectories for a surface with an step edge in the direction perpendicular to the plane of incidence (note the different length scale on the two axis). The increase in attractive potential is -0.1 eV for the solid contour lines. The trajectory calculations for a deposition angle of 80 (with respect to the surface normal) start at 20 Å above the surface. (b) Calculated adatom flux at the surface, normalised to a homogeneous atom flux far above the surface. The dashed line shows schematically the adatom flux in case of classical shadowing without steering.

FIG. 3. Profile of the specular SPA-LEED peak acquired after deposition of 0.5 ML Cu at 80 with the substrate at 250 K.

Growth Anisotropy and Pattern Formation in Metal Epitaxy

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Evidence for the formation of growth induced, ordered checkerboardlike arrangements of mesas has been obtained. These patterns develop on a metal substrate with square symmetry after deposition of tens of monolayers. Its origin is traced back to laterally anisotropic advance rates of island edges in combination with slope selection. The foundation for the mesa arrangement is already laid just after coalescence of the adatom islands in the *first* monolayer. The results are exemplified in a high resolution surface diffraction study for the growth of Cu on Cu(001). [S0031-9007(97)02309-0]

PACS numbers: 68.55.Jk, 61.14.Hg, 68.35.Bs, 81.10.–h

The formation of ordered structures on surfaces during epitaxial growth on surfaces has recently obtained much attention. One prominent result of studies in this field is that in a nonequilibrium system diffusion controlled growth may lead to well defined dendritic patterns [1,2]. Particularly nice examples of this phenomenon have recently been reported by both Hohage *et al.* [3] and Brune *et al.* [4] for metal islands on metal fcc (111) substrates. Like fractals, these islands have a branched, rugged structure. In contrast to fractals, the branches develop along preferred directions, i.e., along ⟨110⟩ directions on the (111)-oriented substrates, leading eventually to triangular envelopes of the ramified islands. The formation of dendrites rather than fractals, i.e., of *orientationally ordered* instead of random island structures, has been traced back to strong *diffusion anisotropies* at the extremities of the branches. This applies, in particular, to an asymmetric diffusion rate of atoms around island corners [3,4]. In this Letter we present evidence for the fact that diffusion anisotropy not only leads to ordering in the *two-dimensional* regime, but that this interrelationship applies more generally. We show for the growth of tens of monolayers (ML) of Cu on Cu(001) that also ordering in *three dimensions* occurs due to growth anisotropies. In particular, our high-resolution low energy electron diffraction data reveal the formation of a checkerboard pattern existing of Cu mesas with quite well defined slopes. This self-organization occurs in a broad temperature regime in which both the average distance between the mesas and their preferred slopes depend distinctly on the substrate temperature. As a remarkable and important phenomenon we emphasize that the base structure develops already immediately after coalescence in the first monolayer at a deposit of approximately 0.7 ML (monolayer). Our results are believed to apply in general for square substrate orientations.

The experiments have been performed in a home-built He atom diffraction (TEAS) apparatus equipped with an Omicron high-resolution SPALEED (spot profile analysis low energy electron diffraction) instrument, an ion gun, a CMA-Auger system, and several deposition facilities. The transfer widths of the SPALEED—and the TEAS device amount to about 1500 and 500 Å, respectively. The *ex situ* desulfurized Cu crystal has been further prepared in UHV by numerous cycles of sputtering with 800 eV Ar^+ ions and prolonged heating at about 1000 K. This procedure allows standard mean terrace widths well exceeding 1000 Å. Copper has been sublimated from a thoroughly desulfurized disk, heated from the rear by means of electron bombardment. The growth experiments described in this Letter have been performed mostly at a substrate temperature of ~250 K and with a deposition rate of about 0.01 ML/s. Immediately after deposition the temperature of the sample has been quenched rapidly in order to suppress undesired diffusion. The SPALEED data have been acquired with the substrate held at 100 K. The profiles of the specular peak (higher order peaks behave similarly) have been taken near destructive interference conditions.

Figure 1 displays a representative example of the profile of the specular SPALEED spot acquired after deposition of tens of monolayers of copper. In this case the deposit amounts to about 20 ML. The profile shows an extremely rich structure. The intensity at the Bragg position exhibits a pronounced minimum. This indicates that many layers are exposed to the probing electron beam: the growth proceeds in a multilayer fashion revealing restricted interlayer diffusion (see below, Fig. 3). The spot profile exhibits four well developed maxima (lobes), peaked roughly in the ⟨100⟩ azimuthal directions. The general aspect of the image is reminiscent of some earlier published data [5,6], which were obtained for the same growth system. The obtained spot profile thus shows a well developed fourfold symmetry as should be expected for the Cu(001) surface, which is illustrated best by the contour plot in the inset. For low intensities the contour assumes a fourfold symmetric shape. As is well known from textbooks on diffraction, they are indicative of a

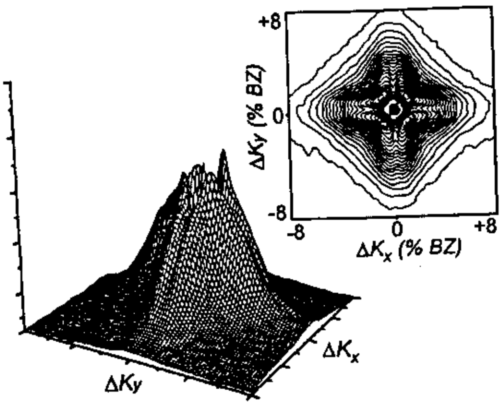

FIG. 1. Profile of the specular LEED spot after deposition of 20 ML Cu at a rate of ~0.01 ML/s with the substrate at 246 K. Inset: contour plot of the same data.

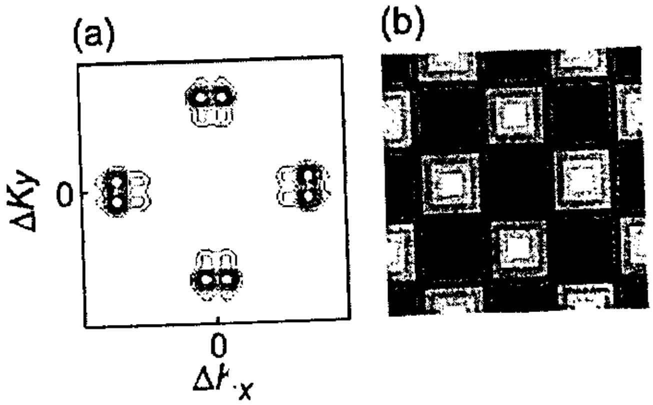

FIG. 2. Calculated specular spot profile (a) for diffraction from a checkerboardlike arrangement of regular pyramids (b). Grey scale refers to various terraces.

square shape of the adatom islands (see Fig. 4 and discussion). The ledges of the adatom island structures are preferentially oriented along the close packed ⟨110⟩ directions. The pronounced four-lobed structure demonstrates the presence of strong order along the ⟨100⟩ directions, i.e., along the diagonals of the surface projected adatom islands. A particularly interesting feature is the appearance of a previously unnoticed feature in the diffraction spot profile: the evolution of couples of well developed parallel ridges along the ⟨110⟩ directions. These ridges are quite clear in the contour plot. They appear as maxima at distances which do not depend on the parallel components of the wave vector change, Δk_x and Δk_y. These well developed ridges occur in a broad temperature range, roughly varying from 100 to 300 K and in a wide coverage regime (~15–40 ML). Their separation becomes increasingly smaller with increasing substrate temperature. These well developed ridges represent the key evidence for the formation of a checkerboardlike arrangement of copper mounds. The separation between the ridges relates reciprocally to a distance scale between the centers of the mounds. We think of a mound as being a mesa, i.e., a flattened pyramid the slope of which becomes gradually better defined with increasing deposition. In order to better understand these features we made a few qualitative calculations.

Figure 2(b) shows a cartoon of idealized pyramids arranged on a perfect checkerboard. Their base plates have a square shape. Figure 2(a) shows a contour plot of the calculated coherent diffraction pattern, under destructive interference conditions, from this arrangement of pyramids. One obtains four couples of closely separated spots; their positions are determined by the ideal superlattice on which the pyramids are based. Thus the distance between the spots is reciprocally related to the distance between the pyramid centers. The number of diffraction spots with appreciable intensity will be determined by the shape of the terraces. For ideal pyramids this implies that per facet orientation, roughly speaking, only two spots will carry measurable intensity. In order to mimic the reality on the surface more closely one has to adopt distributions with finite widths both for the slope and the lateral separation. For a finite distribution of slopes of the facets one should therefore expect the peaks to lie on two parallel lines. So from a comparison of the calculated result and the experimental peak profile one immediately concludes that the slope of the developing pyramids has a rather broad distribution as one should expect for developing mesas. In addition, the checkerboard pattern will not be as ideal as in the cartoon. The introduction of some lateral disorder leads to a broadening of the diffraction spots. The overall result of introducing statistics leads eventually to the parallel ridges appearing in the experimental diffraction spot profile.

Upon further deposition (around 80–100 ML) we also observe facet peaks in the diffraction pattern (not shown in the framework of this Letter). In close agreement with Refs. [7] and [8] we find (113), (115), and (117) facets depending on the substrate temperature: below about 180 K, between about 180 and 280 K, and between about 280 and 300 K, respectively. At and beyond this point the stripes in the spot profiles start to disappear, beginning at large $\Delta k_{\parallel}$ values (i.e., at small length scales).

Our data appear to be also consistent with earlier theoretical results obtained by Plischke and Siegert [9,10]. These authors have, triggered by the growth instabilities for Cu/Cu(001) observed by Ernst *et al.* [7,8], proposed a recipe, which leads to slope selection, i.e., to the growth of pyramidlike structures, under typical conditions of molecular beam epitaxy. Important ingredients in this description are two postulations artificially introducing both lateral growth anisotropy and slope selection. The first gives rise to the growth of square islands, while the latter takes account for the evolution of slopes. Simulations led to the formation of pyramids arranged on a checkerboardlike pattern [9,10]. Our present experimental results are to a high degree consistent with their results. A consequence of this scenario is an increase of the separation between the pyramids at large depositions. Indeed, Stroscio *et al.* [11] have shown for the system Fe/Fe(001) that at large depositions the characteristic separation between the moundlike structures increases. This is a well accepted feature of a growth instability: Finally only one huge pyramid would be observed. Thus ultimately after full development of the facets the smaller mounds will be overgrown and the checkerboard pattern will gradually disappear. Such behavior is consistent with our observations as mentioned above. Below we address the issue of which mechanism is the key contributor to the formation of the checkerboard pattern. It turns out that the basic ingredients are already present just after coalescence of the submonolayer adatom islands. For this purpose we will closely inspect the initial growth behavior.

Figure 3 shows the height of the specular He peak, measured under destructive interference conditions, during the deposition of copper. Initially the He-peak height oscillates revealing quasi layer-by-layer growth [12,13]. This result agrees with data obtained previously by various authors [14–18]. The oscillations are strongly damped, indicative of restricted interlayer mass transport and finally even disappear revealing a crossover to a quasimultilayer growth regime. The latter growth mode implies that nucleation in the next layer takes place before coalescence occurs [12,19,20]. This behavior requires that interlayer diffusion is hampered by the existence of an Ehrlich-Schwoebel barrier hindering the descent of adatoms onto a lower terrace. We will concentrate next on the development of the surface morphology as a result of submonolayer deposition where the growth essentially proceeds in a two dimensional fashion (cf. Fig. 3).

Figure 4(a) shows a contour plot of the specular peak obtained after deposition of 0.5 monolayer copper. The measured peak profile constitutes a nice example of an electron diffraction analog of optical Fraunhofer diffraction from square apertures [21]. The structureless sharp ring around the specular peak indicates that the separation between adatom islands has a narrow distribution and reveals the absence of azimuthal preference for finding neighboring islands. The fourfold structure outside the ring reflects the size distribution and the dominant orientation of the square islands. Global inspection of the contour plot immediately leads to the conclusion that the steps of the square adatom islands are preferentially oriented along ⟨110⟩ directions. The formation of these preferentially square shaped islands, even at relatively low substrate temperatures, indicates that the barrier for diffusion of adatoms along the ⟨110⟩-oriented ledges is low. Note that the island size distribution does not affect the ring shape in this peak profile. The existence of square adatom and vacancy islands on Cu(001) has recently been observed also with STM [22,23].

We consider now the peak profile obtained after deposition of 0.7 monolayers copper shown in Fig. 4(b). This image is representative for the situation just after coalescence of the adatom islands in the first layer. The peak profile now shows a pronounced fourfold symmetry, not only in the fourfold symmetric structure outside the ring, but also in the ring itself: The ring has maxima in the ⟨100⟩ directions. While the step edges are still oriented in the ⟨110⟩ direction, the initially isotropic distribution of island separations has changed into a distribution which is peaked in the ⟨100⟩ directions. Thus the preference for ordering along ⟨100⟩, which prevails at much higher coverages (Fig. 1) and is an important factor in the development of the checkerboard pattern, is already present directly after coalescence of the islands in the *first* monolayer. The reason for this phenomenon is easy to explain. Obviously, the reordering which could take place as a result of coalescence *during deposition conditions* is difficult to describe. Very likely the anisotropic island edge advance rates play a decisive role: The equilibrium shaped square islands grow in the ⟨100⟩ direction $\sqrt{2}$ times faster than in the ⟨110⟩ direction. Since the initial separation between islands is isotropic, coalescence of the adatom islands takes place preferentially along their diagonals, i.e., along ⟨100⟩ azimuths. The details of the subsequent course of events have to be analyzed further. A possible scenario might be that the remaining vacancy

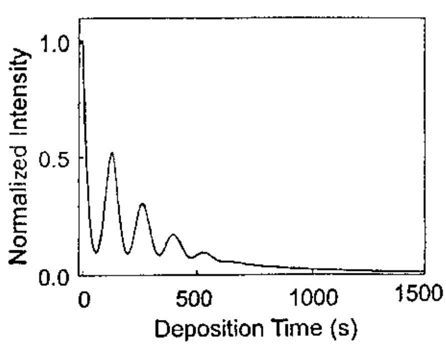

FIG. 3. He-specular peak height during deposition of Cu with the substrate at 250 K.

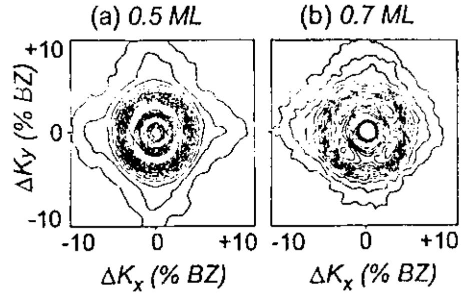

FIG. 4. Contour plots of specular LEED spot after deposition of 0.5 (a) and 0.7 ML (b) 250 K.

clusters (i.e., yet uncovered areas of the original substrate) will tend to assume a square equilibrium shape, reducing the order along ⟨110⟩ directions. The preference for correlations of adatom structures along the diagonal (⟨100⟩) directions is even reinforced when nucleation in the second layer takes place. This has to take place in the coverage interval between the onset of coalescence and deposition of a monolayer equivalent. The precise moment depends on the height of the Ehrlich/Schwoebel barrier, the substrate temperature, and the deposition rate.

As a consequence of the above outlined scenario for the evolution of the growth front the separation of the ridges (see Fig. 1) must be intimately related to the length scale already set during the initial nucleation stage on the freshly prepared surface. Figure 5 shows an independent check of this feature: It shows a plot of the log of the ridge separation and of the $\Delta k_{\parallel}$ values corresponding to the maximum of the ring structure [Fig. 4(a)] versus the reciprocal substrate temperature. Indeed the slopes are identical and, moreover, agree nicely with those found previously [5,6,7,16]. A closer inspection shows that the quantitative length scales also agree nicely.

In summary, we have shown that *isotropic* surface diffusion of single adatoms, in combination with the formation of equilibrium shaped (square) adatom islands leads to laterally anisotropic growth rates. Combined with a finite Ehrlich-Schwoebel barrier this gives rise to the formation of checkerboard patterns of Cu mesas on Cu(001). The base for this ordering phenomenon in tens of monolayers thick deposits is already laid just after coalescence of the growing adatom islands in the first monolayer. The observed features are believed to be a general property of unreconstructed (001) surfaces whenever the three conditions mentioned above are fulfilled.

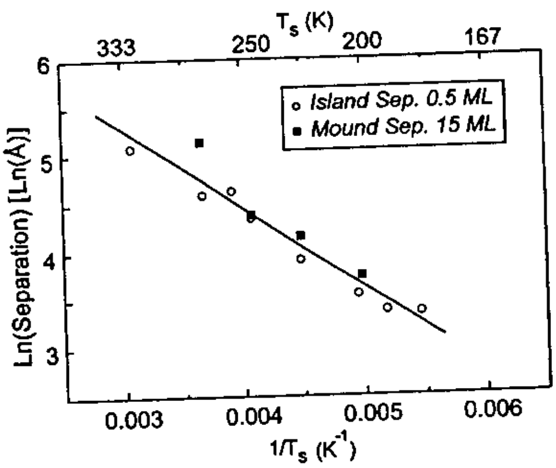

FIG. 5. FWHM of the ring structure (○) and the separation of the stripes (■) (see text) after deposition of 0.5 and 15 ML, respectively, versus the reciprocal substrate temperature.

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