Observation of a Growth Instability during Low Temperature Molecular Beam Epitaxy

H.-J. Ernst, F. Fabre, R. Folkerts, and J. Lapujoulade
Commissariat à l'Energie Atomique Saclay, DRECAM/SRSIM, 91191 Gif-sur-Yvette, France

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The growth of a Cu(100) crystal has been investigated with helium atom beam scattering in real time and examined in the light of the dynamical scaling hypothesis. The associated exponents have been determined. The analysis of both terrace and step correlations during growth provides a detailed scenario of how a singular surface can transform into an arrangement of vicinal surfaces upon unstable growth, resulting in a pyramidlike surface profile. The sides of these pyramids are composed of the (113) and (115) Cu surface for deposition at 160 and 200 K, respectively.

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The growth of a crystal via molecular beam epitaxy (MBE) is often not perfect. While in heteroepitaxial systems geometric or chemical mismatches may influence the growth process, deviations from perfect growth in homoepitaxial systems are exclusively determined by kinetic parameters. The stochastic nature of the impinging flux, and possibly the nucleation process itself, induce a roughness, which is generally counterbalanced by a smoothing or ordering process due to mobility of deposited atoms. At low deposition temperatures, the competition between fluctuation and coarsening can reach an equilibrium on a short range scale only, but globally the roughness continues to develop. A detailed structural characterization during the evolution of the interface is of considerable practical interest due to the application of thin films in a wide variety of areas in science and technology.

Much theoretical effort has also been devoted to growth processes recently, because they are thought to be an example of a dynamical mechanism that drives a system into a spatially and temporally scale invariant state [1,2]. Within this scaling approach, the evolving growth front is proposed to have a self-affine form. Two characteristic lengths can be defined: The width of the interface w (the rms fluctuation about the mean height), and a correlation length ξ parallel to the surface, which is associated with the horizontal cutoff to scaling. ξ and w are assumed to increase as simple power laws in time, $w \propto t^{\beta}$ and $\xi \propto t^{\beta/\alpha}$. The exponents α and β depend on specific growth models [1,2]. Thus, their experimental determination and comparison with theory can be valuable in identifying the dominating underlying kinetic processes.

Of course, a wealth of studies have been performed on crystal growth, but it is only very recently that experiments have been analyzed in the light of the dynamical scaling hypothesis [3]. While scaling has been observed, no clear identification of the experimentally determined exponents with available theories seems to be established. One may suspect that the use of heteroepitaxial deposition systems or buffer layers requires additional elements in growth models that go beyond existing theories. In this paper we analyze the homoepitaxial growth on a nonreconstructed surface of a single crystal. Thus, this experiment is as close as possible to the conditions applied in existing simulation work.

The use of helium atom beam scattering as a probe provides a means to monitor the deposition process in "real time," i.e., during the evolution of the rough growth front. Since this technique can distinguish between scattering from terraces and step edges, it offers not only the possibility to study height correlations of terraces, but also to obtain complementary information from step correlations [4]. From the investigation of terrace scattering, we determined the growth exponents, $\alpha \approx 1$ for deposition at 200 and 160 K, while β amounts to $\approx \frac{1}{2}$ at 200 K and $\approx \frac{1}{4}$ at 160 K. The analysis of step correlation reveals that, starting from the singular (001) surface, the interface evolves into a state that is characterized by "pyramidlike" structures, whose sides correspond to the Cu(115) vicinal surface at 200 K and the (113) vicinal surface at 160 K. This instability has been predicted by a new class of growth models [5,6], which take into account an asymmetry in the vertical and horizontal mobility, the Schwöbel effect [7].

The experiments were performed with the Ramses II helium atom beam spectrometer at Saclay. The Cu crystal has an average residual terrace width of about 700 Å. Cu was evaporated from a radiatively heated Cu disk (12 mm diam), located about 10 cm in front of the sample. The incident flux was 120 sec/monolayer (ML) [8]. The evolution of the growth front was monitored by taking diffraction patterns during deposition. In these scans, the angle of the impinging Cu flux with respect to the crystal varied continuously between about +30° and −30°. In some experiments Cu was deposited at a fixed angle close to the surface normal. No difference in the diffraction pattern was observed for these evaporation modes.

The central hypothesis of a dynamical scaling in space and time asserts that $H(r,t) = \langle [h(r,t) - h(0,t)]^2 \rangle$, the height difference correlation function, has the form [1,2,6]

$$H(r,t) = 2w^2(t) g(r/\xi(t)) \tag{1}$$

with $g(x) = x^{2\alpha}$ for $x \ll 1$ and $g(x) = 1$ for $x \gg 1$. Here $h(r,t)$ denotes the height of the interface above position r parallel to the substrate at time t. The scattering from a

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multilevel interface can be separated into two parts [9]. The intensity shows a sharp component (which in the following we call "the delta part," because its half-width is only determined by the instrumental resolution), superimposed on a broader, near specular contribution ("the diffuse part"). The delta part reflects the properties of the height difference correlation function at large distances, and contains therefore information on the exponent β, while the diffuse part reflects the nature of the interface on a local scale. Specifically, the shape of the diffuse part reflects the "power spectrum" of the roughness, and gives therefore information on the exponent α.

For the determination of the width of the interface as a function of coverage, the delta and diffuse scattering close to the in-phase condition must be separated with very good accuracy. Therefore, we proceeded as follows: A given amount of Cu was evaporated, and the crystal immediately quenched to about 150 K in order to freeze in mobility. Close to the in-phase condition, the delta intensity is expected to vary as $I(Q_z) \propto \exp[-w^2(t)Q_z^2]$ with the perpendicular momentum transfer $Q_z(\mathrm{mod}2\pi/c)$ [9,10]. The intensity of the delta part as a function of perpendicular momentum transfer could be reasonably well described by this Gaussian shape for coverages greater than about 3 ML. Figure 1 shows that the evolution of the width of the interface with time can be described by a power law behavior. The associated exponents are $\beta=0.26$ at 160 K and $\beta=0.56$ at 200 K. Figure 1 demonstrates as well that elevated temperatures do not necessarily lead to smaller interface widths; for a given coverage the width of the interface is smaller for deposition at 160 K as compared to deposition at 200 K.

The diffuse scattering close to the out of phase condition gives information of the height difference function on a local scale [9]. One of the assumptions expressed in the scaling ansatz is the time independence of the height difference correlation function at short range; see Eq. (1). In accord with our original finding [11], we observe the appearance of "sidebands" at very low coverage. With increasing deposition time, the sidebands broaden and merge into just one peak, which does not undergo any further changes for coverages above about 50 ML [12]. In principle, one could determine the exponent α from the line shape analysis of the diffuse scattering in this stationary state, but we were not able to determine α unambiguously; the line shapes at both temperatures could be fitted for α in a range from 0.6 to 1. An alternative method to deduce α is to analyze the FWHM of the diffuse scattering as a function of perpendicular momentum transfer [9], shown in Fig. 2. The functional form [9] of this variation strongly suggests that at both temperatures α is close to 1.

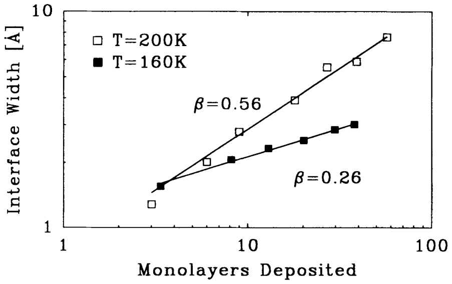

FIG. 1. Width of the interface w as a function of deposition time. Lines are fits to a power law.

Growth models have been classified as conservative, for which the dominant relaxation mechanisms are various types of diffusion processes, and nonconservative, in which the formation of voids and overhangs and desorption of deposited particles is allowed [1]. Recent work argues that the MBE process will always be described by nonconservative models in the *asymptotic limit* [2]. These theories predict typically $\alpha < 0.4$, and a scaling relation $\alpha + \alpha/\beta = 2$ is obeyed. The evolution of the growth front in the early time scaling regime is still a matter of ongoing discussions. While Kessler *et al.* [2] find that it is described by the linear version of the Kardar-Parisi-Zhang equation, Yan [2] finds better agreement with the linear diffusion equation [1]. The latter conclusion has been drawn also for the early time behavior of conservative models [1]. The linear diffusion equation produces the exponent found in this experiment at 160 K, $\alpha=1$, and $\beta=\frac{1}{4}$ [1].

Recently, a third class [5,6] of growth models has been introduced that describes instabilities which might occur in the course of the growth process due to an asymmetry in the vertical and horizontal mobility of adatoms due to an excess energy barrier to diffusion at step edges [7]. In

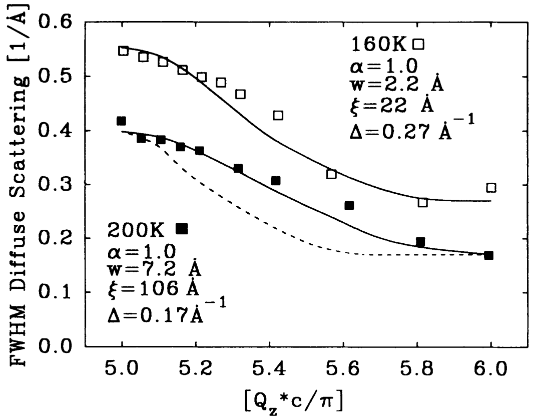

FIG. 2. Variation of the FWHM of diffuse scattering after deposition of about 60 ML as a function of perpendicular momentum transfer ($c=1.803$ Å). The in phase broadening Δ (not present in a kinematical diffraction theory) is due to finite resolution, inelastic processes, and scattering from isolated defects. We assumed that this independent contribution does not change with phase. The fits (solid lines) are for $\alpha=1$, include lattice effects [9], and are based on the phenomenological scaling function [9] $H(r,t)=2w^2(t)\{1-\exp[r/\xi(t)]^{2\alpha}\}$. The values for the width of the interface w were taken from Fig. 1. A smaller α would result in a steeper decrease close to antiphase, as demonstrated by the dashed line, which is for $\alpha=0.5$.

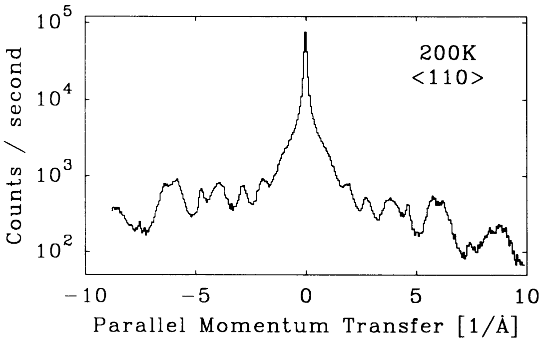

FIG. 3. Angular distribution after deposition of 60 ML Cu on Cu(100) at 200 K. The incident wave number is $k_i = 11.2$ Å^{-1}. The diffraction peaks at large parallel momentum transfer are associated with step edge scattering.

a seminal paper, Villain [5] pointed out that this Schwöbel barrier would lead to unstable growth, resulting in a pyramidlike surface profile, composed of terraces of uniform width. We believe that, for deposition at 200 K, we have encountered exactly this situation [13]. Clearly, $\alpha \approx 1$ would be consistent with a pyramidlike profile [14], and when interlayer mass transport is completely inhibited, β should be $\frac{1}{2}$ [15]. In the following, we provide further support.

Up to now all conclusions about structural correlations have been drawn from the near specular diffuse scattering, which is dominated by scattering from terraces. Helium scattering may provide complementary information from the investigation of far from specular scattering associated with the scattering from steps [4]. This is demonstrated in Fig. 3. In order to identify unambiguously possible correlations, we have performed measurements for different perpendicular momentum transfers. Surprisingly, as Fig. 4 demonstrates, the positions of the observed diffraction peaks associated with step edge scattering are found to show the characteristics expected for diffraction from a *vicinal* surface: the peak locations follow the diffraction rods of the (115) and $(\bar{1}\bar{1}5)$ surface. We made a similar observation for deposition at 160 K (not shown [12]), but at that temperature the vicinal corresponds to the Cu(113), and the diffraction peaks were much weaker and broader [12]. These observations reveal that, at least at 200 K, the morphology is characterized by a sharp width distribution of terraces, which serves as the basic building block of the evolving rough interface in the form of pyramidlike structures. This state of the evolving interface is only reached after deposition of about 50 ML. Measurements during deposition reveal that the diffraction peaks (except those which are in phase) shift with increasing coverage. Figure 5 shows the position of a selected diffraction peak associated with step scattering as a function of time. It is seen that the terrace length runs through a sequence from longer to shorter terrace widths, until it locks into a steady state (at about the same coverage for which the diffuse scattering in the out of phase condition becomes stationary [12]). This real time observation allows us to conclude that the angle between the base and the sides of the pyramids increases in the course of the deposition process and finally saturates to the previously mentioned limiting values above about 50 ML.

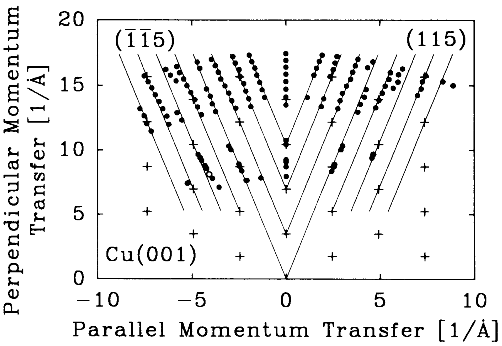

FIG. 4. Location of diffraction peaks on varying the perpendicular momentum transfer. Crosses denote bulk reciprocal lattice vectors in the ⟨110⟩ azimuth of the (001) surface. The diffraction rods correspond to the (115) and $(\bar{1}\bar{1}5)$ vicinal Cu surface. The data point marked by an open circle has been followed as a function of deposition time (see Fig. 5).

We note that the diffraction peaks associated with step edge scattering are only observed in the ⟨110⟩ azimuth, and not along the ⟨100⟩ direction, for which only the delta and the diffuse part are seen. This means that even after depositing 50 ML, steps are still running along the close packed directions [4].

The amplitude of the variation of the FWHM of the diffraction peaks along the rods is found to be much larger at 160 K as compared to 200 K [12], which indicates that the pyramidlike structures are less well developed at the lower temperature. This suggests that the inhibition of interlayer mass transport is less effective and might explain our finding that β is smaller at the lower temperature. Zhang *et al.* [6] have shown that β is a function of the height of the excess energy to diffusion at step edges, dropping from $\frac{1}{2}$ for perfect Schwöbel barriers to lower values, when the height of the barrier is decreased. At present, it is not clear if the height of the barrier itself changes, or if the Schwöbel barrier simply becomes less effective, when islands become small and ramified, as suggested by Kunkel *et al.* [16]. For the Cu on Cu(100) system, we have previously shown that at 160

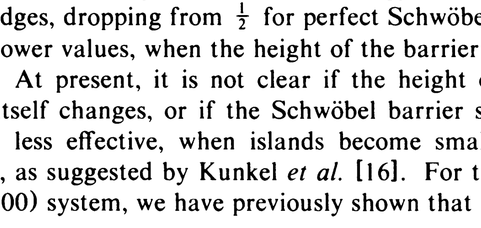

FIG. 5. Position of selected diffraction peaks associated with step edge scattering as a function of coverage, for deposition at 160 and 200 K.

K islands are indeed smaller and step edge diffusion is largely negligible [11,17].

In view of the result of Zhang *et al.* [6], one might suspect that unstable growth with a nonperfect Schwöbel barrier could produce both $\alpha \approx 1$, $\beta < 0.5$, and a not so sharp distribution of terrace widths. In that case, our finding $\beta \approx \frac{1}{4}$ and $\alpha \approx 1$ at 160 K could be by coincidence that of the linear diffusion equation [1]; it is therefore not an unambiguous indication that the kinetics is governed by this equation at that temperature.

We note that for the Cu system, intensity oscillations of the specularly reflected beam have been observed in this temperature range [8]. This is in contrast to the Pt on Pt(111) case [16], for which the total absence of oscillations has been explained in terms of the suppression of interlayer mass transport. A subsequent scanning tunneling microscopy (STM) study [18] revealed the formation of pyramidlike structures under the condition. However, the evolution of the morphology *during* deposition could not be addressed with the STM. Our real time characterization of the statistical properties of the evolving growth front provides a detailed scenario of how a singular surface can transform into an arrangement of vicinal surfaces through unstable growth. We emphasize that this happens, although intensity oscillations in the specular are observed. One may speculate [5,6] that the Schwöbel barrier and this kind of instability on singular surfaces are much more common than previously thought.

Nevertheless, some questions remain open. Why do the sides of the pyramids consist of those particular terrace lengths found and not others? Does diffusion alone explain why they change with temperature? An interesting conclusion has been drawn from molecular statics calculation by Smilauer, Wilby, and Vvedensky [6] and Spanjaard and Desjonquères [19]. In fact, the excess barrier to diffusion at step edges should not make it difficult for atoms to hop down, but to approach the step edge. Also step-step interaction may be important. Deposition on stepped surfaces might help to clarify these open questions. This work is in progress.

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[13] Models appropriate for thin film growth via sputtering or CVD, for which atoms are deposited at arbitrary angle, predict an instability due to shadowing; see C. Roland *et al.*, Phys. Rev. Lett. **66**, 2104 (1991); R. P. U. Karunasiri *et al.*, Phys. Rev. Lett. **62**, 788 (1989). In MBE, atoms come in at fixed angle, usually normal. In our real time measurements, the angle of the impinging Cu flux was varying during deposition. For the determination of the width of the interface, the angle was kept fixed at near normal. We found the He scattering angular distributions to be the same in both cases. Thus, we conclude that in this experiment the shadowing effect does not play a significant role.

[14] For a perfect pyramid, $H(r) \propto r^2$, for r smaller than half the base of the pyramid. This means $\alpha = 1$; see Eq. (1).

[15] The exponent β found at 200 K is even larger than 0.5, that expected when interlayer mass transport is completely inhibited. A value larger than this is possible only when "uphill" diffusion is allowed. $\beta = \frac{1}{2}$ is within the 95% confidence limit of the experimentally determined value.

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