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SURFACE PROFILE EVOLUTION ABOVE AND BELOW THE ROUGHENING TRANSITION[1]

W. Selke[2], P.M. Duxbury[3]
Institut für Festkörperforschung Forschungszentrum Jülich D-52425 Jülich, Germany

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The relaxation towards equilibrium of a profile imprinted on a crystal surface is studied above and below the roughening transition of that surface. Evaporation dynamics and surface diffusion are considered. Experiments and various theoretical approaches, including continuum theories and simulations, show significantly different features for profile shapes and decay times on rough and smooth surfaces.

1. Introduction

In equilibrium a surface is microscopically smooth at low temperatures while it becomes rough at and above the roughening transition temperature, T_R [1,2]. This phase transition has been studied extensively, both experimentally and theoretically, for stepped (or vicinal) crystal surfaces ("step roughening") and for surfaces of low Miller indices. Macroscopically, the transition is characterized by the disappearance of a facet of a given orientation from the equilibrium crystal shape.

The effect of roughening on the relaxation dynamics for surfaces has attracted much attention in recent years, initiated by the systematic experimental investigations of Bonzel et al. [3] on the flattening of one–dimensional grooves. By etching, a periodic profile is imprinted on a crystal surface. During the relaxation process, the grooves acquire, roughly speaking, either a sinusoidal shape or display flat parts ("broadening") at the top and bottom of the profile. Indeed, these two different types of profiles may be explained by the fact that the surface is either above or below its roughening transition temperature, T_R [4].

The healing of grooves has been studied extensively theoretically, using phenomenological descriptions and Monte Carlo simulations of SOS models. Particle transport by evaporation– condensation and surface diffusion has been considered. The interested reader is referred to the brief reviews [3, 5].

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[2] e-mail address: IFF072@DJUKFA11.BITNET

[3] Permanent address: Department of Physics and Astronomy and Center for Fundamental Materials Research, Michigan State University, East Lansing, MI 48824-1116, USA

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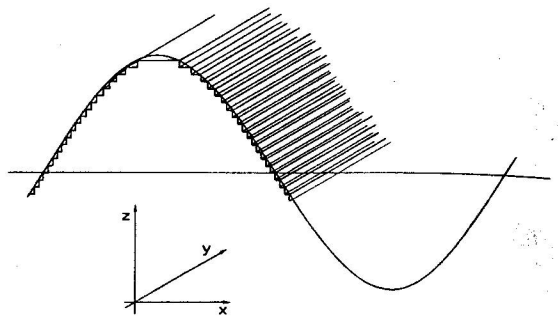

Fig. 1. Geometry of the grooved surface.

In this contribution, we shall present additional results, especially on comparing continuum theories to simulational data, above and below roughening. Above roughening, $T > T_R$, the classical description of Mullins is recovered in the limit of small amplitude of the sinusoidal profile, compared to the wavelength of the grooves [6]. Results are given in Section 2 together with extensions of Mullins' theory. In Section 3, the evaporation kinetics below roughening is considered, pointing out some open questions. In particular, the atomistic nature of the problem, as realised in the simulations, is seen in the dynamics of the top terrace and of its bounding steps, with meandering of the steps as well as forming and shrinking of islands [7]. We introduce a simpler model for the top terrace dynamics, in which we analyse the decay of a single terrace, bounded by two wandering steps. Results are presented in Section 4. A short summary concludes the article.

2. Flattening above roughening

A. Mullins' theory

The flattening process of one–dimensional periodic grooves, see Fig. 1, was described in a continuum theory by Mullins [8], assuming a small slope everywhere on the profile, isotropic surface free energy and the mobility being also independent of the orientation of the surface. With these assumptions, Mullins showed that the time evolution of the profile $z(x,t)$ (x refers to the direction of the modulation) is determined, for evaporation–condensation ($e-c$), by the diffusion equation

$$\frac{\partial z}{\partial t} = z_t = E\frac{\partial^2 z}{\partial x^2} = E z_{xx} \tag{1}$$

where E is a temperature dependent coefficient. In case of surface diffusion ($s-d$) the flattening process is described by the equation

$$z_t = -F\ z_{xxxx} \tag{2}$$

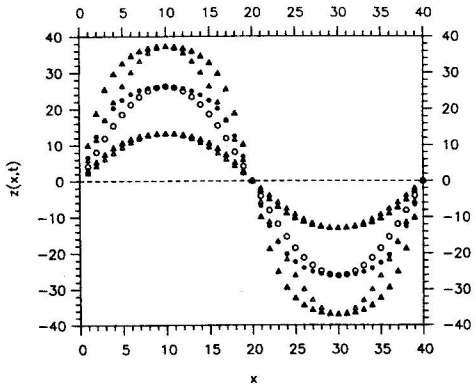

Fig. 2. Profiles $z(x,t)$ in the one–dimensional SOS model in the case of surface diffusion at $k_BT/J = 0.8$ with initial amplitude $A_0 = 45.5$, comparing Monte Carlo data (full symbols) with the pure sine form (open symbols).

with the constant F being proportional to the diffusion coefficient. Accordingly, an initial sinusoidal profile evolves, for evaporation–condensation, as

$$z(x,t) = z_0 \sin\,(2\pi x/L) \exp\,(-E(2\pi/L)^2 t) \tag{3}$$

and, in the case of surface diffusion, as

$$z(x,t) = z_0 \sin\,(2\pi x/L) \exp\,(-F(2\pi/L)^4 t) \tag{4}$$

i.e. the profile shape remains a pure sine wave, and the relaxation is purely exponential, with the relaxation time, τ, being proportional to $L^2(L^4)$ for $e-c(s-d)$, L is the wavelength of the groove.

B. Simulations and extensions of Mullins' theory

The predictions of Mullins' theory have been tested by simulating the two–dimensional SOS model at $T > T_R$ [7] and the one–dimensional SOS and Gaussian models [6]. In one dimension, the roughening temperature $T_R = 0$. These models characterise the surface by a height variable, h_ℓ, at site ℓ. In the SOS model, the interaction between neighbouring sites (ℓ, m) is given by the bonding energy J, with the Hamiltonian

$$\mathcal{H} = \Sigma_{\ell,m}\ J|h_\ell - h_m|, \tag{5}$$

while in the discrete Gaussian model the Hamiltonian has the form

$$\mathcal{H} = \Sigma_{\ell,m}\ J(h_\ell - h_m)^2\ . \tag{6}$$

In the simulations, Mullins' theory is nicely recovered when the amplitude, A, of the profile is sufficiently small compared to the wavelength, L, of the groove. Corrections to that theory manifest themselves in a broadening of the profile shape, compared to

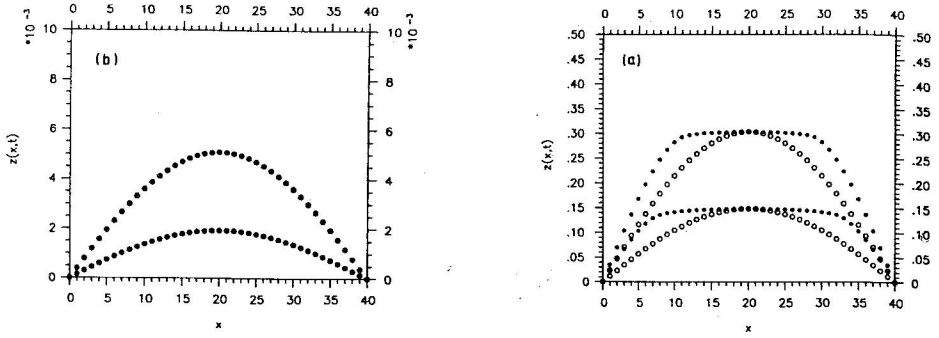

Fig. 3. Numerical solution of the continuum theory, equations (8) and (9), for the profile with $C\mu(\Theta) = 1$ and $\Theta_0 = 0.05$ (full dots), compared to the sinusoidal form (circles). The initial profile shape is sinusoidal with the amplitude being 0.5. Both the broadening (a) and the approach to Mullins' limit (b) are shown.

the sinusoidal form, and in deviations from the purely exponential decay law, where the effective relaxation time may be larger or smaller than the asymptotic one (for a detailed discussion, see Ref. 6). An example is depicted in Fig. 2, showing a pronounced broadening of the profile at large amplitude.

Prerequisites for Mullins' theory are the small slope of the profile, i.e. $A/L \ll 1$, and the isotropic surface free energy, $\sigma(\Theta)$, and mobility, $\mu(\Theta)$, where Θ is the angle describing the orientation of the surface. In case of the one–dimensional SOS model, it follows from the exact expression for $\sigma(\Theta)$ that the free energy is (nearly) isotropic for

$$A/L \ll \exp\,(-J/k_BT) \tag{7}$$

Indeed, the simulations show a crossover to Mullins' predictions when this condition is satisfied [6]. The anisotropy in the mobility (if present at all above roughening) does not appear to alter these conclusions (see also below).

Mullins' theory may be extended in a straightforward way by allowing arbitrary smooth slopes of the profile as well as anisotropic surface free energy[9] and mobility[10]. We restrict our discussion to the case of evaporation kinetics, as there it is possible to compare in detail with direct Monte Carlo simulations both above and below roughening. In the case of evaporation-condensation, the extension of (1) to include arbitrary slopes and anisotropy leads to,

$$z_t = C\mu(\Theta)z_{xx}[\sigma(\Theta) + \sigma''(\Theta)]/(1 + {z_x}^2) \tag{8}$$

where C is a constant, and $\sigma''(\Theta) = d^2\sigma/d\Theta^2$ with the angle $\Theta = \tan^{-1}(z_x)$.

An increase of σ with angle Θ is expected to lead to a broadening of the profile, because the flat surface is favoured, while an increase of the mobility with Θ may result in a sharpening of the profile, because the sides of the profile, having a larger inclination, decay faster than the top or bottom. This picture of conflicting tendencies

in the profile shape due to the anisotropic surface free energy and mobility is confirmed by our numerical analyses of equation (8).

For simplicity, we set $C = 1$ and assume

$$\sigma(\Theta) = (\Theta^2 + {\Theta_0}^2)^{1/2} \tag{9}$$

For isotropic mobility, taking $\mu(\Theta) = \text{constant} = 1$, the initial sinusoidal profile broadens; at later times, when $A/L \ll \Theta_0 \sim exp(-J/(T - T_R))$, the profile becomes, in the Mullins' limit, sinusoidal again. An example is depicted in Fig. 3. The corresponding temporal decay also resembles qualitatively the Monte Carlo findings, with deviations from the exponential form at early times. By taking into account an anisotropic mobility, similar to (9), one may easily monitor a crossover between a sharpening and a broadening of the profile, typically accompanied by a rapid change in the time scale. For instance, we observed the scenario where the initial sinusoidal profile first sharpened, then broadened and finally recovered the sinusoidal shape. However, in the Monte Carlo simulations the sharpening in the profile was not seen, giving additional evidence that the anisotropy of the mobility plays no significant role above roughening for the systems considered.

3. Evaporation kinetics below roughening

Below roughening, the surface free energy on vicinal surfaces $\sigma(\Theta)$ displays a cusp at $\Theta = 0$ [1,2], which is approximated by $\sigma(\Theta) \sim G_1|\Theta| + G_3|\Theta|^\gamma$. The first term is the free energy due to isolated steps, while the second term is due to step-step repulsion. If step fluctuations mediate the step-step repulsion, $\gamma = 3$. In the thermodynamic limit, the linear Mullins' theory is no longer valid, and several modified continuum theories have been suggested [4,5,10,11]. In the following we shall consider the case of $(e - c)$. Due to the singular nature of $\sigma(\Theta)$ below roughening, there is a $\delta(\Theta)$ singularity in $\sigma''(\Theta)$ there. However Spohn has argued that because $\mu(|\Theta|) \sim |\Theta|$ at small $|\Theta|$ in the case of evaporation-condensation and in the thermodynamic limit, the $\delta(\Theta)$ is removed (note that due to finite size effects in both the free energy and mobility, Spohn's argument is not entirely convincing - see below). Accepting this analysis and making the small slope approximation, one finds

$$z_t = D\ {z_x}^2\ z_{xx} \tag{10}$$

where D is a constant. As noted before by Lancon and Villain [4], this equation implies a sharpening of the initial sinusoidal profile near its top and bottom, where the deviation from the amplitude $\delta z(\delta x, t)$ behaves like $\delta z \sim |\delta x|^{4/3}$. The simulated profile shapes, for the two–dimensional SOS and Gaussian models, are in marked contrast to that prediction, showing, at amplitudes close to integers, a broadening near the top and bottom [7].

The discrepancy between continuum theory and the Monte Carlo simulations is illustrated in Fig. 4, depicting new data for the SOS model, well below $T_R(k_BT/J = 0.8$, while $k_BT_R/J \approx 1.25)$. The discrepancy may be either due to finite–size effects in the

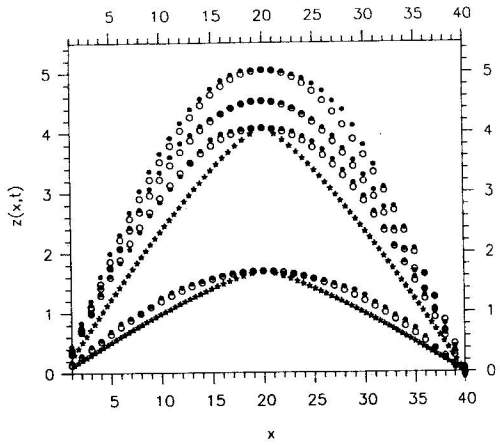

Fig. 4. Profiles $z(x,t)$ obtained from simulating the two–dimensional SOS model of size 80×1000, $A_0 = 9.5$, at $k_BT/J = 0.8$ with evaporation kinetics (full dots), compared to purely sinusoidal shapes (circles) and the prediction of the continuum theory, equation (10) with $D = 1$ (stars).

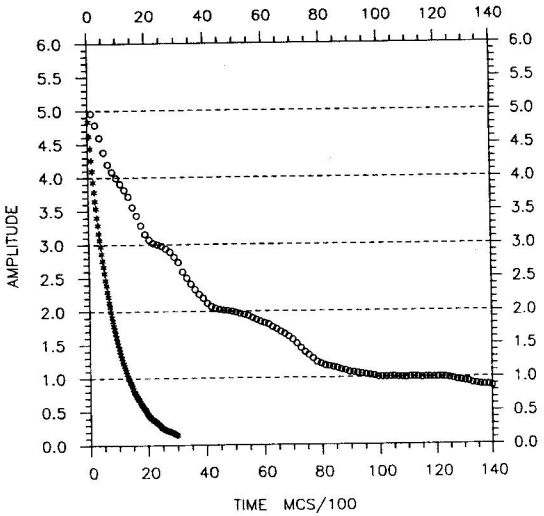

Fig. 5. Time dependence of the amplitude, obtained from simulations with evaporation dynamics for SOS models of size $80 \times M$, $M = 1$ (asterisks) and 1000 (circles), at $k_BT/J = 0.8$, with initial amplitude $A_0 = 5.5$. In the one–dimensional case, an average over 25000 realisations was taken. The time is measured in MC steps per site.

simulations, as suggested by Spohn [10], or it may indicate a possible inadequacy of that continuum theory. Actually, the prior simulations were performed at, typically,

$0.25 \lesssim A/L \lesssim 0.025$, with L ranging from 20 to 140. The broadening was observed for single realizations, and it persisted by averaging over many realizations or by doing a coarse graining in time [7]. A systematic study of finite–size effects would be presumably rather involved, as inferred from the finite–size analyses for the scaling behaviour of the relaxation time with the wavelength, see below and Ref. 7. In any event, we did new simulations for SOS systems of rectangular size, $L \times M$, being rather large in the transverse direction, $M = 200$, 500 and 1000, with the wavelength $L = 40$, 80, and 160, and the initial amplitude A_0 of the discretised sine function being $A_0 = 5.5$ and 9.5, at $k_BT = 0.8J$. We found no evidence for a sharpening of the kind predicted by the continuum theory, see also Fig. 4. In fact, the broadening of the profile of a single realization near its top and bottom seems to become more pronounced with increasing L, at constant amplitude. The broadening occurs when the top terrace is nearly flat, bounded by meandering steps to the neighbouring terraces. When these two steps annihilate and islands are formed on the top terrace, which eventually shrink and vanish, the profile takes an almost sinusoidal form. After dissolving the islands, the top terrace is nearly flat again, with the height of the profile being reduced by one lattice constant. In that way, one observes shape fluctuations which are accompanied by two distinct time scales in the flattening process, reflecting the step wandering and the shrinking of islands, see Fig. 5 and Ref. 7.

To quantify possible finite–size effects, we also considered the mobility $\mu(\Theta = 0)$, defined by

$$\mu(\Theta = 0) = v/h \tag{11}$$

where v is the velocity of an advancing surface under the action of an external field h, with h being small [10]. In simulations of SOS models, the change in energy for an elementary Monte Carlo move may be reduced by h, if the height of a site would be increased by one, while it remains unchanged by lowering the height by one, thereby favouring adsorption (condensation) against desorption (evaporation).

From standard nucleation theory, one expects, below roughening, for an infinite surface

$$v \sim \exp(-h/k_BT) \tag{12}$$

giving rise to the vanishing mobility, mentioned above. For finite systems of $N \times N$ sites, one may easily see, by comparing the energy barriers for advancing and receding surfaces in SOS models, that

$$v \sim hN^2\, e^{-NJ/k_BT}/k_BT \tag{13}$$

i.e. the mobility approaches zero exponentially as the sample size increases. We confirmed this behaviour in simulations, which also show rather drastic corrections to the linear relation between the field and the growth velocity at larger fields, leading to a quicker advancement of the surface. However, in the profile evolution problem, finite size effects are much stronger than that implied by equation (13), as the top terrace is bounded by two steps which effectively means that the mobility of the top terrace is really a mobility at a small but finite angle $\Theta \sim 1/w_t$, where w_t is the width of the top terrace. Since below roughening, we expect $\mu \sim |\Theta|$, the top terrace mobility, μ_t,

for grooves of wavelength, L, scales to zero at most as $\mu_t \sim 1/w_t > 1/L$. This is a very slow approach to zero (much slower than (13)). In addition there are also finite size corrections to the surface free energy, so even in the thermodynamic limit it is not clear that the mobility singularity is strong enough to remove the δ function singularity as suggested by Spohn[10]. It is clear that a more careful study of finite size effects is necessary before one can be sure that even the continuum theory predicts a sharpening in the thermodynamic limit.

Aside from the problem of finite size effects, the Monte Carlo data illustrates the fact that there are two distinct time scales which are important in profile decay below roughening. There is a rather slow time scale during which the two steps bounding the top terrace wander but do not touch, and a faster time scale, beginning when the two top steps first touch and islands begin to form. Once the islands form on the top terrace, each island has a net curvature which drives a relatively rapid island evaporation. We have made some preliminary attempts to develop a theory incorporating these two time scales and one such attempt is as follows.

Assuming preservation of the sinusoidal profile shape after lowering the amplitude by one lattice constant (such truncated sinusoidal shapes have been also observed experimentally [12]), the extent of the flat top terrace may be approximated by

$$w_t(A) = \frac{L}{\pi} \cos^{-1}\,(A/A+1) \tag{14}$$

A taking integer values. The time needed to reduce the amplitude by one, Δ, should include the two timescales, step meandering and island shrinking, see Fig. 5. Neglecting memory effects, these times are expected to scale with the extent of the top terrace, w_t. Assuming the general scaling behaviour

$$\Delta \approx G\, w_t^a \tag{15}$$

where G is a temperature–dependent coefficient, implies the scaling of the amplitude

$$A(t, L) = \bar{A}(t/L^a) \quad . \tag{16}$$

From the prior simulations, an effective scaling exponent a_{eff} could be determined, depending rather strongly both on temperature and system size, especially the wavelength L. Its value is clearly larger than in the linear Mullins' case, $T > T_R$, where $a = 2$; a lower bound $a \gtrsim 3$ was given in[7].

From equations (14) and (15), and going over to a continuum description, one obtains

$$t = -\left(G\frac{L}{\pi}\right)^a \int_{A_0}^{A} \left[\cos^{-1}\left(\frac{x}{x+1}\right)\right]^a dx \tag{17}$$

where A_0 is the initial amplitude of the sinusoidal profile.

The resulting time dependence of the amplitude $A(t)$ agrees surprisingly well with our new simulational data, for moderate wavelengths at $k_BT/J = 0.8$, by adjusting the coefficient G and setting $a = 3$, see Fig. 6. However, the Monte Carlo data correspond

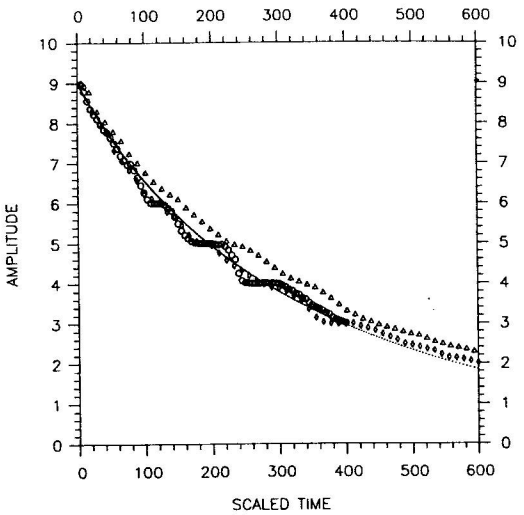

Fig. 6. Amplitude of the groove vs. scaled time, $t = t_{MC}(80/L)^a/10$, with t_{MC} denoting the number of MC steps per site; a is chosen to be 3. Monte Carlo data for systems of sizes 80×1000 (triangles), 120×500 (diamonds) and 160×500 (circles), at $k_BT/J = 0.8$, are compared with equation (16), setting $a = 3$ (dotted line).

to rather small values of w_t, and the value of a found may be invalid for larger system sizes. To gain more insight into the physics controlling the value of a, we have studied a simpler problem of two wandering steps which annihilate and this we now describe.

4. The annihilation of two wandering steps

We consider the SOS model on a rectangular lattice of $(L+2) \times M$ sites with the initial height configuration

$$h_{xy}(t=0) = \begin{cases} 0 & x = 1, L+2 \\ 1 & \text{otherwise} \end{cases} \tag{18}$$

We then monitor the time evolution of this isolated terrace. We fix the heights at the x–boundaries, $h_{1y}(t) = h_{L+2y}(t) = 0$, and take periodic boundary conditions in the y–direction. This geometry introduces two steps which fluctuate, touch and form islands, and finally the system reaches the equilibrium flat state. The "amplitude" of the step, A_s, is given by

$$A_s(t) = \max_x \langle \Sigma_y\; h_{xy}(t)/M \rangle \tag{19}$$

where the brackets $\langle\ \rangle$ denote the thermal average.

Using $(e-c)$ kinetics, simulations were done for systems below roughening ($k_BT/J = 0.8$, $M = 200$, and L ranging from 4 to 32) and above roughening.

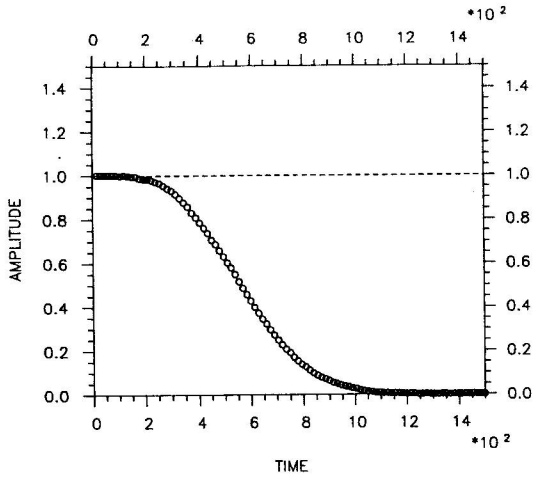

Fig. 7. Amplitude vs. time, measured in MC steps per site, for the two-step SOS model, equation (17), using Monte Carlo techniques with evaporation kinetics. The system size is 18×200; an average over 100 realisations was performed.

Below roughening, one readily distinguishes two stages; an example is depicted in Fig. 7. In the first stage, the amplitude A_s fluctuates around one. A typical configuration consists of two wandering interfaces, separating the "0" regions from the "1" region in the center of the system. In the second stage, which initiates when the two interfaces collide, A_s decreases quickly. This behavior is similar to that of the top terrace of the grooved surface. From analytic considerations [13, 14], related simulations [15, 16] and experiments [17] one expects that the width of an initially straight one-dimensional interface of length M grows as $t^{1/4}$, after a short time transient and well before saturation to its maximal value proportional to $M^{1/2}$. Accordingly, the time needed for the collision of the two interfaces, t_c, is expected to scale like L^4, provided M is sufficiently large.

Our data show pronounced corrections to the expected asymptotic behaviour. The effective scaling exponent, defined by

$$\alpha_{eff} = d \ln t_c / d \ln L \quad , \tag{20}$$

with the definition $A_s(t_c) = 0.95$ is found to increase from about 2.5, for the smallest systems, $L = 4$ and 8, to about 3.1, for the largest systems, $L = 24$ and 32. The continuous change in α_{eff} reflects, presumably, a transient behaviour, enhanced by the boundary-limited fluctuations of the interface. Interestingly, these values for α_{eff} are rather close to those for the effective scaling exponent a_{eff} for grooves with top terraces of similar extent. Therefore, one may speculate that both scaling exponents, a and α, approach asymptotically the value of 4 (we tacitly assumed that the meandering stage leads to the dominant time scale as compared to the dynamics of the islands). This

hypothesis is supported (at least for α) by a stochastic-Langevin-equation model which we have developed. In this model, a meandering wall lies between a perfectly reflecting boundary and a perfectly absorbing boundary. If the interface starts out near the repelling wall, fluctuations drive it toward the absorbing wall, which is at a distance W from the repelling wall, and eventually the interface is stuck to the absorbing wall. This system clearly shows, after some initial behaviour, the $t^{1/4}$ time scale associated with the movement of the interface away from the repelling wall, provided the transverse length $M > W^2$. There is also a shorter timescale, during which "island" shrinking occurs, which initiates when part of the interface first sticks to the absorbing wall. This implies that the overall time to stick to the absorbing wall scales as W^4.

Of course, additional analyses are needed to substantiate the suggestion that the asymptotic exponent is 4, especially for the grooved surface consisting of an array of meandering and interacting steps.

Above roughening, the amplitude of a single step, $A_s(t)$, is observed to decay exponentially, as is the case for the grooved surface.

From the analogy to the problem of two annihilating steps, one may readily explain the non-monotonic dependence of the relaxation time of the grooved surface on the transverse direction M. Certainly, in the one-dimensional limit, $M = 1$, the flattening occurs most rapidly, see Fig. 5. For intermediate values the decay is observed to proceed most slowly, because the wandering of the steps is limited by an amount proportional to $M^{1/2}$. Finally at large M one expects a slow decrease in the decay time due to the greater probability of a "rare" large step fluctuation leading to premature islanding of the top terrace.

5. Summary

The equilibration of grooved surfaces has been studied using various theoretical approaches, including extensions of continuum theories and Monte Carlo simulations. New results have been obtained for particle transport by evaporation-condensation.

Above roughening (by going either to the one-dimensional situation or to higher temperatures), the observed broadening of the profile shape, as compared to the sinusoidal form, and deviations from the exponential decay law may be reproduced by an extension of the classical theory of Mullins, taking into account the anisotropy of the surface free energy.

Below roughening, modifications of the continuum theory are needed, which predicts a sharpening of the profile shapes, to reproduce the profiles found in Monte Carlo simulations. For instance, finite-size effects in the inclination-dependent mobility seem to play an important role.

To explain quantitatively the scaling behaviour of the relaxation time with the wavelength, L, of the groove, it is necessary to combine the annihilation dynamics of the top terrace with the repulsive spreading of the steps on the sides of the profile. For evaporation-condensation, just considering the top step annihilation leads to the scaling $\tau \sim L^4$, which gives an exponent much larger than the linear prediction 2.

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