Growth of Patterned Surfaces

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During epitaxial crystal growth a pattern that has initially been imprinted on a surface approximately reproduces itself after the deposition of an integer number of monolayers. Computer simulations of the one-dimensional case show that the quality of reproduction decays exponentially with a characteristic time which is linear in the activation energy of surface diffusion. We argue that this lifetime of a pattern is optimized if the characteristic feature size of the pattern is larger than $(D/F)^{1/(d+2)}$, where D is the surface diffusion constant, F the deposition rate, and d the surface dimension. [S0031-9007(97)04838-2]

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Modern techniques of manipulating crystal surfaces allow one to imprint structures down to the atomic size on them. With the tip of a tunneling microscope one can arrange adsorbed atoms in a pattern. In heteroepitaxial growth a two-dimensional array of quantum dots may form on the nanometer scale. With masking techniques arbitrary patterns with features as small as a micrometer along the surface and atomic size perpendicular to it can be fabricated. If one buries such a pattern under an overlayer growing in layer-by-layer mode, the pattern will approximately reproduce itself periodically at the completion of each layer. The question arises as to how this propagation of a pattern is influenced by the growth conditions. A theoretical understanding of such temporal correlations began to emerge only recently [1–5]. In the following, the propagation of a pattern will be discussed for the simplest case that the influence of elastic strain on surface diffusion is negligible and that diffusion across a step edge from an upper to a lower terrace is not inhibited by Ehrlich-Schwoebel barriers [6].

Experimentally, it has long been known that layer-by-layer (or Frank–van der Merwe) growth persists up to a time $\tilde{t}$, after which the characteristic growth oscillations are damped out and the surface becomes rough. Recently, it has been shown that $\tilde{t}$ depends on the growth conditions [2,7–9], i.e., the surface diffusion constant D and the deposition rate F, with a power law $(D/F)^{\delta}$. Clearly, a pattern can at most survive as long as the surface grows layerwise. In fact, the lifetime of a pattern is much shorter than $\tilde{t}$, as we are going to show: It depends on D/F only logarithmically rather than with a power law.

The lifetime of a pattern in layer-by-layer growth depends also on the feature size r. We shall identify the length scale that is important in the context of pattern decay as $\ell_0 \sim (D/F)^{1/(d+2)}$. For feature size $r \gtrsim \ell_0$, the lifetime increases rather abruptly.

Propagation probability.—To simplify the discussion, we assume that the grown crystal is simple cubic [10] and that overhangs and defects can be neglected. Then, the surface configuration at a given time t can be represented by a height function $h(x,t)$ with x indexing the lattice sites on a d-dimensional substrate. In the following, time will be measured in numbers of deposited monolayers.

When the growth process is initiated at time $t = 0$ with some structure $h(x,0)$, consisting of, e.g., islands of a particular shape on an otherwise flat substrate (a *pattern*), new islands will form on top of the initial ones, and all of them will expand laterally due to the attachment of atoms at their edges. Therefore the pattern will be deformed during this early stage of growth. However, the original pattern will nearly be reproduced after the deposition of one monolayer, because the new layer nucleates preferentially near the centers of islands in the previous layer. As this correlation extends over long times [1], one expects the approximate reproduction of the pattern at later times as well. It makes sense to ask which fraction of the pattern is propagated through t monolayers. We call this fraction the propagation probability $p(t)$. It is given by

$$p(t) \equiv \left\langle \prod_{s=1}^{t} \delta_{h(x,s),h(x,0)+s} \right\rangle. \tag{1}$$

The brackets denote averaging over different lattice sites x and different realizations of the growth process. $\delta_{i,j} = 1$ if $i = j$ and 0 otherwise denotes the Kronecker delta.

By defining $p(t)$ in this way, we measure the *deterministic* reproduction of the initial pattern $h(x,0)$, seen stroboscopically after deposition of $1, 2, \ldots,$ monolayers in the comoving frame. It is *deterministic* in the sense that a given site x is counted as "surviving" after time t only if it has survived through *all* previous times $1, \ldots, t$. Of course, the height might also regain its initial value by chance once it has left it, but this is a stochastic process and contributes to the noisiness of the pattern. Thus the survival of *information* is not described properly by the propagation probability (1), but by an appropriate entropy. It will be dealt with in a longer paper.

If the probability of return to the initial height (in the comoving frame) at time t was considered, irrespective of the values at intermediate times, one would probe a

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two-point function. Such a two-point function is expected to decay algebraically for long times. More precisely, it should scale like the value of the height distribution function at the average height. For self-affine surfaces the width of the height distribution increases like t^{β}. Thus the probability to recover the initial height after the deposition of t monolayers should decay as $t^{-\beta}$.

In contrast to this, the t-point function $p(t)$ decays exponentially; see Fig. 1. Although the exact evaluation is nontrivial, this result is easily made plausible: The fraction $p(t+1)$ of surviving sites at time $t+1$ equals the number of surviving sites at time t, $p(t)$, times the probability to propagate to the next layer. Assuming that this probability can be identified with $p(1)$, independent of the surface configuration, which at time t, of course, differs from the initial one, the exponential decay

$$p(t) = \exp(-t/t_c) \tag{2}$$

follows immediately. We shall show below that the propagation probability does depend on the surface configuration. However, this dependence is so weak that the surface evolution during the lifetime t_c of a pattern hardly affects its exponential decay (2).

The main purpose of this Letter is to investigate the dependence of t_c on the microscopic growth parameter D/F and the feature size of the pattern. Let us first discuss two limit cases. For $D/F \to 0$, the sites are not coupled by diffusion and the appropriate description of the growth process is given by the random deposition model [11], where atoms are deposited randomly onto the substrate and remain at the deposition site forever. In this model, the fraction $S_j(t)$ of surface sites in layer j after time t is a Poisson distribution, $S_j(t) = \exp(-t)t^j/j!$. Therefore, $p(1) = S_1(1) = \exp(-1)$, so that $t_c = 1$.

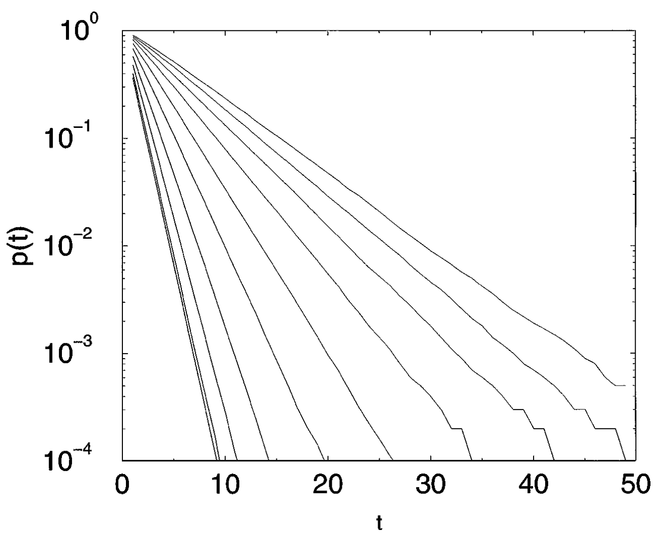

FIG. 1. Propagation probability $p(t)$ of a flat surface as a function of a number of deposited monolayers t for different values of $D/F = 0, 10^0, 10^1, \ldots, 10^8$ from left to right. The characteristic time of the exponential decay increases with increasing D/F.

For $D/F \gg 1$, a monotonous increase of t_c as a function of D/F is expected. When the initial "pattern" is simply a flat surface, one expects $t_c \to \infty$ for $D/F \to \infty$, because layer-by-layer growth persists forever for infinitely high diffusion constant. The computer simulations of molecular beam epitaxy (MBE) on a one-dimensional substrate, to be presented in the next section, confirm this picture and show a dependence

$$t_c \sim \ln(D/F) \tag{3}$$

with a cutoff at $t_c = 1$ for small D/F.

In the following we shall discuss three different initial patterns: (1) A completely flat surface, (2) a rough surface as it evolves from the flat one after time $\tilde{t}$, when the oscillations due to layer-by-layer growth have died out, and (3) a periodic modulation of the surface with a fixed feature size. The first arises as a natural limit of a pattern with a characteristic feature size $r \to \infty$. The second represents the simplest generic configuration for which the growth kinetics has no periodic time dependence any more. Both will be used to study the pattern decay process systematically in the next section. Finally, the third choice will lead to the optimization condition for pattern survival and will be studied afterwards.

Model and simulation results.—Atoms are deposited onto a one-dimensional substrate of typical size $L = 10^4$ with a rate of F atoms per unit time and area. Atoms with no lateral neighbor are allowed to diffuse with diffusion constant D. Atoms with lateral neighbors are assumed to be immobile, so that, e.g., dimers are immobile and stable. Growth commences with a flat substrate, $h(x,0) = 0$ for all sites $x = 1, \ldots, L$. (The other initial configurations will be discussed below.) On deposition at x, $h(x,t)$ is increased by one.

Figure 1 confirms the exponential decay (2) of the propagation probability. The initial configuration survives the better the higher the value of D/F, as expected. The lifetimes $t_c(D/F)$ are shown in Fig. 2. For small D/F the lifetime approaches 1 as derived above for random deposition, and for large D/F it increases like $\ln(D/F)$.

The results for a rough surface as initial pattern are very similar. The propagation probability (1) was averaged over 100 different initial patterns, all obtained by depositing 50, respectively, 200 monolayers on a flat substrate. For the values of D/F considered here the periodic oscillations of the surface morphology have stopped by then, as shown in [2]. Interestingly, the lifetime of a rough surface pattern is shorter than that of the flat surface (see Fig. 2), but we could not observe any difference between the rougher surface (200 monolayers) and the less rough one (50 monolayers). This will become plausible, when considering how the lifetime of a pattern is affected by the feature size.

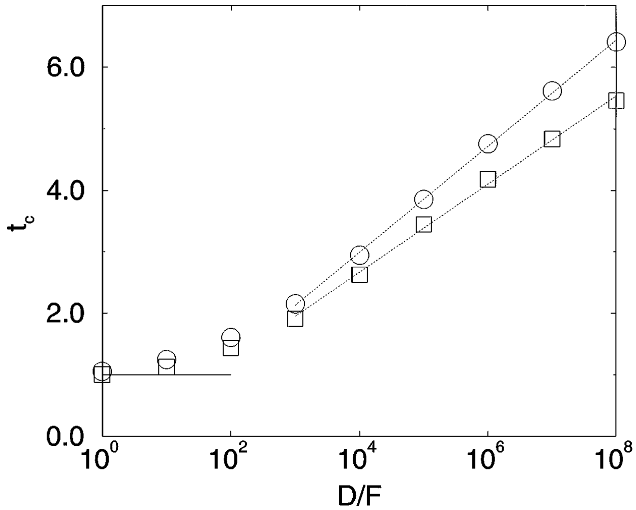

FIG. 2. Pattern lifetime t_c as a function of D/F for a flat (∘) and a rough (□) surface. The solid line indicates the minimum lifetime $t_c = 1$ for $D/F \to 0$. The dotted lines represent logarithmic fits to the last five decades of D/F.

Periodic patterns.—In this section, we study the dependence of the lifetime on the typical feature size of an artificially prepared initial configuration. To this end, it is useful to recall the different length scales associated with ideal MBE.

The island distance or diffusion length ℓ is a function of D/F,

$$\ell \sim (D/F)^{\gamma}. \tag{4}$$

The exponent γ depends on the substrate dimension, on the size of the critical nucleus, and on the possible fractality of the islands [8,9,12]. Its numerical value for the simulations presented here is $\gamma = 1/4$.

The only dimensionless length scale ℓ_0 which can be constructed from the dimensionful parameters D and F is

$$\ell_0 \sim (D/F)^{1/(d+2)}. \tag{5}$$

Physically, this length scale comes from comparing the diffusion time to the adatom arrival time on an area of size l^d [8,13]. ℓ_0 and ℓ are submonolayer quantities.

Finally, the layer coherence length $\tilde{\ell}$ depends on D/F like

$$\tilde{\ell} \sim (D/F)^{4\gamma/(4-d)}; \tag{6}$$

see [2]. This is not a submonolayer quantity, as $\tilde{\ell}$ appears as the typical length scale after the oscillation damping time, i.e., after deposition of $\tilde{t}$ monolayers.

In order to study the length scale dependence of the lifetime, we use a periodically modulated surface

$$h(x, t = 0) = \Theta\big(\sin(\pi x/r)\big), \tag{7}$$

where $\Theta(x) = 1$ if $x \geq 0$ and 0 otherwise is the Heaviside function.

The measurements of the lifetime as a function of the initial wavelength r for different D/F (see Fig. 3) show that, for r greater than a characteristic value depending on D/F, survival is strongly enhanced. Above this value, the lifetime depends only a little on the feature size. For small r, the lifetime seems to saturate for increasing D/F. These findings can be understood in the following way.

The mechanism of transporting the memory of the surface structure from one monolayer to the next is that nucleations take place near the center of islands that have already formed one layer below. If the feature size r is chosen so small that nucleations cannot take place on top of the pattern, this mechanism is suppressed and consequently the lifetime of the pattern is reduced. To suppress nucleations, the distance between sinks for adatoms, i.e., the feature size r, has to be chosen so small that a freshly deposited adatom diffuses to the nearest sink (i.e., a distance r), before the next atom is deposited within the area $\sim r^d$. This is the case for $r \lesssim \ell_0$. Hence, ℓ_0 should be the length scale found in Fig. 3.

The scaling plot Fig. 4 shows that the characteristic length above which the survival is prolonged scales like $(D/F)^{0.32\pm0.01}$, which suggests a length scale proportional to $(D/F)^{1/3}$. This is in accordance with the argument given above for the characteristic length scale being ℓ_0. To assure that the characteristic length scale in Fig. 3 is not $\tilde{\ell}$, which for the parameters studied here also is proportional to $(D/F)^{1/3}$, we studied epitaxial growth with a critical nucleus of 2 instead of 1, which influences $\tilde{\ell}$, but not ℓ_0. This analysis shows that indeed ℓ_0 is the characteristic length scale. Details on this will be published in a longer paper.

The saturation of t_c for small r as a function of D/F can now easily be understood: If $r \ll \ell_0$, the memory of

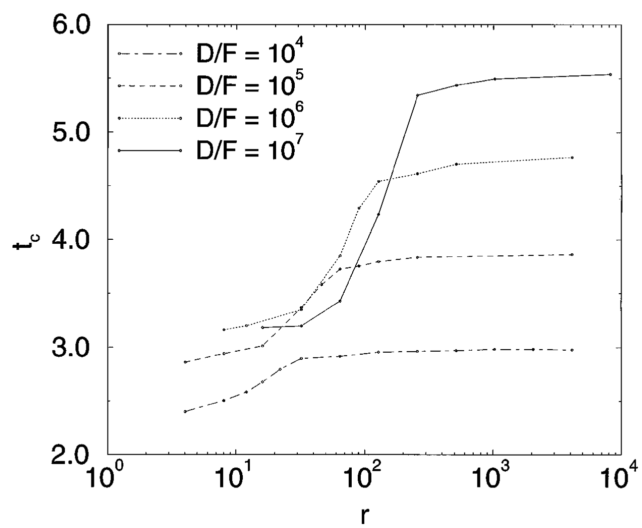

FIG. 3. Lifetime t_c as a function of the feature size r of the initial surface modulation for $D/F = 10^4, 10^5, 10^6$, and 10^7. Around a characteristic feature size depending on D/F, the lifetime increases rapidly.

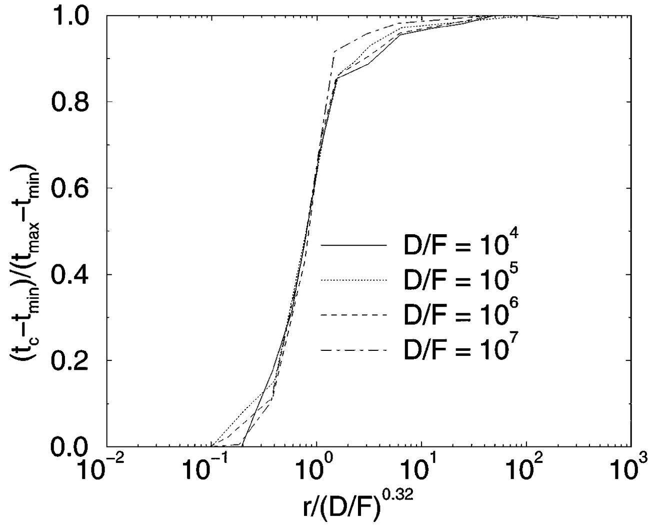

FIG. 4. Data from Fig. 3 with normalized lifetimes, and feature size r scaled with $(D/F)^{0.32}$.

the initial periodic pattern is destroyed already after the deposition of one monolayer. The surface will be half filled with islands which have a typical size ℓ much larger than r. Then the fraction of sites which propagated by just one lattice constant is about 50%, irrespective, how large D/F is.

The faster decay of a rough surface compared to a flat surface as the initial pattern can be made plausible with the following reasoning: The feature size of rough surfaces may be identified with the typical terrace size ℓ. For the simulations presented here, where the critical nucleus was 1, ℓ is smaller than ℓ_0. Therefore the faster decay of the pattern is consistent with our findings for periodic patterns.

In conclusion, we have shown that a pattern decays exponentially fast with a lifetime proportional to $\ln(D/F)$. With the Arrhenius law, $D \sim \exp(-E/k_B T)$, the lifetime decreases linearly with the energy barrier E for surface diffusion. The lifetime of a pattern is optimal if the feature size of the pattern is larger than $(D/F)^{1/(d+2)}$.

An important extension of this work would be the study of the two-dimensional case. Whereas it is natural to expect an exponential decay of the propagation probability, the dependence of the lifetime on the microscopic growth parameters is an open question.

In this paper, we neglected barriers for interlayer transport (Ehrlich-Schwoebel barriers) [6]. The memory mechanism will be enhanced by them, but the instability [14] associated with them will tend to make pattern reproduction worse. The competition between these two effects is well worth studying as in many materials interlayer transport is inhibited.

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