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Kinetic roughening of vicinal Si(001)

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Abstract

The kinetic roughening behavior of vicinal Si(001) surfaces is studied with scanning tunneling microscopy. By analyzing the height–height correlation function of the Si layers that have been grown we found, in the case of islands growth, an algebraic roughening behavior with a roughness exponent of 0.68 ± 0.05 . In the step flow mode, however, we found non-algebraic roughening behavior.

1. Introduction

Kinetic roughening in growth processes has attracted much attention recently [1,2]. This type of roughening, which emerges in non-equilibrium growth processes, is very interesting for several reasons. Firstly, it is of great technological importance in the design of surfaces with special roughness properties. A roughness tuned by the growth mode would be the ideal case. In most cases, however, one prefers that the surface of the thin film is as flat as possible. Secondly, it provides an excellent test case for theories of fractal growth, dynamic scaling and self-organized criticality in non-equilibrium situations. Furthermore, it is expected that in many other cases, e.g. the growth of tumors, flame fronts and colloid aggregates, the same kind of phenomena occurs.

During the growth process the roughness of the surface increases and a self-affine surface is

expected to develop [2]. We consider a part of a two-dimensional surface of extent L^2 which is initially flat ($t = 0$). During the growth process the surface becomes rougher and rougher. The surface can be described by a single-valued function $h(\mathbf{r}, t)$ which refers to the column height at position $\mathbf{r}$ and time t . The root-mean-square of the height fluctuations $\sigma(L, t)$ is a quantitative measure of the surface width and is defined by

$$\sigma(L, t) = [\langle h^2(\mathbf{r}, t) \rangle - \langle h(\mathbf{r}, t) \rangle^2]^{1/2}. \quad (1)$$

In the absence of any characteristic length or time scale, it is expected that initially $\sigma(L, t)$ grows with some power of the time t :

$$\sigma(L, t) \sim t^\beta. \quad (2)$$

The exponent β describes the growth of the correlations with time along the growth direction. The length L is the maximum spatial extent to which the correlations can grow parallel to the surface. As soon as the correlations have reached this maximum, they are not able to extend any further, and the width $\sigma(L, t)$ saturates at a constant value. The surface is scale invariant and

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the saturation value of σ is expected to obey a power law dependence on L ,

$$\sigma(L, t) \sim L^\alpha. \quad (3)$$

The dependence of σ on t and L given above can be rewritten in the form of one simple equation representing the dynamic scaling behavior [3]:

$$\sigma(L, t) \propto L^\alpha f\left(\frac{t}{L^{\alpha/\beta}}\right), \quad (4)$$

where α/β is the dynamic scaling exponent and $f(x)$ the scaling function defined as

$$f(x) \sim \begin{cases} x^\beta & x \ll 1 \\ \text{const} & x \gg 1 \end{cases}. \quad (5)$$

Numerical simulations of the Eden model and the ballistic deposition model have shown that the width indeed obeys the scaling relation of Eq. (4) [2,3].

An alternative way to describe the scaling behavior is given by the height–height correlation function. The height–height correlation function is defined as

$$G(\mathbf{r}, t) = \langle |h(\mathbf{r}', t') - h(\mathbf{r}' + \mathbf{r}, t + t')|^2 \rangle^{1/2}. \quad (6)$$

$G(\mathbf{r}, t)$ scales in the same way as the width except that L has to be replaced by r :

$$G(\mathbf{r}, t) \propto r^\alpha f\left(\frac{t}{r^\beta}\right), \quad (7)$$

with $f(x)$ analogous to Eq. (5). The scaling behavior of the surface has been extensively studied for a number of growth models trying to verify the scaling relations of Eqs. 4 and (7) and to determine the exponents α and β .

It is convenient to treat the problem of the time evolution of a growing surface in the continuum limit. Kardar, Parisi and Zhang [4] proposed the following non-linear Langevin equation in order to describe the growth process:

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h + \lambda (\nabla h)^2 + \eta(\mathbf{r}, t). \quad (8)$$

This equation is known as the KPZ equation [4]. The first term on the right-hand side describes surface relaxation and the second one refers to the lowest-order non-linear term that can appear in the interface growth equation. Finally, the last

term is a Gaussian random variable which mimics the local variations in the deposition rate.

In the trivial case $\nu = \lambda = 0$, the KPZ equation describes a simple uncorrelated random deposition model in which no spatial correlations are present. The only relevant correlations in the height fluctuations of the surface emerge in the time dependence ($\beta = 1/2$).

For $\lambda = 0$, the KPZ equation describes a random deposition process with finite surface diffusion and can be solved exactly leading to the scaling exponent $\beta = (3 - d)/4$ and $\alpha = (3 - d)/2$ below a critical dimension $d_c = 2 + 1$. In $d = 1 + 1$ dimensions, the result $\alpha = 1/2$ and $\beta = 1/4$ is found [5,6].

In the most general case $\lambda \neq 0$ and $\nu \neq 0$, the KPZ equation has only been solved in $d = 1 + 1$ leading to $\alpha = 1/2$ and $\beta = 1/3$ [4]. These values are in agreement with the Eden model [7–9] and the ballistic deposition model [10–12]. In $2 + 1$ dimensions, however, the KPZ equation cannot be treated by renormalization-group techniques and has to be solved numerically. Recent numerical solution of the KPZ equation [13] gives the following results for the exponents: $\beta = 0.25$ and $\alpha = 0.39$, respectively. These exponents are close to the values obtained in the restricted solid-on-solid model [14–16] but not in agreement with the Eden and the ballistic deposition model.

Recently, an anisotropic generalization of the KPZ equation has been proposed [17]. In this model it is suggested that, in the case of growth processes on a vicinal surface, algebraic scaling behavior as defined in Eq. (4) will only be found for layers grown in the island formation mode. Furthermore, a morphological transition to logarithmic roughness is predicted in the case of step flow growth [17]. In this anisotropic KPZ equation a growth rate depending on the step density was necessary to explain the above-described morphological transition. Desorption of atoms during molecular beam epitaxy (MBE) is, however, a very unlikely phenomenon even for a low step density, i.e. very flat surfaces.

The aim of our experimental study was to determine whether there is a distinct difference in roughening behavior between the island growth mode and the step flow mode. The STM, which

turns out to be a proper technique to characterize rough surfaces [18–21], was used for analyzing Si(001) surfaces grown in island formation and step flow mode.

The Si(001) surface reconstructs by forming dimers that are arranged in parallel rows. The dimer rows can be oriented along two possible perpendicular directions, depending on the plane where the crystal is cut. A slight misorientation (smaller than $2\text{--}3^\circ$) with respect to the [001] direction in the [110] (or $\bar{1}10$) direction results in a stepped surface with two types of monatomic step edges. One type of step edge runs parallel to the dimer rows of the upper terrace and is quite straight, whereas the other type that runs perpendicular to the dimer rows of the upper terrace is rough, i.e. exhibits many thermally excited kinks [22–24].

In the case of a high deposition rate and low substrate temperature most deposited atoms meet another adatom and form an island before they reach a step edge. This island continues to grow when its size is greater than the size of a stable nucleus (in the case of Si(001) the stable nucleus is 2 atoms [25]).

If, however, the deposition rate is low enough and/or the substrate temperature is high enough the deposited atoms can easily coalesce with a step edge before meeting another adatom on the terrace. This is the so-called step flow mode, and the other growth mode described above is referred as island growth mode.

2. Experimental

The Si(001) samples we have used are 0.15° and 0.52° misoriented in the [001] direction towards the [110] direction giving rise to terrace lengths of about 500 and 150 Å, respectively. The most vicinal samples (0.52° off) were used in the case of step flow growth, whereas the others (0.15° off) were used in the case of island growth. Before loading the samples in the ultra-high vacuum (UHV) chamber with base pressure below 1×10^{-10} Torr they were ultrasonically rinsed in ethanol. The UHV chamber was equipped with an Auger apparatus (cylindrical mirror analyzer)

and a home-built Si evaporator described elsewhere (see Ref. [26]). Prior to the cleaning of the Si sample, the sample and holder were outgassed at 800–900 K for several hours. The Si samples were cleaned inside the UHV chamber by repeated cycles of ion bombardment (Ar ions, 800 eV) and annealing at 1100 K. This cleaning procedure resulted in atomically clean Si(001) 2×1 surfaces [27]. The scanning tunneling microscopy (STM) measurements were performed in air after the growth processes and rapid quenching to room temperature with a home-built STM. It must be noted, however, that the fact that these experiments were performed “after the event” on quenched surfaces in air does not seem to influence the roughness of the substrate [20].

Images of 200 by 160 points, covering areas ranging from 21 by 17 nm² up to 1068 by 854 nm², were obtained at typical tunnel currents of 1–3 nA and sample biases of 1–2 V. For vicinal Si(001) surfaces with a misorientation of 0.5° towards [110] it has been shown [28,29] that at a deposition rate of a few monolayers per minute and a substrate temperature of about 750 K epitaxial growth proceeds initially via preferential step flow growth of the rough B-type step edge, leading to the formation of a single-domain surface. During continued growth under the same conditions island growth also begins to play a role, however, step flow is still the dominant growth mode [29]. At higher substrate temperatures it is reasonable to assume that the island growth will even be more suppressed.

In order to grow Si layers in the step flow mode as well as in the island formation mode we have selected two significantly different growth conditions:

- For the Si layers grown (on 0.15° off Si(001)) in the island formation regime a substrate temperature of 600 K, a deposition rate of about 30 monolayers per minute and a total deposited amount of approximately 3000 monolayers were selected.
- For the second samples which were grown in the step flow mode (on a 0.52° off Si(001) substrate) these parameters were respectively: a substrate temperature of about 950 K, a deposition rate of about 10 monolayers per minute and a

total deposited amount of approximately 1000 monolayers.

3. Results and discussion

The height–height correlation function, see Eq. (6), is used for analyzing the STM images. For a

fixed time t the height–height correlation function depends only on r and is then defined as [1,2]:

$$G(\mathbf{r}, t) = \langle |h(\mathbf{r} + \mathbf{r}', t) - h(\mathbf{r}', t)|^2 \rangle. \quad (9)$$

If the scaling behavior as proposed in Eq. (7) is

Fig. 1. (a) STM image ($570 \text{ nm} \times 460 \text{ nm}$) of vicinal Si(001) after growing in the island growth mode. The sample bias was 2 V. (b) STM image ($420 \text{ nm} \times 350 \text{ nm}$) of vicinal Si(001) after growing in the step flow mode. The sample bias was 2 V.

satisfied the asymptotic behavior of $G(r, t)$ for a fixed growth time can easily be derived:

$$G(r, t) \begin{cases} \sim r^{2\alpha} & \text{for } r \ll \xi \\ = \text{const} & \text{for } r > \xi \end{cases} \quad (10)$$

For a fixed time t , $G(r, t)$ increases as $r^{2\alpha}$ (for small r). The spread of the fluctuations in the direction parallel to the Si(001) surface is characterized by the correlation length ξ . This shows how far the effect of a perturbation can get after some time t (growth time) along the surface. For r values which go beyond ξ the height–height correlation saturated at constant value.

The STM images of the samples grown in step flow and island formation modes are shown in Fig. 1. The surface of the sample grown in the step flow mode is relative smooth as compared to the film grown in the island formation mode. From these STM images the height–height correlation is calculated using a grid of 200 (fast scan direction) by 160 points. The tilt of the images is removed by fitting a plane through the data using the least-squares method and subtracting this plane from the data. The height–height correlation function is calculated only in the fast scan direction for r values smaller than $r = L/2$. A log–log plot of the height–height correlation function calculated from the STM images of Fig. 1 is shown in Fig. 2. The height–height correlation function for the sample grown in the step flow mode (squares) behaves as was described by Eq. (10). It shows a linear increase for small r and saturates to a constant value at large r . To determine the exponent α a best fit is made through the values in the algebraic regime. To decrease the error, α is calculated for 50 STM images taken on different places of the sample and the values are averaged resulting in $\alpha = 0.68 \pm 0.05$.

For films grown in the step flow mode we did not find the algebraic roughness regime but instead we found a more logarithmic-like behavior (logarithmic behavior is only observed over one decade). From Fig. 2, it is obvious that the behavior of the sample grown in the step flow mode is not algebraic and significantly different from the behavior shown in the island growth mode. Thus

Fig. 2. Log–log plot of the height–height correlation function $G(r, t)$ versus r . Squares: island growth mode; circles: step flow mode.

films grown in step flow mode show no scaling behavior and are not self-affine.

As already mentioned before our experimental results are in qualitative agreement with the theoretical predictions of Wolf [17]. Wolf showed that the description of growth at vicinal surfaces leads to an anisotropic generalization of the KPZ equation. With the aid of dynamical renormalization he found a sharp morphological transition from algebraic roughness in the island growth mode to logarithmic roughness in the step flow mode.

In the description of Wolf, however, a growth velocity depending on the miscut angle was necessary for the occurrence of this morphological transition. In our case such a growth velocity depending on the vicinality (a desorption rate depending on the miscut angle) is very unlikely due to the relative low substrate temperature.

Without any assumptions we therefore speculate that the morphological transition between island growth and step flow has its origin in the dimension of the system. Island growth occurs in a two-dimensional regime ($d = 2 + 1$), whereas step flow can be regarded as an effective one-dimensional ($d = 1 + 1$) growth system.

In a recent experimental STM study of step flow growth on vicinal Si(001) Wu et al. [30]

presented clear evidence for scaling behavior in $d = 1 + 1$ dimensions. The roughness and dynamical scaling exponents extracted by them are even in good agreement with predictions of a simple ballistic aggregation model in $d = 1 + 1$ dimensions.

4. Concluding remarks

In summary, a transition from algebraic roughness in the island growth mode to a non-algebraic behavior in the step flow mode is found in the case of homoepitaxial growth on vicinal Si(001).

For the island growth mode an algebraic roughness regime is observed which saturates at a constant value for large r . The roughness exponent α is estimated to be 0.68 ± 0.05 . This value is not in agreement with predictions based on numerical simulations of the Eden model [6–8], the ballistic deposition model [9–11] or the KPZ equation [4,12].

For the surface grown in the step flow mode we find a non-algebraic roughening behavior. We speculate that these observations can be understood by a difference in dimension: island growth is growth in $d = 2 + 1$ dimensions, whereas step flow growth can be regarded as growth in $d = 1 + 1$ dimensions.

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