Crossover from BKT-Rough to KPZ-Rough Surfaces for Crystal Growth/Recession

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Abstract. We found a crossover from a Berezinskii-Kosterlitz-Thouless (BKT, logarithmic)-rough surface to a Kardar-Parisi-Zhang (KPZ, algebraic)-rough surface for growing/recessing vicinal crystal surfaces in the non-equilibrium steady state using the Monte-Carlo method. We also found that the crossover point from a BKT-rough surface to a KPZ-rough surface is different from the kinetic roughening point for the (001) surface. Multilevel islands and negative islands (island-shaped holes) on the terrace formed by the two-dimensional nucleation process are found to block surface fluctuations, which contributes to making a BKT-rough surface.

1. Introduction
Surface roughness is a key property of the surface to control the growth velocity. When the surface is smooth, it grows in the manner of a two-dimensional (2D) nucleation process; whereas when the surface is rough, it grows continuously.

The surface roughness is measured by a surface width \( W(L; t) \). For a fluctuation surface, using the surface height \( h(x, y) \), the surface width is defined by

\[
\sqrt{g W(L, t)} = \sqrt{\langle [h(x, y) - \langle h(x, y) \rangle]^2 \rangle},
\]

where \( t \) is time, \( g \) is \( 1+p^2 \), \( p \) is the mean surface slope, and \( \langle \rangle \) is an ensemble average. The surface width \( W(L, t) \) for a kinetically roughened surface is known to satisfy a Family-Vicsek scaling relation [1, 2]:

\[
W(L, t) \sim L^{\alpha} f(L^{-z} t), \quad z = \alpha/\beta,
\]

where the \( \alpha \), \( \beta \), and \( z \) exponents are referred to as the roughness, growth, and dynamic exponents, respectively. In the non-equilibrium steady-state (in the limit \( t \to \infty \)), the surface width \( W \) becomes \( W(L) \sim L^\alpha \).

The surface growth equation with a non-linear term under a symmetry principle consideration was first proposed by Kardar, Parisi, and Zhang (Kardar-Parisi-Zhang, KPZ) [4]. For a 2D surface in 3D, the exponents are obtained numerically as \( \alpha = 0.3869 \), \( \beta = 0.2398 \), and
Figure 2. Top-down views and side views of the simulated surfaces at $4 \times 10^8$ MCS/site. System size: $40\sqrt{2} \times 40\sqrt{2}$. (a) and (b) BKT-rough. Number of steps $N_{\text{step}} = 8$. $p = 8/(40\sqrt{2}) \approx 0.141$. (c) KPZ-rough. $N_{\text{step}} = 64$. $p = 64/40\sqrt{2} \approx 1.131$.

$z = 1.6131$ [5, 6] (KPZ-rough surface). The values of the exponents have been observed for directed polymers, as well as other systems in the KPZ universality class.

However, for crystal growth, the observed exponents are different from the KPZ exponents [2, 7]. The reason why the exponents on the crystal growth are different from the KPZ values is not understood yet.

On the other hand, at the equilibrium, a smooth 2D surface becomes rough at a roughening transition temperature $T_R$. The universality class of the transition is Berezinskii–Kosterlitz–Thouless (BKT) class with $W^2(L)$ being logarithmically divergent ($\alpha = 0$).

In this article, the crossover from a BKT-rough surface to a KPZ-rough surface for interface-limited growth is studied using the Monte Carlo method based on the restricted solid-on-solid (RSOS) model [8, 9, 10] with a discrete Hamiltonian equivalent to the 19-vertex model.

2. RSOS Model

The adopted model to study is a restricted solid-on-solid (RSOS) model [8, 9, 10] with a discrete Hamiltonian,

$$
\mathcal{H} = \sum_{\{m,n\}} \left\{ \epsilon |h(m+1,n) - h(m,n)| + |h(m,n+1) - h(m,n)| \right\} - \Delta \mu h(m,n) + NE_{\text{surf}},
$$

where $h(m,n)$ is the height of the surface at a site $(m,n)$ (Fig. 1), $\epsilon$ is the microscopic ledge energy, $N$ is the total number of unit cells on the (001) surface, $\Delta \mu$ is a driving force for crystal growth, and $E_{\text{surf}}$ is the surface energy per unit cell. Here, “restricted” means that the surface-height difference between nearest-neighbour sites is restricted to 0, and ± 1. At equilibrium, $\Delta \mu = 0$. When $\Delta \mu > 0$, the crystal grows; whereas when $\Delta \mu < 0$, the crystal recedes.

Vicinal surfaces of a (001) surface tilted towards the $\langle 111 \rangle$ direction are considered using the Monte Carlo method for non-conserved$^1$ systems with the Metropolis algorithm. The external parameters are temperature $T$, $\Delta \mu$, number of steps $N_{\text{step}}$, and the linear size of the system $L$. The surface slope $p = N_{\text{step}}a/L = \tan \theta$, where $\theta$ is the tilt angle from the (001) direction.

$^1$ Here, “non-conserved” indicates that the number of crystals is not conserved.
Several snapshots of the vicinal surfaces obtained by the Monte Carlo method are shown in Fig. 2. It should be noted that the “desorption process” is included to reach equilibrium.

3. Driving Force and Temperature Dependence of Surface Width

![Graphs showing driving force dependence of the surface width](image)

Figure 3. Driving force dependence of the surface width [11]. (a), (b), and (c) show $gW^2/\ln L$ vs. $\Delta\mu/\epsilon$. (d), (e), and (f) show $\sqrt{gW}/L^{0.385}$ vs. $\Delta\mu/\epsilon$. (a), (c), (d), and (f): surface slope $p = 3\sqrt{2}/8 \approx 0.530$, tilt angle $\theta = 27.9^\circ$. (b) and (e): surface slope $p = \sqrt{2}/2 \approx 0.707$, tilt angle $\theta = 35.3^\circ$. Reverse triangles in (a) and (d): $\Delta\mu$ negative and $L = 80\sqrt{2 a}$ ($a = 1$).

Figure 3 shows the $|\Delta\mu|$ dependence of the scaled surface width for several temperatures. The roughening temperature of the (001) surface is $T_{R(001)} = \epsilon = 1.55 \pm 0.02$ [9, 12], whereas the roughening temperature of the (111) surface $T_{R(111)}$ is infinite. The temperature in (c) and (f) is higher than $T_{R(001)}$. When $|\Delta\mu|$ is small, the surface is BKT-rough; whereas when $|\Delta\mu|$ is large, the surface is algebraic-rough. Then, a crossover-driving-force $\Delta\mu_{co}$ is introduced. As the temperature is higher, $\Delta\mu_{co}$ becomes larger.

The size dependence of the surface width was also studied [11] to obtain the roughness exponent $\alpha$. The obtained values of exponents were all less than the value expected for the KPZ. However, the slope in the $\ln W(L)$ vs. $\ln L$ became slightly steeper as the size became larger. Hence, we concluded that the fixed point of the algebraic-rough surface is the KPZ universality class.

It should be noted that the driving force for the kinetic roughening $\Delta\mu_{kr}^{(001)}$ [13] on the (001) surface is $\Delta\mu_{kr}^{(001)}/\epsilon = 1.15$ ($k_B T/\epsilon = 0.4$) and 0.65 ($k_B T/\epsilon = 0.63$) in the present study.

In the case of crystal recession, the 2D nucleus on the (001) terrace at $\Delta\mu/\epsilon = 1$ is a negative square nucleus. Here, an ad-hole, a negative-island, and a negative-nucleus are, respectively, a vacancy on the terrace, an island made by a vacancy, and a negative-island with a critical size.
Figure 4. Slope dependence of $W$ [11]. $p = \tan \theta$. $g = 1 + p^2$. $\bar{T} = k_B T/\epsilon$. (b) and (d) $k_B T/\epsilon = 0.4$.

4. Slope Dependence of Surface Width

In Fig. 4 (a) and (c), blue and light blue marks show the width less than the roughening temperature of the (001) surface. When the angle decreases to zero, the width converges to zero because the (001) surface is smooth. The dark solid line and the light broken line are expressed by the equation:

$$\sqrt{gW} = (0.32 + 0.06 \ln p)\sqrt{\ln L}$$  \hspace{1cm} (4)

However, near the (111) surface, the marks deviate from the solid line but agree with the broken line. The dark broken line and the light solid line are expressed by the equation.

$$\sqrt{gW} = [0.233 + 0.0886 \ln(\sqrt{2} - p)]\sqrt{\ln L}.$$  \hspace{1cm} (5)

The black and pink marks show the surface width higher than the roughening temperature of the (001) surface. Hence, when the angle decreases to zero, the width has a finite value because the (001) surface is BKT-rough.

Therefore, at equilibrium, the squared $W$ depends on the surface slope as expressed by two lines based on Eqs. (4) and (5) approximately.

Fig. 4 (b) and (d) show slope dependence of the scaled surface widths in the non-equilibrium steady state. As expected, marks with different sizes agree with the broken line in the lower figure. The line is expressed by the equation.

$$\sqrt{gW} = [0.327 + 0.026 \ln(\sqrt{2} - p)]L^{0.374}$$  \hspace{1cm} (6)

Therefore, the vicinal surface with a large tilt angle and a large driving force for crystal growth is KPZ-rough.
Unexpectedly, for small angles, the marks in the upper figure are more agreeable. This indicates that the surface is the BKT-rough when the tilt angle is small. For the vicinal surface with a large tilt angle, the vicinal surface consists of the (111) terrace surface and steps with the side surface being the (001) surface (Fig. 2). However, ad-atoms and islands are prohibited on the (111) surface due to the RSOS restriction. On the other hand, for the vicinal surface with a small tilt angle, ad-atoms and islands are formed on the (001) terrace surfaces. Some parts of ad-atoms and the islands which are the same height-level as the steps enhance the step fluctuations; whereas, due to the RSOS restriction, some parts which are of different height-levels from the steps suppress the step fluctuations.

Therefore, the multi-level islands formed by 2D nucleations or thermal capillary fluctuations on the (001) surface make the surface BKT-rough. A similar change of the universality class caused by the multi-level islands is the change of the Ising class to BKT class for the roughening transition at equilibrium.

5. Discussions
When there exists point-contact-type step-step attraction between the second-nearest-neighbour lattice-sites, the elementary steps condensate at low temperatures [12, 14] at equilibrium. The assembled elementary steps form a faceted macrostep. We also studied the driving force dependence of the surface width for the vicinal surface with faceted macrosteps. In the non-equilibrium steady-state at sufficiently low temperatures, the vicinal surface grows in the polynucleation process at the edge of the faceted macrosteps. When the driving force becomes larger, the mean height of the faceted macrosteps becomes smaller [15, 16, 17]. For sufficiently large driving force, we found that the steps are well separated and become KPZ-rough surface [18] as seen in the present study.

Conclusions
For the RSOS model with a discrete Hamiltonian under a non-equilibrium steady state without surface diffusion or volume diffusion:

- There is a crossover point $\Delta \mu_{co}$ between the BKT (logarithmic)-rough surface and the KPZ (algebraic)-rough surface. The $\Delta \mu_{co}$ is different from the kinetic roughening point $\Delta \mu_{kr}$.
- A step flow growth or recession leads intrinsically to a KPZ-rough surface.
- The ad-atoms, ad-holes, and their clusters on terraces, which block the step advancement and recession, are relevant for making the BKT-rough surface.

Acknowledgement
This work was supported in part by the Collaborative Research Program of the Research Institute for Applied Mechanics, Kyushu University.

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