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A Kinetic 14-Vertex Model for \((2+1)\)D-Crystal Growth

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Abstract. — Some dynamic properties of a fourteen-vertex model defined on the bcc(001) surface are investigated with Glauber kinetics. Exact results are given for small-size versions of the model. Using Monte-Carlo simulations on large systems and the Becker-Doering law behaviour of the growth rate in the 2D-nucleation theory, a non-equilibrium phase diagram is presented.

1. Introduction

Most phenomena in nature fall in the non-equilibrium category. Accordingly, the study of non-equilibrium statistical mechanics becomes highly imperative and received much attention from physicists over the few past years. However, even the simplest phenomenon in the field remains so far not completely well understood contrarily to the equilibrium case. The steady states of these phenomena often display fascinating properties unexpected under equilibrium conditions. They thereby appear more interesting than the myriad intermediate states leading to them. They are also simpler and more often uniquely defined. Crystal growth is special in the field and has attracted much interest in recent years due to its relevance in advanced technologies. On the fundamental level, most interest arises from the fact that growth models often show a non-trivial dynamic scaling from which there emerges their universal classification in analogy with critical phenomena [1–3].

In this paper, I address a non-trivial problem of crystal growth in a lattice vertex model which can be defined on a bcc(001) surface or a fcc(110) surface. The bcc(001) surface, for instance, has been studied at equilibrium [4] and in growth [5] within the body-centered solid-on-solid (BCSOS) approximation where the height difference between neighbouring atoms is \(S = \pm a/2\), \((a\) is the lattice spacing parameter\). In the new growth model some local surface configurations where \(S\) can take the values \(\pm 3a/2\) are introduced in addition to the usual jumps \(\pm a/2\). Such modification of the BCSOS model is mainly motivated by the aim to get a more realistic description of real surfaces for growth properties. In fact, at low temperature where surface diffusion is practically frozen, some multisteps are physically possible on crystal surfaces. Therefore, a realistic model of the bcc(001) surface may, at least, allow the hollow

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site of a three-atom triangular island to be an equilibrium site. Such equilibrium site has been, for instance, observed during the Molecular Dynamics simulation of silver atoms diffusion on silver (110) at low temperature with the tight-binding model [6]. Although the consideration of the exact position of this equilibrium site on the surface violates the solid-on-solid (SOS) condition, its physical existence at low temperature suggests that double steps of height $3a/2$ are permissible in a SOS model of the bcc(001) surface.

The new growth model obeys the SOS approximation and is defined such that when two next-nearest neighbour positions are occupied in a layer, their centre is considered as an equilibrium site and may be occupied. The resulting surface is exactly mapped onto a 14-vertex model where 8 anomalous vertices are added to the well-known 6 vertices (referred to as normal vertices) of the two-dimensional version of the ice model [7]. Normal and anomalous vertices are given different energies. The equilibrium properties of the 14-vertex model have been recently studied by Barbero et al. [8] by means of transfer matrix and finite-size techniques. They found that the bcc(001) surface in the 14-vertex model undergoes a thermal roughening transition with some unusual features which emerge from the behaviour of some physical quantities such as the single and double step energies, the specific heat, etc. The smooth-rough transition line found in the parameter space presents a cusp. The whole nature of this transition line is still not very clear.

In the present work, I essentially study qualitatively the effect of an applied driving force on the thermal roughening transition of the bcc(001) surface in the 14-vertex model. The growth model appears as an extension of the kinetic 6-vertex model extensively studied in references [5, 9]. The reader should then refer to these references for more details on the description and the different approaches used in the solutions of the present model. The time evolution of the surface is described by a master equation which is solved exactly for small samples. An accurate Monte-Carlo simulation algorithm is developed to study large systems. Results on the growth velocity (at fixed driving force) as a function of the temperature $T$ and the ratio $\delta/\epsilon$ ($\delta$ is the anomalous vertex energy and $\epsilon$ denotes the 6-vertex energy) evidence some unusual features which lead to a rather complicated growth phase diagram. In fact, using the Becker-Doering law behaviour of the growth rate (at fixed driving force) in the 2-D nucleation theory [10], a qualitative phase diagram is presented on relatively large systems. It emerges that the cusp found in the thermal roughening transition line [8] is transformed into an unusual reentrant domain in the non-equilibrium phase diagram.

For relatively small values of the ratio $\delta/\epsilon$, the anomalous vertex density in the early stage of the growth is found to increase logarithmically with the deposit thickness.

In Section 2, the model and its solution are presented. Section 3 deals with results and discussions.

2. The Stochastic Model

2.1. Description of the Model. — The model is defined on the bcc(001) surface. On this surface, the height subconfigurations (a, b, c) of Figure 1 are allowed whereas those in (d, e, f) are forbidden. This holds for all subconfigurations obtained by rotating (a, b, c) and (d, e, f) by $n\pi/2$, ($n = 1, 2, 3$). The topmost atom in (d, e, f) is assumed in the present model to be in a physically unstable equilibrium position and must undergo a transient diffusion to one of the nearest equilibrium sites as defined in (a, b, c). Considering (d, e, f) as permissible may lead to a rather complicated growth model not easily tractable numerically.

Single and double steps are then present on the surface. Such surface is exactly mapped onto a 14-vertex model (see Figs. 2a, b). The mapping consists in drawing arrows on the dual lattice in such a way that the higher level atom lies in the right-hand side of the arrows [4].
The arrows with closed head is a set of three single arrows with open head. Accordingly, at each vertex, the "ice rule": number of arrows in equal number of arrows out, holds. The anomalous vertices (vertex numbers 7, . . , 14) appear whenever the height difference between the atoms at both sides of an arrow is 3 in units of a/2. The energy $e_i$ of a vertex of type $i$ is defined as: $e_i = 0$ for $i \in [5, 6]$; $e_i = \varepsilon$ for $i \in [1, 4]$ and $e_i = \delta$ for $i \in [7, 14]$. The energies $\varepsilon$ and $\delta$ are defined positive and assumed not to depend on the temperature. Accordingly, the ground state of the surface is twice degenerate. The two ground configurations are obtained by alternating vertices 5 and 6 in both in-plane directions. In the special case where all energies have the value zero, the ground state is infinitely degenerate.

To each surface configuration corresponds a 14-vertex configuration and vice-versa. Different surface configurations are parted into groups subdivided into subgroups in which configurations are related to each other via translational symmetry. The latter arises from the periodic boundary conditions applied on the vertices, which comprise also helicoidal conditions for stepped surfaces. A group of configurations is associated to a surface with well-defined Miller indices $(h, k, l)$. Each group is characterized by two time-conserved integers $Dh$ and $Dv$. If $n_1/n_2$ denotes the number of arrows pointing to the West/East in a row of vertices and $m_1/m_2$ the number of arrows pointing to the North/South direction in any column, $Dh = n_1 - n_2$ and $Dv = m_1 - m_2$. These numbers give the level difference between (vertical and horizontal) opposite sides of the surface. They are related to the Miller indices as defined in reference [5]. For the system of size $N = 4$, the group (0, 0) comprises 916 subgroups;
(0, 2) has 449 subgroups; (0, 8) has only one subgroup which contains two passive configurations (no evolution site). The group (−2, 2) contains 156 subgroups and (2, 4) has 14 subgroups. In general, all groups having the same |DH| and |DV| are related via rotation (of angle π/2) or reflection symmetry and have the same time evolution. The present study will be essentially restricted to the group (0, 0). The classification of possible surface configurations into groups has been widely used in the literature in similar context [9, 11, 12].

In equilibrium as in dynamics, a statistical time-dependent weight \( P(\sigma, t) \) is associated to each 14-vertex configuration. The dynamics is specified when one fixes the transition rates \( W(\sigma, \sigma') \) from a configuration \( \sigma \) to a configuration \( \sigma' \) through a kinetic equation:

\[
\frac{dP(\sigma, t)}{dt} = \sum_{\sigma'} (-W(\sigma, \sigma')P(\sigma, t) + W(\sigma', \sigma)P(\sigma', t)).
\]

This relation expresses that the rate of change of \( P(\sigma, t) \) is given by the difference between the flux into \( \sigma \) from other configurations \( \sigma' \) and the flux out of \( \sigma \) to others. These transitions occur in the present model by atom deposition and evaporation on the crystal surface considered as Markov processes. Each transition changes the surface 14-vertex configuration on four vertices which are used to define growth/evaporation kink subconfigurations (see [5, 9]). The transition rates follow Glauber kinetics [13] and are defined as in references [5, 9]:

\[
C^a = \exp(\Delta \mu/k_B T)/(1 + \exp(\Delta E/k_B T))
\]

for deposition and

\[
E^a = 1/(1 + \exp(\Delta E/k_B T))
\]

for evaporation, where \( \Delta \mu \) represents the driving force applied to the surface, \( \Delta E \) the change in the surface 14-vertex energy during the evolution of the kink of type \( (a) \). The factor \( 1/k_B T \) describes the coupling of the system to a heat bath at the temperature \( T \). One can check that these rates satisfy a detailed balance condition with respect to the 14-vertex energy at the considered temperature. In the absence of net growth, the steady state distribution may correspond to the Boltzmann distribution. The above rates and the master equation (1) completely specify the dynamics of the system at the “microscopic level”.

2.2. Exact Results on Small Samples. — The same method has been considered in references [9, 11, 12]. Configurations of a table \( N \times N \) (of size \( N \)) are classified into groups \( S_i \) \( (i \) is the index of the group) subdivided into subgroups \( S^m_i \) \( (m \) is the index of the subgroup). During growth, the surface configuration runs from one subgroup to another within the same group. This results from the ergodic nature of the growth process within a group and enables one to determine a transition matrix between subgroups. The weight of the subgroup \( S^k_i \) is defined at time \( t \) as:

\[
P^k_i(t) = \sum_{\sigma \in S^k_i} P(\sigma, t).
\]

For a given active group \( i \), the eigenvectors \( P^k_i(\ast) \) corresponding to the eigenvalue 0 are used to compute physical quantities in the steady state, \( e.g. \) the growth rate per surface site for the group \( i \) defined by:

\[
G(i) = (1/N^2) \sum_{\alpha,k} (C^a p^a(i,k) - E^a q^a(i,k)) P^k_i(\ast)
\]

where \( p^a(i,k)/q^a(i,k) \) denotes the multiplicity of the growth/evaporation kink of type \( (a) \) in \( S^k_i \).
2.3. Monte-Carlo Simulation. — The simulation appears easier than the difficult problem of subgroup classification. It always starts from a flat configuration. At each Monte-Carlo step, all growth and evaporation kinks are listed. The total condensation rate $S_1(\sigma, t)$ and evaporation rate $S_2(\sigma, t)$ are calculated and their sum $S(\sigma, t)$ evaluated. Then an active kink is randomly selected and its evolution accepted with the probability $C^a/S(\sigma, t)$ for condensation and $E^a/S(\sigma, t)$ for evaporation of the kink of type $(a)$. Actually, this algorithm is not very efficient; however, it gives very accurate results in agreement with exact calculations up to several digits. The growth rate is calculated using the real time averaging procedure described in references [5,9,11,12]. For small values of $\delta/\epsilon$, the number of Monte-Carlo steps needed to reach the steady state strongly increases. For $N = 32$, this number is about $6 \times 10^5$.

3. Results and Discussions

The present model displays several non-trivial behaviours when one attempts to study the growth kinetics for all values of the ratio $\delta/\epsilon$. The case $\delta/\epsilon \leq 1$ is interesting from the statistical mechanics point of view, although it appears somewhat non-physical. As in reference [5], finite-size effects are found to be very weak for all growth parameters when the system size $N$ is larger than 30. At large values of $\delta/\epsilon$, the anomalous vertex density is very small on the growing surface and the model tends to the kinetic 6-vertex model whose properties are somewhat known numerically [5,9]. In the following results, the temperature and the driving force are given in units of the thermal roughening transition temperature of the BCSOS model $(k_B T_c = \epsilon/\log(2))$.

The master equation (1) is solved exactly for $N = 4$, group $(0, 0)$. This involves a transition matrix of size $916 \times 916$ between subgroups. In Figures 3a, b, the exact growth rates are given for $N = 4$ at fixed driving force $\Delta \mu = 1.5$. Although the system size is quite small, some basic features of the growth model are found. In Figure 3a, the thermal evolution of the growth rate for several values of $\delta/\epsilon$ is displayed. Simulation results for $N = 4$, group $(0, 0)$ are compared with the exact calculations and an agreement is found up to 3 or 4 digits.

The growth rate shows a maximum at a critical value $T_c(\delta, \Delta \mu)$ after which it decreases. For varying $\delta$ at fixed temperatures, another maximum of the growth rate is found. One interesting feature is that with increasing $\delta/\epsilon$, the temperature $T_c$ decreases, then increases and further decreases again. One might therefore expect to obtain a non-trivial behaviour of the transition line between different growth modes: layer-by-layer and continuous. From Figures 3a, b, one realizes that the maximum growth rate in the present model is reached for $\delta/\epsilon \approx 0.9$ around $T = 1$. Support to this general result is found from the investigation of other systems. However, the existence of physical systems corresponding to such ratio $\delta/\epsilon$ is questionable. While exact results do give some insight into the real features of the model, they are however of little value for the understanding of the large scale properties of the model. In Figure 3c, the simulated growth rates are given for $N = 32$, group $(0, 0)$ and $\Delta \mu = 1.5$. The same trend of the steady growth rate $G$ found in the last two figures is recovered. The increasing part of $G$ may be associated to a layer growth domain according to Becker-Doering’s law [10]. In this framework, the growth rate mainly behaves as $\exp(-\beta E_0/3\Delta \mu)$, where $E_0$ is the step free energy per unit length which is temperature-dependent and $\beta$ denotes the inverse temperature $1/k_B T$. Beyond that region, the growth may become continuous, following the Wilson-Frenkel law [14]. When the exponential prefactor (Zeldovich factor) $(\beta \Delta \mu)^\gamma$ with $\gamma \approx 1$ is considered, one actually finds that only a part of the increasing region of $G$ does correspond to a decrease of the step energy $E_0$ when the temperature is raised. However, the dominant exponential factor is sufficient to qualitatively describe the change in the growth modes. Using $T_c$ as a transition temperature to the continuous mode at fixed $\Delta \mu$, two non-equilibrium
Fig. 3. — In a), the temperature dependence of the exact (full lines) and the simulated (open circles) growth rates are displayed. Numbers given on different curves correspond to the values of $\delta/\epsilon$. The system considered is $N = 4$, group $(0, 0)$. Simulation results have been averaged over two independent runs. In b), the exact growth rate is plotted against $\delta/\epsilon$. Numbers on the curves give the values of the temperatures considered. In c), the simulated growth rates are given for $N = 32$, group $(0, 0)$. The different values of $\delta/\epsilon$ used in the calculations are those displayed on the curves.

Fig. 4. — Non-equilibrium phase diagram for $N = 16$, group $(0, 0)$ at two values of the driving force $\Delta \mu$ (a); $\beta$ is the inverse temperature $1/k_B T$. In b), the same phase diagram is displayed for $N = 32$, group $(0, 0)$.

transition lines are given in Figure 4a at two driving forces for a system of size $N = 16$. The fundamental remark which emerges is that the transition line for $\Delta \mu = 1$ has the same form as the smooth-rough transition line found for $\Delta \mu = 0$ [8], except that here, the cusp found at equilibrium is transformed into a reentrant region in the non-equilibrium phase diagram. With increasing driving force at fixed $N$, the rough phase domain where the continuous growth prevails advances in the layer growth region. One might expect the whole layer growth domain to disappear when the driving force will go to infinity at any ratio $\delta/\epsilon$. In fact, at large driving
force or high temperature, the active kink energies become somewhat irrelevant. Hence, all kinks are almost equally eligible for evolution. Particles will land anywhere on the surface provided the height difference restriction on neighbouring sites allowed in Figures 1a, b, c holds. This situation of course will lead to a rough surface. In Figure 4b, another phase diagram is given. Similarly to what happened above, the increase of the system size $N$ plays the same effect as the driving force, with the exception that, here, a layer growth is expected for a system of infinite size. Due to the fact that finite-size effects are small for $N = 32$,
the transition line in the thermodynamic limit will not be qualitatively different from the one displayed in that figure when the same driving force is considered. One should remember that any growing surface is rough at least logarithmically. The surface smoothness discussed here is related to the possibility of layer growth of the surface. In the layer growth region, the surface may grow by the periodic formation of 2D-supercritical nuclei [10].

In the right part of the phase diagram where the transition line is almost horizontal, the model is equivalent to the kinetic 6-vertex model [5]. In fact, the anomalous vertices are given higher energy and become exceedingly rare on the surface. In the left part where $\delta/\epsilon$ is much less than unity, anomalous vertices proliferate on the surface. This, due to the height difference constraint on neighbouring sites, plays locally some pinning effect on the surface. Then, slow growth and an almost flat surface result. Around the line $\delta/\epsilon = 0.9$, normal and anomalous vertices are almost equally probable on the surface. Accordingly, particles can land on any growth site. Such situation leads to the roughening of the surface along this line, at least up to some critical value of $\beta \epsilon$ which depends on the driving force and the system size. This physically justifies the unusual feature of the phase diagram along this line.

A direct investigation of the surface morphology at three different points of the phase diagram is performed. In the rough phase, the surface is found to be really rough (see Fig. 5a). The black blots observed correspond to holes on the surface. They are numerous and sparse. In the left part of the smooth phase, one can realize from Figure 5b that the surface is not really flat. Large flat regions separated by deep holes are found. Such result is somewhat expected. In fact, in this region at equilibrium [8], a DisOrdered Flat (DOF) domain is found and the real nature of the smooth-rough transition is still not very clear. In the right part of the layer growth region, the growing surface is found to be very smooth. In Figure 5c, a typical surface configuration is given. For the physical parameters considered, the surface is growing in a one-cluster mode: only one big island is evolving on the surface.

In Figure 6, the anomalous vertex density $d$ is investigated at large supersaturation ($\Delta \mu/k_B T = 10$). $d$ increases and rapidly saturates. For relatively small values of $\delta/\epsilon$, the increase is almost logarithmic in the time (taken here as the surface thickness).
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