Universal Behavior of the Equilibrium Crystal Shape near the Facet Edge.

I. A Generalized Terrace-Step-Kink Model

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A generalized version of the terrace-step-kink (TSK) model is introduced to study in detail the shape of a crystal near the facet edge. The model is analyzed by the transfer-matrix method combined with the lattice fermion approach. As a result, behavior of the curvature near the facet edge is found to have unexpected new features: finite Gaussian curvature jump with universal amplitude and vanishing transverse curvature with universal exponent.

§1. Introduction

The equilibrium crystal shape (ECS) depends on the temperature \( T \). At high temperatures above the roughening temperature \( T_r \), there is no facet (flat plane) and we have a rounded ECS. On the contrary, at \( T = 0 \), the ECS is enclosed only by facets (flat planes). In the intermediate range of temperature \( 0 < T < T_r \), the ECS is composed of both facets and curved areas. In the recent development of the study of the ECS, the central issues have been the critical behavior of ECS: 1) the faceting transition at \( T_f \) accompanying a finite curvature jump, and 2) the Gruber-Mullins-Pokrovsky-Talapov (GMPT) type behavior (3/2 th-power law) of the ECS profile near the facet edge at \( T < T_r \) which has been known in the theory of commensurate-incommensurate transitions in physisorbed system. Important point is that the above behaviors 1) and 2) are expected to be 'universal'; the amount of curvature jump at \( T_r \), after suitably normalized, is the same for a wide class of models and so is the value of the GMPT exponent 3/2. This universal nature, although yet to be verified experimentally, gives the study of the ECS much interest and importance. In our view, the present status of understanding about the thermal behavior of the ECS including the above-mentioned critical behavior is far from satisfactory. This situation can be improved by detailed studies on various aspects of the problem.

The purpose of this series of papers is to study the ECS in the intermediate temperature range \( 0 < T < T_r \) in more detail, focusing attention on the curvature of the ECS near the facet edge. In this paper, the first of the series, we introduce a generalized terrace-step-kink (TSK) model to discuss the interface properties for general interface orientation. In contrast to the former treatments by use of the 'original' TSK model, where only the profile of the ECS along a single direction is drawn, present model and its solution allow us to obtain the ECS as a two-variable function \( z = z(x, y) \) of the coordinates \( x \) and \( y \). In particular, we can calculate full curvature tensor, not just its single component, which is indispensable to discuss the geometry of the ECS.

We adopt the transfer matrix method called the lattice fermion method to analyze the generalized TSK model. The lattice fermion method was first proposed by Izuyama and Akutsu for the statistical mechanics of biomembranes. Afterward, this method was applied to the discussion of the commensurate-incommensurate transitions.

The paper is organized as follows. In §2, the
generalized TSK model and its solution technique are explained. In §3, the solution of the model is given. The procedure to obtain the ECS from the solution is also presented. In §4, the behavior of the curvature near the facet edge is investigated in detail. New features (finite jump of Gaussian curvature, singular behavior of the 'transverse' curvature) of the critical phenomena at the facet edge are found. The last section is devoted to summary of this paper.

§2. Generalized TSK Model and Transfer Matrix Method

We consider a simple cubic crystal below the roughening temperature where the crystal shape is made up of both facets and curved surfaces. We set up a Cartesian coordinate system such that the \(x-y\) plane is parallel to the \((0, 0, 1)\) facet and the origin is at the center of the facet. For convenience, \(x\) and \(y\) directions are chosen to be the crystal axes. Then the crystal shape near the facet is described as \(z = z(x, y)\). Determination of the equilibrium crystal shape (ECS) requires the knowledge of orientation-dependent interface tension. This can be achieved by calculation of the specific interface free energy of a system with fixed average gradients \(p_x(=\partial z/\partial x)\) and \(p_y(=\partial z/\partial y)\). Since the analysis based on the original TSK model is restricted to the case \(p_r=0\), we introduce a generalized TSK model which enables us to discuss the interface properties for general interface orientation with \(p_t \neq 0\).

In the TSK picture of the two-dimensional interface, microscopic configuration of a tilted surface near the \((0, 0, 1)\) facet is described in terms of terraces parallel to the facet and the steps connecting these terraces. The steps are infinitely long and running in some average direction. The step density and the step direction angle determine the average interface orientation. Namely, denoting the unit height of the step (=atomic distance of the crystal) by \(d\), the step density by \(\rho\), and the step direction angle relative to the \(y\)-direction by \(\theta\), we have \(\rho_x = -d \cdot \rho \cdot \cos \theta\) and \(\rho_y = -d \cdot \rho \cdot \sin \theta\). In what follows, we shall only consider an interface tilted in the neighborhood of the \(x\)-direction with \(\rho_x \leq 0\) and \(|\theta| \leq \pi/4\). Then the step we consider is the down step connecting the lower terrace at larger \(x\) to the higher one at the smaller \(x\) (Fig. 1).

The partition function of the system is calculated as the weighted sum over possible step wanderings. We restrict the configuration space such that the steps do not overlap with each other. In the lattice fermion method, we start from the random walker representation of steps. By regarding the wandering of a step as a space-time path of a random walker, our system is converted to that of non-overlapping multiple random walkers. To simplify the treatment, we take into account the kinks with smallest length (=the atomic distance \(a\) of the crystal). Then we have two types of kinks, the right kink and the left kink. The right kink (resp. left kink) connects the step at smaller (resp. larger) \(y\) to the right neighbor step at larger (resp. smaller) \(y\) (Figs. 2 and 3). Then the surface slopes in the \(x\) direction and the \(y\) direction are obtained from the step densities and the densities of both types of kinks. In the original TSK model, the densities of two types

![Fig. 1. Terraces and down steps with kinks. The configuration of the steps is regarded as the space time trajectories of the random walkers on the one-dimensional lattice. The \(y\) axis corresponds to the time axis.](image1)

![Fig. 2. The right kink. The right kink corresponds to the movement of the random walker with \(\tau = 1\).](image2)
of kinks are equal hence the surface normal is always in the $x-z$ plane. The generalized TSK model, which we introduce here, allows us to investigate the surface with an arbitrary normal near $(0, 0, 1)$ direction.

For convenience, we set the lattice constant to be unity. The two-dimensional system size is chosen to be $N$ $(x$-direction) $\times M$ $(y$-direction). In the random walk picture, $y$-coordinate is regarded as the time which is discrete, and $x$-coordinate represents the position of the random walkers in one-dimensional lattice. The number of the steps is denoted by $n$. The number of the random walkers is also $n$ at any time step.

In order to count the number of the right kinks and of the left kinks, we introduce three 'orbitals' on each lattice site$^{37,38}$ according to the idea of Izuyama.$^{40}$ The orbital state is denoted by $\tau$ with $\tau = 1, 0, -1$. At any time step, the $j$th random walker ($j = 1, \cdots, n$) occupies an orbital state $\tau_j$ on the lattice site $x_j$. The random walker with $\tau = 0$ remains on the same site at the next time (see Fig. 4). This costs an energy $\delta$ which is the excitation energy of a step per unit bond length in the $y$-direction. The random walker occupying the orbital state $\tau = 1$ on the site $x$ moves to the right neighbor site $x+1$ at the next time. The trajectory of this random walker corresponds to two steps connected by a right kink. The random walker with $\tau = -1$ moves to the left neighbor site $x-1$. The trajectory of this random walker corresponds to a step with a left kink. See Figs. 2 and 3. At one time step, the random walker with $\tau = \pm 1$ costs an energy $\delta + e$. The extra energy $e$ is the excitation energy for a kink formation. The above three 'moves' are the basic elements of each step.

In the lattice fermion approach, essential information is contained in the one-particle transfer matrix $T(j)$ acting on the space of $j$th particle$^{39,40}$. It is defined by$^{37,38}$

$$ T(j) |(x_j; \tau_j); j> = \xi_j \sum_{\tau_j} |(x_j + \tau_j; \tau_j); j>,$$  

(2.1)

where $|(x_j; \tau_j); j>$ is a state vector for the $j$th particle occupying the orbital $\tau_j$ at site $x_j$. In the above, $\xi_j$ is defined by

$$ \xi_j = \exp \left[ \beta (\mu + \mu_+ - \delta - e) \right],$$  

(2.2a)

for $\tau = \pm 1$, and

$$ \xi_0 = \exp \left[ \beta (\mu - \delta) \right],$$  

(2.2b)

with $\beta = (k_B T)^{-1}$. Three quantities $\mu$, $\mu_+$, and $\mu_-$ are the chemical potentials for the steps ($\mu$), for the right kinks ($\mu_+$), and for the left kinks ($\mu_-$). The eigenvalue $A_k$ for $T(j)$ is found to be

$$ A_k = 0,$$  

(2.3a)

or

$$ A_k = \sum_{\tau} \xi_j e^{i k \tau}.$$  

(2.3b)

For $A_k = \sum_{\tau} \xi_j e^{i k \tau}$, the eigenstate is given by

$$ |k; j> = \frac{1}{\sqrt{3}} \sum_{\tau} |(k; \tau); j>,$$  

(2.4)

where we define the 'Bloch orbital' $|k; \tau); j>$ for the $j$th random walker by

$$ |(k; \tau); j> = \frac{1}{\sqrt{N}} \sum_{x=0}^{N-1} e^{-i k x} |(x; \tau); j>.$$  

In the above $k = (2\pi v/N)$ with $v$ being the in-
teger satisfying $-N/2 < \nu \leq N/2$. The other eigenvalue $\Lambda_k = 0$ does not contribute to the partition function. Thus, we can confine ourselves to the vector space spanned by the r.h.s. of (2.4). From now on, $\Lambda_k$ and $\langle k ; j \rangle$ represent the r.h.s. of (2.3b) and of (2.4), respectively.

We take the fermion representation to evaluate the eigenvalues of the transfer matrix. Note that the non-overlapping condition of $n$ random walkers is satisfied when we choose a totally antisymmetric state $\Psi_\nu(n)$ which is a linear combination of the states $\{ \prod_{\nu=1}^{N} | k_{\nu} ; j_{\nu} \rangle \}$.\textsuperscript{39,40} We denote the vector space spanned by the antisymmetric states $\{ \Psi_\nu(n) \}$ by $V_\nu(n)$ which is isomorphic to a space of fermions and is called fermion space. The lattice fermion approach is the one where we seek the eigenvectors in the fermion space $V_\nu(n)$, which has been known to work well for the problems with non-overlapping conditions in two dimensions.\textsuperscript{36,39,40}

We denote the fermion operator which creates (resp. annihilates) the state $\langle k \rangle$ by $\alpha_k$ (resp. $\alpha_k^\dagger$). We denote the ‘initial state’ (resp. ‘final state’) for $n$ random walkers by $| (x_1 ; \tau_1) , \cdots , (x_n ; \tau_n) \rangle$ (resp. $| (x'_1 ; \tau'_1) , \cdots , (x'_n ; \tau'_n) \rangle$). Then, the grand partition function is rewritten as

$$\Xi = \sum_{n=1}^N \frac{1}{n!} \left[ \sum_{(x_1 ; \tau_1), \cdots , (x_n ; \tau_n)} \langle (x_1 ; \tau_1) , \cdots , (x_n ; \tau_n) | \right]$$

$$\times A^n | (x_1 ; \tau_1) , \cdots , (x_n ; \tau_n) \rangle,$$

where the transfer matrix $A$ is given by\textsuperscript{38-40}

$$A = \prod_{x \in \{1\}} | \Lambda_x |^{\alpha_x \alpha_{x'}},$$

and the summation is taken over all possible initial and final states. The thermodynamic potential $f$ is obtained from the grand partition function (2.5) as

$$f(\mu, \mu_{+1, \mu_{-1}}, T) = -k_B T \lim_{N,M \to \infty} \frac{1}{NM} \ln \Xi.$$

(2.7) and the crystal shape $z = z(x, y)$. It is easy to see that the following relations hold:

$$\frac{\partial f}{\partial \mu} = -\tilde{\rho},$$

$$\frac{\partial f}{\partial \mu_{+1}} = -\tilde{\sigma}_R,$$

$$\frac{\partial f}{\partial \mu_{-1}} = -\tilde{\sigma}_L.$$ 

(3.1)

In the above, $\tilde{\rho}$, $\tilde{\sigma}_R$ and $\tilde{\sigma}_L$ are the equilibrium densities of the steps, the right kinks and the left kinks, respectively. The quantities $\tilde{\rho}$ and $\tilde{\sigma}_L - \tilde{\sigma}_R$ directly determine the slopes $p_x$ and $p_y$ as

$$p_x = -\tilde{\rho},$$

$$p_y = -(\tilde{\sigma}_L - \tilde{\sigma}_R).$$

(3.2)

From (3.1) and (3.2), we have

$$\frac{\partial f}{\partial \mu} = p_x,$$

$$\frac{\partial f}{\partial \mu_{+1}} - \frac{\partial f}{\partial \mu_{-1}} = p_y.$$ 

(3.3)

Since the slopes $p_x$ and $p_y$ are defined as $p_x = \partial z / \partial x$ and $p_y = \partial z / \partial y$, respectively, eq. (3.3) are rewritten as

$$\frac{\partial f}{\partial \mu} = \frac{\partial z}{\partial x},$$

$$\frac{\partial f}{\partial \mu_{+1}} - \frac{\partial f}{\partial \mu_{-1}} = \frac{\partial z}{\partial y}.$$ 

(3.4)

The solution of (3.4) which fits with our choice of the coordinate system is given by

$$z = \frac{1}{\lambda} f,$$

$$x = \frac{1}{\lambda} \mu,$$

$$y = \frac{1}{\lambda} \mu_{+1} = \frac{1}{\lambda} \mu_{-1},$$

(3.5)

where $\lambda$ is a constant. Thus, the crystal shape $z(x, y)$ is given by

$$z(x, y) = \frac{1}{\lambda} f(\lambda x, -\lambda y, \lambda y, T).$$

(3.6)

The constant $\lambda$ is related to the volume of the crystal but does not affect the shape of the crystal, which corresponds to the Lagrange

§3. Calculation of the Equilibrium Crystal Shape from the Generalized TSK Model

Before writing down the solution of the generalized TSK model, we mention the relation between the thermodynamic potential $f$ in...
multiplier in the Andreev's method. We then define the scaled variables \( X, Y \) and \( Z \) by
\[
X = \lambda x, \\
Y = \lambda y, \\
Z = \lambda z.
\]
In terms of the scaled variables the crystal shape is given by
\[
Z(X, Y) = f(X, -Y, Y, T). \quad (3.6)'
\]
Let us now proceed to calculate the thermodynamic potential from the largest eigenvalue of the transfer matrix \( A \). In general, the eigenvalue of the transfer matrix \( A \) is complex. However, for a fixed number of steps \( n \), the eigenvalue \( E_0(n) \) which has the largest absolute value is a positive real number due to the Perron-Frobenius theorem. The thermodynamic potential is obtained as
\[
f(\mu, \mu_+1, \mu_1, T) = -k_B T \lim_{N,M \to \infty} \frac{1}{NM} \ln Z \\
= \min_{\rho} \tilde{f}(\rho), \quad (3.8)
\]
where \( \rho \) is the step density \( n/N \) and the function \( \tilde{f}(\rho) \) is defined by
\[
\tilde{f}(\rho) = -k_B T \lim_{N \to \infty} \frac{1}{N} \ln E_0(n). \quad (3.9)
\]
In order to obtain the thermodynamic potential, we should calculate the largest eigenvalue \( E_0(n) \) of the transfer matrix \( A \) for a fixed step density \( \rho \).

From eqs. (2.2) and (3.5), the Boltzmann weights \( \xi_0 \), \( \xi_1 \) and \( \xi_{-1} \) are expressed as
\[
\xi_0 = \exp \{ \beta(X - \delta) \}, \\
\xi_1 = \xi_0 e^{-\beta Y} (\xi = e^{-\beta \rho}), \\
\xi_{-1} = \xi_0 e^{\beta Y}. \quad (3.10)
\]
The eigenvalue \( A_k \) for \( T(j) \) is written as
\[
A_k = \xi_0 (1 + 2 \zeta \cosh \omega), \quad (3.11)
\]
where
\[
\omega = \omega_k(Y) = \beta Y - ik. \quad (3.12)
\]
Then we find
\[
A_k^* = A_{-k},
\]
and
\[
|A_k|^2 = \xi_0^2 + \xi_1^2 + \xi_{-1}^2 + 2 \xi_0 (\xi_1 + \xi_{-1}) \\
\times \cos k + 2 \xi_1 \xi_{-1} \cos 2k.
\]
It is easy to see that \( |A_k|^2 \) has the maximum at \( k = 0 \). For the purpose of studying the ECS near the facet edge, only the \( \hat{\rho} = 0 \) property of the system is necessary. Thus, the eigenstate \( |\Phi_0(n)\rangle \) corresponding to the eigenvalue \( E_0(n) \) is constructed by making the fermions occupy the states around \( k = 0 \):
\[
|\Phi_0(n)\rangle = \prod_{|k| \leq k_f} \alpha_k |0\rangle,
\]
where \( k_f = \pi \rho \) and the vacuum state \( |0\rangle \) is defined by \( \alpha_k |0\rangle = 0 \) (for all \( k \)). Then the largest eigenvalue \( E_0(n) \) is given by
\[
E_0(n) = \prod_{|k| \leq k_f} A_k = \prod_{0 \leq k \leq k_f} |A_k|^2. \quad (3.13)
\]
From (3.11) and (3.13), the function \( \tilde{f}(\rho) \) is obtained as
\[
\tilde{f}(\rho) = (\delta - X) \rho - \frac{k_B T}{2\pi} \int_0^{k_f} \left[ R(\omega_k(Y)) \right] dk,
\]
with
\[
R(\omega) = \frac{A_k}{\zeta_0} = \ln (1 + 2 \zeta \cosh \omega). \quad (3.15)
\]
Using the result (3.14), we obtain the equilibrium crystal shape through (3.6)' and (3.8).

\section{Curvature near the Facet Edge}

When \( \rho = 0 \), \( \tilde{f}(\rho) \) given by (3.14) is expanded as
\[
\tilde{f}(\rho) = A \rho + B \rho^3 + \ldots, \quad (4.1)
\]
where the coefficients are given by
\[
A = A(X, Y) = \delta - X - k_B T R(\omega(Y)), \quad (4.2)
\]
and
\[
B = B(Y) = \frac{k_B T}{12} \frac{\partial^2}{\partial k^2} [R(\omega) + R(\omega^*)]_{k=0} = \frac{k_B T}{6} \pi^2 R''(\beta Y). \quad (4.3)
\]
From (3.6)', (3.8) and (4.1), the crystal shape is given by
\[
Z(X, Y) = \lim_{\rho \to 0} \tilde{f}(\rho) = \lim_{\rho \to 0} (A \rho + B \rho^3). \quad (4.4)
\]
When $A(X, Y)>0$, the minimum is attained at $\rho=\bar{\rho}=0$ with $Z=0$. That is, the region $A(X, Y)>0$ corresponds to a facet. When $A(X, Y)<0$, we have $\bar{\rho}>0$ and the crystal surface is curved. The contour of the facet is given by $A(X, Y)=0$. Namely, we have the equation which describes the facet contour:

$$X = X_c(Y),$$

where

$$X_c(Y) = \delta - k_b^a TR(\omega_0(Y)) = \delta - k_b^a T \ln (1 + 2\zeta \cosh \beta Y).$$

(4.6)

We should note here that since we neglect the kinks whose lengths are larger than 1, equation (4.6) is correct in the region $|Y| = 0$. From (4.4), we obtain the crystal shape near the facet contour:

$$Z(X, Y) = -\frac{2}{3\sqrt{3}} \frac{1}{B(Y)} (X - X_c(Y))^{3/2},$$

$$Z(X, Y) = 0, \quad (X \geq X_c(Y)) \quad (0 \leq X \leq X_c(Y))$$

(4.7)

Near the facet edge $(X_c(0), 0)$, the curvature in the $x$-direction is given by

$$K_x = \frac{\partial^2 Z(X, Y)}{\partial X^2} \bigg|_{X = X_c(0), Y = 0} = \frac{1}{2\sqrt{3}} \frac{1}{B(0)} (X - X_c(0))^{-1/2},$$

(4.8)

which shows the well-known Gruber-Mullins-Pokrovsky-Talapov singularity. The curvature in the $y$ direction is obtained as

$$K_y = \frac{\partial^2 Z(X, Y)}{\partial Y^2} \bigg|_{X = X_c(0), Y = 0} = \frac{1}{3} \frac{1}{B(0)} X_c''(0)(X - X_c(0))^{1/2}.$$

(4.9)

In contrast to $K_x$, $K_y$ approaches zero as $X \to X_c(0)$. This vanishing property of the 'transverse' curvature and its characteristic exponent $1/2$ have not been discussed in literature.

From (4.8) and (4.9), it is clear that the product $K_x K_y$ remains finite in approaching the facet edge. Noting that the $x$-direction and the $y$-direction are the principal axes of the curvature tensor near the facet edge, the product $K_x K_y$ is the Gaussian curvature $K$ of the surface. Thus we see that the Gaussian curvature of the curved area near the facet edge $(X_c(0), 0)$ is given by

$$K = K_x K_y = \frac{1}{6} \frac{X_c''(0)}{B(0)} = \frac{\beta^2}{\pi^2}.$$  

(4.10)

Since $K = 0$ on the facet, we have found a finite jump of the Gaussian curvature at the facet edge.

Furthermore, we can show that the amount of the curvature jump $\beta^2/\pi^2$ is the same at any point on the facet contour. Let us note that the Gaussian curvature is defined as the determinant of the curvature tensor. This leads to the following expression of the Gaussian curvature $K$ near the facet edge:

$$K = \beta^2 \pi^2 (X - X_c(Y) + 0).$$

(4.15)

As can be seen from the above derivation, the equations (4.12)-(4.14) are essential. We should remark here that in these equations the details of the model do not appear. In fact, whenever the eigenvalue $\lambda_i$ is a function of $\omega = \beta Y - ik$, the relations (4.13) and (4.14) are satisfied. For example, in more generalized models where the excitation of long kinks are
permitted, the eigenvalue \( \Lambda \) is also a function of \( \omega \) if the excitation energy of the kink is proportional to its length.

\section{Summary}

In this paper, we have investigated the equilibrium crystal shape (ECS) by use of the generalized terrace-step-kink (TSK) model which enables us to discuss the detailed property of the ECS below the roughening transition temperature. We have focused the analysis on the property of the ECS near the facet edge. As a result we find unexpected new features in the critical behavior of the ECS near the facet edge. Namely, the finite jump of the Gaussian curvature

\[
K = \begin{cases} 
\frac{\beta^2}{\pi^2}, & \text{on the curved surface}, \\
0, & \text{on the facet},
\end{cases}
\]

and the behavior of the ‘transverse’ curvature

\[
K_T \sim (X - X_c)^{1/2}(X \to X_c, X_c; \text{facet edge}),
\]

are found. Important point is that the precise value \( \beta^2/\pi^2 \) of the curvature jump is the same at all points on the facet contour (and so is the exponent 1/2 of the transverse curvature). As was noted at the end of §4, the fact that these features do not depend on the details of the present model suggests the universal nature of these behaviors. In fact, we have verified these features also in the BCSOS model\(^{41}\) and have found a general argument for their universality. In the next paper of the series, we will give full account on these points.

It is interesting to compare the value of the Gaussian curvature jump with that of the well-known universal curvature jump\(^{37}\) at \( T_R \): \[
\Delta(k_B T \sqrt{K}) = \frac{1}{\pi} \quad \text{(at the facet edge)},
\]

\[
\Delta(k_B T_R \sqrt{K}) = \frac{2}{\pi} \quad \text{(at } T_R \text{)}.
\]

We see ‘by a factor 2’ difference between the two. It is then natural to expect that these two behaviors can be ‘unified’. A more generalized model including up steps and step loops as well as down steps may be suited for this purpose. In such a model, two kinds of fermions are necessary to describe both the down steps and the up steps. Furthermore, we should admit the ‘pair annihilation’\(^{36}\) of the fermions to take into account the step loops. In spite of difficulty caused by these generalizations, certain aspects of the model can be analyzed. Details will be published elsewhere.

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