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# The morphology of crystalline surfaces in the presence of attractive step interactions 

Metin Ozdemir $\dagger$<br>Department of Physics, University of Cukurova, 01330 Adana, Turkey

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#### Abstract

We study the equilibration of an initial surface misoriented relative to a high-symmetry (low-energy) direction. The surface considered consists of parallel monatomic ledges separated by terraces. Both repulsive and attractive interactions between step pairs are taken into account. Repulsive interactions between steps are assumed to vary as $\ell^{-2}$ and attractive interactions to vary as $\ell^{-1}$ where $\ell$ is the average step separation between neighbouring steps. Attractive interactions lead to step bunching, and as a consequence the resulting morphology is in the form of macro-steps separated by large flat terraces. The time dependence of flat parts that separates the macro-steps (step bunches) is not expressible as a simple analytic function.


## 1. Introduction

The study of the morphology of solid surfaces is important in a number of surface processes such as crystal growth, epitaxy, and etching. Even for a qualitative comprehension of these processes it is necessary to understand the equilibrium structure of surfaces. The equilibrium shape of the crystal is determined by minimizing the surface free energy [1,2]. The surface tension $\gamma(\hat{n})$ (surface free energy) is not an analytic function of surface orientation $\hat{n}$ below its roughening temperature $T_{R}$ [3], the temperature at which the cusps in the surface tension disappear or equivalently the temperature above which a free energy associated with an atomic step can no longer be defined. As a consequence of the anisotropy of the surface free energy, a surface of minimum area is not necessarily the morphology required to minimize it. Indeed, the thermodynamics of solid surfaces predicts that surfaces of arbitrary orientation can be thermodynamically unstable with respect to the break-up, or faceting, into surfaces of different orientations, namely the 'hill-and-valley' structure [4] (see figure 1). The existence of such formations has been observed experimentally [5-8]. At the temperatures of interest in this study, namely at temperatures well below the roughening temperature, the surface consists of mainly monatomic steps separated by terraces. Steps are important entities in growth and equilibration processes because these processes occur only through the movement of steps on the surface. One way of characterizing the transition of the surface from a morphology of high energy to a morphology of low energy but not necessarily the minimum surface area is to consider explicitly the behaviour of atomic steps on the surfaces and investigate their motion in the course of the surface advancing towards the equilibrium shape. The motion of steps occurs through the attachment of atoms to or detachment of atoms from the step edges (see figure 2). To fully explore the motion of monatomic steps, a detailed knowledge
$\dagger$ E-mail address: metoz@pamuk.cu.edu.tr.
(a)

(b)


Figure 1. Break-up of a flat surface into a 'hill-and-valley' structure to minimize the surface free energy. For the case depicted, the flat surface in (a) is unstable with respect to faceting and undergoes break-up into a hill-and-valley structure in (b) whereby the surface tension is minimized. The requirement for break-up is that the average surface orientation $A_{0} \hat{n}_{0}=$ $A_{a} \hat{n}_{a}+A_{b} \hat{n}_{b}$ must be conserved and there must be a reduction in the surface free energy $A_{0} \gamma\left(\hat{n}_{0}\right)>A_{a} \gamma\left(\hat{n}_{a}\right)+A_{b} \gamma\left(\hat{n}_{b}\right)$ [4].


Figure 2. The labelling of steps and terraces and the definitions of $k_{ \pm}$, the kinetic coefficients for adatom attachment to or detachment from a step from or to the two bounding terraces. Steps whose heights always decrease (or increase) as one goes in a certain direction on the surface are called 'like' steps. The macroscopic orientation of the surface must be conserved in this process.
of the form of step-step interactions is essential. Repulsive interaction between pairs of 'like' steps on a vicinal surface which goes as the inverse square of the average step separation may
result from elastic [9-11] or entropic [12] interactions. Entropic interactions, studied first by Gruber and Mullins [12], result from the wandering of steps caused by the thermal excitation of kinks at non-zero temperatures, which increases the configuration entropy of steps and thereby reduces the step free energy. Besides the repulsive interactions, experimental studies of equilibrium shapes dictate that there must exist attractive interactions between steps which fall off inversely with the average distance between steps, for which unfortunately a satisfactory theoretical explanation is not yet completely achieved. Saenz and Garcia [13] indicated the necessity of having such an interaction in order to explain the experimentally observed [14] equilibrium shape of gold crystallites. Further experiments on the equilibrium shape of indium crystallites by Metois and Heyraud [15] also suggest that inclusion of such a term gives a better fit of theoretical predictions to the observed shapes. Other recent experimental [16] and theoretical [17] studies also do not exclude the possibility of attractive interactions between step pairs. Scanning tunnelling microscopy experiments on $\mathrm{Cu}(100)$ surfaces by Ibach and coworkers [16] clearly demonstrate that attractive interactions between steps must exist to account for the equilibrium shape observed. Redfield and Zangwill [17] predict an attractive interaction which falls off as the square of the distance between the steps, while an attractive interaction varying as the inverse of the distance between the steps was predicted earlier by Yamamoto and Izuyama [18] to explain the experimental study mentioned earlier [14]. Accordingly, the subject of this paper is the investigation of the final equilibrium morphology of an initial crystalline surface misoriented relative to a low-energy direction and consisting of monatomic steps when, in addition to entropic repulsive interactions, an attractive interaction of the form $\ell^{-1}$, where $\ell$ is the average step separation, exists between step pairs. It is expected that while repulsive interactions only lead to the flattening of the surface in the direction that corresponds to the minimum-surface-energy orientation, inclusion of attractive interactions must create step bunches on the surface in the form of macro-steps separated by large flat portions as observed in the experiments on Si surfaces [5-8]. Theoretical studies of this subject were carried out by Tersoff et al [19], who assumed a logarithmic interaction between step pairs, and more recently by Jeong and Weeks [20], who took the dynamics of surface reconstruction explicitly into account.

The plan of this paper is as follows. In section 2 we derive the equations of motion for each individual step on the surface in the presence of both attractive and repulsive interactions between step pairs. Section 3 is devoted to the numerical solution of these equations. The resulting surface morphology and the time dependence of the evolution are presented and compared with experimental results. Section 4 contains conclusions and a summary.

## 2. Equations of motion

In this section, we derive the equations of motion for each step on the surface. We consider a surface misoriented with respect to a high-index plane in one direction only. Hence the geometry is essentially one dimensional and parallel straight steps are separated by terraces. A portion of the surface studied is shown in figure 2. For the geometry considered, the only driving force for the motion of the steps is provided by the step-step interactions. For the case of repulsive step interactions only, the equations of motion were derived elsewhere [21,22] and solved completely [22]. Here we only add an attractive interaction of the form $\ell^{-1}$ between pairs of 'like' steps (see figure 2). Let us consider a one-dimensional surface of length $L=\sum_{n} \ell_{n}$ consisting of atomic steps of height $h$ where $\ell_{n}$ denotes the actual length of each individual terrace $n$. At small angles of misorientation $\theta$ of the actual surface with surfaces of highsymmetry (low-Miller-index) planes along which the surface tension has minima in the form
of cusps, one can expand the projected surface tension $G$ as

$$
\begin{equation*}
G=G_{0}+G_{1}|\tan \theta|-\frac{1}{2} G_{2} \frac{h^{2}}{L} \sum_{n} \frac{1}{\ell_{n}}+\frac{1}{3} G_{3} \frac{h^{3}}{L} \sum_{n} \frac{1}{\ell_{n}^{2}}+\cdots . \tag{1}
\end{equation*}
$$

The first term in the above expansion is the surface energy per unit area of the low-index surface, the second term can be interpreted as the sum of individual-step free energies since the slope $\tan \theta$ is proportional to the step density, and finally the third and the fourth terms are the total attractive and repulsive energies of interaction between pairs of steps, respectively. The constants $G_{i}$ appearing in the above equation are temperature dependent. The dependence of $G_{1}$ on temperature comes from the fact that at non-zero temperatures kinks at step edges may be excited. The formation of kinks in turn allows the step to wander, thus increasing its configurational entropy and hence reducing its free energy. This term is absent above the roughening temperature because a step can no longer be defined. The fourth term in equation (1) represents entropic interactions between step pairs which result from step fluctuations and the restriction imposed on the fluctuations by the impossibility of two steps crossing each other [12]. To be more definite, we consider a step bounded by two solid walls from above and from below. The number of different configurations that can be realized by that step is restricted by the bounding walls; hence the step has less entropy than it would have if the step were free to make fluctuations of any amplitude. The increase in free energy results in an effective repulsive interaction between the steps. The new term in this expansion, the third term, represents an effective attractive interaction between step pairs. The origin of that interaction is not yet completely understood, but is needed to explain the experimental equilibrium shapes of small crystallites noted earlier [13-15]. We will assume that such an interaction between step pairs exists, without providing any solid theoretical justification.

The motion of a particular step on the surface occurs through the attachment of adatoms to or detachment of atoms from the step edges from or to the two bounding terraces (figure 2). Creating such a virtual variation in the surface by a transfer of particles to a step labelled $n$, one can easily show that the variation in the projected surface energy (1) is given by

$$
\begin{equation*}
\delta G=2 \frac{1}{3} G_{3} \frac{h^{2}}{L}\left\{\frac{1}{\ell_{n}^{3}}-\frac{1}{\ell_{n-1}^{3}}\right\} \Delta \ell-\frac{1}{2} G_{2} \frac{h}{L}\left\{\frac{1}{\ell_{n}^{2}}-\frac{1}{\ell_{n-1}^{2}}\right\} \Delta \ell \tag{2}
\end{equation*}
$$

where $\Delta \ell$ is the variation in $n$th terrace length $\ell_{n}$ in the direction perpendicular to the average step orientation. The above equation defines a step potential $\mu=\delta G / \delta N$ [21,22]:

$$
\begin{equation*}
\mu_{n}=2 K_{3}\left\{\frac{1}{\ell_{n}^{3}}-\frac{1}{\ell_{n-1}^{3}}\right\}-K_{2}\left\{\frac{1}{\ell_{n}^{2}}-\frac{1}{\ell_{n-1}^{2}}\right\} \tag{3}
\end{equation*}
$$

where $\delta N$ is the number of particles transferred in the virtual displacement of the step described above, $\Omega$ is the atomic volume, and

$$
K_{2}=\frac{1}{2} G_{2} \frac{\Omega h}{L} \quad \text { and } \quad K_{3}=\frac{1}{3} G_{3} \frac{\Omega h^{2}}{L} .
$$

The steady-state diffusion equation

$$
\begin{equation*}
\frac{\partial C_{n}}{\partial t}=D_{s} \frac{\partial^{2} C_{n}}{\partial x^{2}}=0 \tag{4}
\end{equation*}
$$

where $D_{s}$ is the surface diffusion constant must be solved for each terrace $n$ in order to compute the velocity of the steps. It is assumed that any flux of atoms to the surface from a third dimension is completely arrested, so there are no additional terms in the diffusion equation. The diffusion equation is solved by

$$
\begin{equation*}
C_{n}(x)=a_{n}+b_{n} x \quad x_{n} \leqslant x \leqslant x_{n+1} \tag{5}
\end{equation*}
$$

for each terrace, where $a_{n}$ and $b_{n}$ are constants. Using appropriate boundary conditions following from mass-conservation equations and expressing the velocity of a step edge in terms of the variations of free energy due to particle attachment to or detachment from a step edge from the bounding two terraces, one can solve for the $b_{n}$ given by [22]
$b_{n}=\frac{\mu_{n+1}-\mu_{n}}{\nu D_{s}\left(1 / k_{+}+1 / k_{-}\right)+(\partial \bar{\mu} / \partial C)\left(x_{n+1}-x_{n}\right)}=(\partial \bar{\mu} / \partial C)^{-1} \frac{\mu_{n+1}-\mu_{n}}{d+\ell_{n}}$.
In the above equation, $\ell_{n}=x_{n+1}-x_{n}$ is the width of the $n$th terrace,

$$
d=v D_{s}\left(1 / k_{+}+1 / k_{-}\right)(\partial \bar{\mu} / \partial C)^{-1}
$$

is a parameter in units of length, $v$ is the inverse of the areal density of diffusing particles on the surface [23], $k_{ \pm}$are the kinetic coefficients for adatom attachment to or detachment from a step from or to the two bounding terraces (figure 2) and $\bar{\mu}$ is the chemical potential of an atom adsorbed on a terrace. Only the $b_{n}$ are needed to calculate the velocities of the step edges [21,22]:

$$
\begin{equation*}
v_{n}=v D_{s}\left(b_{n}-b_{n-1}\right) \tag{7}
\end{equation*}
$$

and the equation of motion for each terrace width $\ell_{n}$ takes the form

$$
\begin{align*}
& \dot{\ell}_{n}(t)=v D_{s}\left(b_{n+1}-2 b_{n}+b_{n-1}\right) \\
& \quad=K\left[\left(\frac{\mu_{n+2}-\mu_{n+1}}{d+\ell_{n+1}}\right)-2\left(\frac{\mu_{n+1}-\mu_{n}}{d+\ell_{n}}\right)+\left(\frac{\mu_{n}-\mu_{n-1}}{d+\ell_{n-1}}\right)\right] \tag{8}
\end{align*}
$$

where $K=v D_{s}(\partial \bar{\mu} / \partial C)^{-1}$. In the derivation of these equations, it is implicitly assumed that the steps are above their roughening temperature with the result that a step behaves like a line sink for the particles diffusing across terraces. As one can see from the above equation, the variation of a terrace width depends on the widths of the two neighbouring terraces on each side. An analytical solution of equation (8) in the absence of attractive interactions is possible provided that the initial surface has a certain well defined shape, and in that case the solution is shape preserving [22]. In the presence of attractive interactions also, however, it becomes an equation which is not easy to handle and therefore we will resort to solving it numerically. We can consider two distinct limiting cases for the evolution of terraces depending on the relative values of the length parameter $d$ defined in equation (6) and the average step separations $\ell$; that is, one can consider (a) a 'diffusion-limited' $(d \ll \ell)$ and (b) a 'step-attachment-limited' ( $d \gg \ell$ ) regime of step motions.

Let us briefly mention that in the case of repulsive interactions only, the evolutions of terrace widths for the cases (a) and (b) above go respectively as [22] $\ell_{n}(t) \sim t^{1 / 5}$ and $\ell_{n}(t) \sim t^{1 / 4}$ for an initial surface of sinusoidal corrugation. Similarly, in the presence of attractive interactions only, one can easily show that the evolutions of the terrace widths must go as $\ell_{n}(t) \sim t^{1 / 4}$ for the diffusion-limited regime and as $\ell_{n}(t) \sim t^{1 / 3}$ for the step-attachmentlimited regime for a similar initial surface.

Since equation (8) is not linear, superposition of solutions is not allowed and hence it is not possible to make a satisfactory long-time prediction about the evolution of terrace widths in the presence of both attractive and repulsive interactions using the combined equations given in the previous paragraph. But one can clearly deduce that in the presence of both attractive and repulsive interactions the enlargement of terrace widths cannot continue indefinitely. The step separations have an equilibrium separation given by equation (9) below. Let us define an average step separation as $\ell_{a v}=\sum_{i}^{N} \ell_{i} / N$ where $N$ is the total number of steps on the surface. Along the path towards equilibration, some terrace lengths will reach $\ell_{e q}$, provided that $\ell_{a v}>\ell_{e q}$, after which they will remain in the vicinity of that position. For that reason, after a finite time the surface must converge to a particular morphology and the evolution must
come almost to a halt, after which no appreciable variation in the surface takes place (see section 3). Consequently, a time dependence of terrace widths in the form $t^{\beta}$ where $\beta$ is a positive number cannot be correct for all times. Despite this fact, we tried to fit the numerical solutions for the terrace widths to a function of the form $c_{1}\left(1+c_{2} t\right)^{\beta}$ where the constants $c_{1}$, $c_{2}$, and $\beta$ are treated as adjustable parameters. The results are discussed in section 3 .

## 3. Solution of the equations of motion

In this section we solve equations (8) numerically for a number of different configurations and parameters. The two main parameters of the problem are:
(a) the length $d$ defined in equation (6) which determines whether the evolution proceeds in the 'diffusion-limited' $(d \ll \ell)$ regime or in the 'step-attachment-limited' $(d \gg \ell)$ regime;
(b) the relative strength of the temperature-dependent coefficients of repulsive and attractive interactions, namely the ratio $K_{3} / K_{2}$ defined in equation (3).

In the solution of these equations, periodic boundary conditions are used; that is, it is assumed that $\ell_{1}=\ell_{N}$ where $N$ is the total number of steps. The initial surface is usually chosen as having equally spaced monatomic steps and in that case the topmost step width is chosen to be slightly greater than the rest of the step separations in order to facilitate the evolution.

Before we explain the numerical solutions in detail, let us note that there is an equilibrium value for step separations noted above. If two neighbouring steps ever come to the equilibrium separation, they will remain in that position unless other driving forces arise and force them out of the equilibrium. The equilibrium separation can be obtained from equation (1) as

$$
\begin{equation*}
\ell_{e q}=\frac{2 K_{3}}{K_{2}} \tag{9}
\end{equation*}
$$



Figure 3. The evolution of an initial surface consisting of equally spaced monatomic steps obtained from the numerical solutions of equations (8) for the case where $K_{3} / K_{2}=3, d=1000$, and $\ell_{a v}=10$ atomic units. The height of each surface is 100 atomic units and the horizontal length in each is about $100 \ell_{a v}$. The surface shown at the bottom is the initial one and the one at the top is the shape of the final surface. Shown here are the surface morphologies at different times of the evolution not spaced equally in time. The formation of double, triple, and multi-step bunches can be seen if the sequence of surfaces from bottom to top are examined carefully.


Figure 4. Typical terrace width evolution curves obtained by numerically solving equations (8). The vertical axis is in atomic units while the horizontal axis is in arbitrary units, and the parameters are the same as for figure 3. Shown here is the evolution at initial times. Only a few of the terraces are shown, for clarity. Some terraces quickly approach the equilibrium separation (dotted curve) while others first expand and later approach $\ell_{e q}$ (dashed, solid, and dash-dotted curves). And some terrace widths continue the expansion as shown here (tiny-dashed curve). See also figure 5.
by minimizing the total interaction energy

$$
\begin{equation*}
U(\ell)=-\frac{K_{2}}{\ell}+\frac{K_{3}}{\ell^{2}} \tag{10}
\end{equation*}
$$

of a pair of steps of separation $\ell$. We have chosen $K_{3}>K_{2} / 2$ throughout our calculations.
For all parameter values considered, regardless of whether we are considering the diffusion-limited $\left(d \ll \ell_{e q}\right)$ or step-attachment-limited $\left(d \gg \ell_{e q}\right)$ regime, if the average step separation $\ell_{a v}$ defined in section 2 is greater than $\ell_{e q}$ given in equation (9), then the evolution proceeds qualitatively as follows. First, relatively quickly, pairs of steps at the equilibrium separation $\ell_{e q}$ are developed on the surface (see figures 3 and 4). At the same time, formation of triple steps at a distance of approximately the equilibrium separation away from each other starts. Later quadruple and multi-step bunches begin to appear. These formations can be clearly identified if one carefully examines the sequence of surfaces depicted in figure 3 from the bottom to the top. This process continues and eventually leads to a bunching of steps of almost equilibrium separation given by $\ell_{e q}$ at particular locations of the crystal surface. The speed at which the step bunching takes place and the number of steps that may exist in a step bunch (macro-step) is determined by the initial conditions (initial surface) and by the parameters of the problem. At a given time, double, triple, and multi-step bunches separated by terraces whose lengths are each greater than $\ell_{e q}$ may coexist. The step separation in each bunch is very close to $\ell_{e q}$. Figure 3 shows a typical time evolution of a surface where the initial surface consists of equally spaced monatomic steps. The surfaces depicted are at different times of the evolution and are not spaced equally in time. The surface shown at the bottom is the initial one and that at the top is the final one. The coexistence of step bunches of different sizes mentioned above can clearly be seen. Finally, for the case where the average step separation is less than the equilibrium value ( $\ell_{a v}<\ell_{e q}$ ), no step bunching is observed. Such a surface is
not in equilibrium and cannot reach a stable thermodynamic equilibrium unless, e.g., double or greater step heights are allowed.

Following the above summary, let us first start with the time dependence of the terrace widths $\ell(t)$. There is no simple analytical expression for their time dependence. Our numerical solutions show that while some steps quickly approach the equilibrium separation and remain in the close vicinity of that position throughout the evolution, others start expanding and continue the expansion for a while. Some of those terraces that tended to expand at the beginning may approach the equilibrium separation later, and this usually corresponds to a merging of two macro-steps to form another macro-step of size equal to the combined sizes of merged macrosteps. The process of merging of two (or more) macro-steps, and hence the approach of some of the terraces expanding until that time to the equilibrium separation, continues up to a point where no further coalescence of macro-steps is possible. Then the surface locks to a final shape and thereafter it shows no observable evolution. The final shape is in the form of macro-steps separated by large flat terraces. (See the topmost surface in figure 3.) The number of steps that are present in a macro-step and the lengths of the large terraces separating the macro-steps are primarily determined by the initial shape (i.e. mass conservation) and by the relative strength of the attractive and repulsive step interactions, namely by the ratio $K_{3} / K_{2}$. Typical terrace width variation curves at the very beginning of the evolution are shown in figure 4 . Some terrace widths quickly approach the equilibrium separation (dotted curve) while others first increase then later return to the equilibrium separation (dashed curve). On the other hand, some of the terraces continue expanding (tiny-dashed curve) and some of them may approach the equilibrium separation later in time (solid and dash-dotted curves). A variety of equilibration curves are possible for terrace widths, but only a few representative ones are shown in figure 4 for the purpose of clarity.


Figure 5. Typical terrace evolution curves as in figure 4, but at intermediate times. The parameters and axis definitions are the same as for figure 4 . Note that some of the previously expanding terraces approach the equilibrium separation while some others continue the expansion.

Figure 5 depicts the evolution of terrace widths at intermediate times. Note the similarity with figure 4. Here some of the previously expanding terraces approach the equilibrium separation (solid and tiny-dashed curves) while others continue to expand (dotted and dashed
curves). These expanding terraces actually separate macro-steps on the surface shown in figure 3. At later times some of those terraces that continued the expansion may approach the equilibrium separation through the integration of two macro-steps. This whole process of merging of (macro-) steps repeats until the surface reaches the final morphology (topmost surface) shown in figure 3 .


Figure 6. The complete time evolution of two of the large terraces remaining on the final surface shown in figure 3. The vertical axis is in atomic units and the horizontal axis is in arbitrary units. These and similar curves are fitted to a function of the form $c_{1}\left(1+c_{2} t\right)^{\beta}$ where the constants are variational parameters. From many different fits, the exponent $\beta$ is found to be in the range 0.16-0.22.

Most of the terrace widths ultimately approach $\ell_{e q}$, forming macro-step bunches separated by large terraces. Figure 6 shows the complete time dependence of two typical long flat terraces that survived to the end of the evolution. These are two of the large terraces appearing in figure 3 on the final surface. Note that the terrace widths shown in figure 6 increase quickly at the beginning and more slowly later, and the whole evolution looks like a ladder, i.e. when the overall evolution curves of terrace widths are considered, one sees instant increases in terrace widths followed by long plateaus where the evolution proceeds very slowly (figure 6). These impulsive increases in terrace widths are due to the mergings of pairs of macro-steps occurring in such a way that the separation between the monatomic steps in the resulting macro-step is still the equilibrium separation. But during the merging of the macro-steps, those terrace widths that have already approached the equilibrium separation are sometimes driven out of their equilibrium positions for a short duration and then later return to their original positions. This causes momentary spikes in the terrace widths.

As can be deduced from the foregoing, the time dependence of terrace widths is not expressible as a simple analytic function. A function of the form $c_{1}\left(1+c_{2} t\right)^{\beta}$ where the constants are adjustable parameters is fitted to the terrace evolution curves for large terraces such as the ones shown in figure 6. From many different fits that we made, a value for $\beta$ in the range $0.16-0.22$ is found. It appears that the functional fits are better for short times and the deviation is more pronounced for later times. The numerical evolution curves for terrace widths are more likely to resemble that found by Phaneuf et al [7] experimentally
rather than one given by an exponential function. The evolution takes place rather quickly at the very beginning, slows down at later times, and eventually the surface locks to a final shape after which there is no observable evolution. A similar investigation of crystal surface evolution for surfaces under stress was carried out by Tersoff et al [19] assuming a logarithmic attractive interaction between pairs of steps. Although the driving forces for step bunching are different in each study, the morphology of the final surface that we have obtained in this work is qualitatively very similar to theirs. But the value of the equilibrium step separation depends on different parameters in this work and in their work, and hence the slopes of the surfaces where step bunching took place are different.

We should also note that one can get an idea about the relative strengths of the coefficients $K_{2}$ and $K_{3}$ from the final shape of the crystal surface by recognizing that the slope of the macro-steps relative to the minimum-energy surface depends on the ratio $K_{3} / K_{2}$ through equation (9). Hence an experimental determination of that slope may give one an idea of their relative strengths at a given temperature. Furthermore, the variation of that slope as a function of temperature will provide the dependence of the ratio of these parameters on temperature. Another point that one notices is that the large flat parts and the macro-steps are separated from each other by sharp corners, i.e. the slope of the surface at the intersection of these two regions is not continuous.

Finally, let us note that the final surface morphology appears to be independent of the parameter $d$, although this case was not investigated in very great detail. It does change the time dependence of terrace widths and in fact preliminary work shows that for the step-attachment-limited case the terrace widths are scaled approximately as $\ell(t)=F\left(t /\left(d / \ell_{a v}\right)\right)$ provided that $\ell_{a v}>\ell_{e q}$ is satisfied by the initial surface. However, we have not analysed this behaviour in detail.

## 4. Conclusions

In this paper we have investigated the equilibration of a surface initially oriented relative to a low-energy surface at a small angle and consisting of monatomic steps. The problem is investigated with the presence of both repulsive and attractive interactions between pairs of steps. The resulting morphology is in the form of a 'hill-and-valley' structure predicted long ago by Herring [4]. The time dependence of the large flat parts which separates macro-steps on the surface is not expressible as a simple analytic function. While most of the terrace widths quickly approach and remain in the close vicinity of $\ell_{e q}$, the evolution for those terraces separating two macro-steps proceeds with sharp increases in their widths followed by long slowly varying plateaus. The abrupt increases in the terrace widths are due to the mergings of pairs of macro-steps. A detailed knowledge of step interactions and their temperature dependence is required for a better quantitative investigation of the problem.

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