Step motions on high-temperature vicinal surfaces

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Abstract. The fluctuations of a train of steps on a vicinal surface at equilibrium and during evaporation are studied in the framework of Burton, Cabrera and Frank's theory. Step fluctuations are treated as perturbations of the straight step's shape and within a linear analysis the step morphological stability is investigated in a general way. Previous results of Bales and Zangwill and of Uwaha and Saito are obtained as special cases when asymmetry between kinetic coefficients for adatom attachment at step edges (the 'Schwoebel effect') is included. A strong Schwoebel effect is known to lead to step bunching and to smoothing of an isolated step during evaporation. Here we show that a strong Schwoebel effect leads instead to roughening of steps in a train at evaporation temperatures. Furthermore, we show that the Schwoebel effect is negligible on vicinal surfaces with widely spaced steps at evaporation temperatures. For both reasons, we conclude, in contrast with Uwaha and Saito, that the observed phenomenon of kinetic step smoothing during silicon evaporation cannot be justified by the presence of a strong Schwoebel effect. The experimental situation is discussed and a scenario is proposed that does not invoke a strong Schwoebel effect.

1. Introduction

The dynamics of steps on vicinal surfaces at and out of equilibrium is a subject of substantial current interest, from both the theoretical [1–9] and the experimental [10–14] point of view. Theoretical investigations of the out-of equilibrium case usually take as their starting point the seminal paper by Burton, Cabrera and Frank (BCF) [15], extended so as to include the so-called 'Schwoebel effect' [16]. This refers to a by now well established [10–19] asymmetry in the attachment kinetics of adatoms at ascending versus descending steps. Generally speaking, an adatom trying to stick to a descending step has to climb an energy barrier higher than the simple diffusion barrier. Therefore, the probability of a sticking event will be reduced compared with that of an adatom approaching an ascending step. Such a barrier has many implications concerning the stability of a train of steps, since it is able to induce step bunching [7, 16]. It is also important for the stability of the surface itself, since a flat high-symmetry face becomes unstable when a net up-step surface diffusion current is present [20, 21]. However, the situation is not always as simple as that, since cooperative mechanisms may be devised [19] that favour the overcoming of the step barrier.

The Schwoebel effect may be responsible for two kinds of instability: (i) step bunching in the direction perpendicular to the steps [16]; (ii) step meandering [3]. The former occurs at evaporation and the latter during growth. In the latter case the straight shape of steps becomes unstable because diffusing adatoms attach preferentially to a protruding part of

a non-straight step (as in diffusion-limited aggregation) and mostly from the lower terrace (due to the asymmetric kinetics); if a sufficiently high supersaturation is maintained on the terraces, a protrusion in a step is enhanced (see [3]), whence the instability.

Uwaha and Saito [4] used this same concept of asymmetric attachment kinetics to describe the stabilization of the step shape during sublimation of the crystal [12]. In this case, because of detailed balance the Schwoebel effect mechanism implies that adatoms detach from a step more easily towards the lower terrace than to the upper one. If the adatom density on the terraces is sufficiently reduced by evaporations, step atoms have a stronger tendency to detach from protruding parts, which should lead to damping of fluctuations and smoothing of the step shape (figure 1). This simple argument, however, applies only to isolated steps, since it neglects correlations between neighbouring steps due to the diffusing adatoms. In any case, the phenomenon of step smoothing has been reported on vicinal (111) surfaces of silicon in the evaporation regime (see [12] and the next section). It is this phenomenon that partly motivated our work.
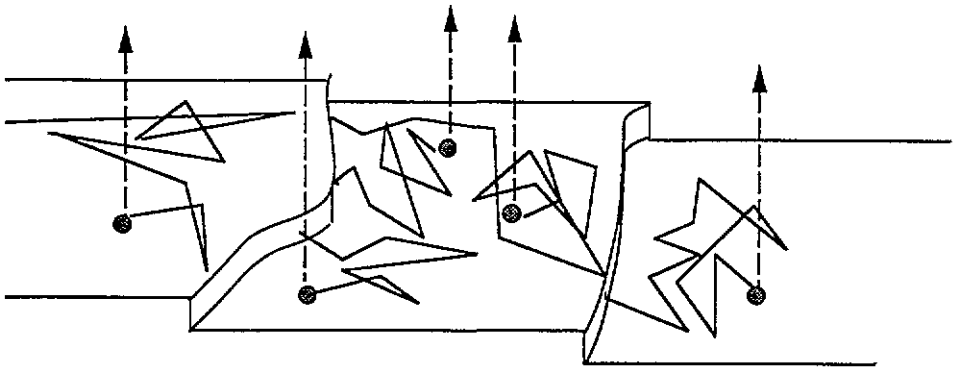

Figure 1. Step fluctuation damping during evaporation, in the presence of a strong Schwoebel effect. Atoms detach preferentially from protruding meanders to the terrace in front, and are not able to cross the step and attach to the upper terrace. This simple scenario may be modified by step coupling through the diffusing adatoms.

The main ambition of the present work is to point out that no specific study of an *evaporating* vicinal surface has yet been made, and that some points are worth reconsidering. Bales and Zangwill [3] investigated the full problem of a step train, but only considered situations where all steps are fluctuating in phase. In the case of growth this 'in-phase' mode turns out to be the most unstable one, so the growth instability is correctly predicted in [3]. In the case of evaporation, however, the most unstable mode will be seen in section 3 to be the 'anti-phase' mode, when any pair of consecutive steps fluctuate in phase opposition. The explicit treatment of the step–step phase relation is therefore crucial. The analysis of Uwaha and Saito [4], which is limited to an isolated step, is thus not satisfactory. In the present article, the relaxation rate of step fluctuations is evaluated for any phase shift.

The second important point that we want to stress is the magnitude of the Schwoebel effect. A very strong effect, which overdamps the fluctuations of an evaporating isolated step, as Uwaha and Saito showed [4], *leads instead in a train to a step-pairing instability and enhances the fluctuations of steps*, as we show in section 5.1. This contrasts with the observation [12] of an evaporating train of steps: (i) the steps remain approximately equidistant; (ii) step fluctuations are damped. In this paper, following [3] we associate with the step-edge potential barrier a typical length, d_S, which can be thought of as the mean

free path of an adatom before it descends the step. If this length is shorter than the interstep distance ℓ, a diffusing adatom is more likely to descend the step than to cross the terrace back to the nearest ascending step. In this case, the Schwoebel barrier is negligible. This scenario is most likely to apply at the high temperatures where sublimation takes place and on the large terraces ($\approx 10^3$ Å) observed in the experiments [12].

The paper is structured as follows. In section 2 we discuss the experimental scenario that we are interested in. In section 3 the linear stability of a fluctuational perturbation on the shape of steps in an array with separation ℓ is addressed. In section 4 we check our formulae against known results for steps at equilibrium. The case of evaporation is discussed in section 5, where we distinguish between an isolated step and a step train, and between strong and weak Schwoebel effects. A discussion of the observed step smoothing and of the possible importance of the Schwoebel effect at evaporation temperatures is proposed in section 6. We conclude with a summary in section 7. Two appendices containing details of calculations close the article.

2. Step flow sublimation of vicinal surfaces: the experiments

When surfaces that are vicinal of a high-symmetry orientation are heated under ultrahigh vacuum (UHV), a temperature region is reached where the steps fluctuate about fixed equilibrium positions. Such fluctuations on a silicon surface vicinal of orientation {111} have been studied by reflection electron microscopy (REM) [11]. During these experiments no atom is deposited, and adatom desorption is negligible, so steps can only exchange matter with surrounding surface regions. Due to the high temperature (900 °C), surface diffusion is fast enough to warrant equilibration of the step distribution. Statistical [11] and dynamical [1, 2] properties of the step fluctuations have been investigated in detail.

On further increasing the temperature (>1000 °C), the adatoms appreciably desorb, the step train starts to move towards the up-step direction in what is known as step flow motion, and the crystal sublimates [12]. In the sublimation regime the surface undergoes a morphological transition, called 'macroscopic facetting' [10, 14]. The steps regroup in many close bands ('step bunching'), where they cannot be resolved by REM, separated by large terraces ('facets') several tens of microns in size. In these terraced regions there is a very weak density of non-bunched steps, with separations ℓ ranging between 100 Å and some microns. It is the fluctuations of these widely spaced steps that are studied, since step bunching is always produced during preparation of the sample (high-temperature cleaning) and persists down to lower temperatures due to very slow kinetics. Note that step bunching is strongly dependent on, and very likely due to, the electric current used to heat the sample [10, 23]. Actually, steps bunch only under DC conditions, and the temperature where this happens depends on the current direction with respect to the step-up and step-down orientation. Step bunching does not occur if AC heating is employed [10], but unfortunately the latter prevents REM observation. Also, spiral steps emerging at screw dislocations appear during sublimation; the spiral rotates regularly with a movement equivalent to step flow.

Step flow and spiral sublimation take place as described by Burton, Cabrera and Frank's theory (see the following section). The observed step velocity is linear in the step–step separation ℓ. According to theory, this implies that the typical length on which adatoms diffuse before desorption, x_s, is much larger than ℓ at all temperatures investigated (up to 1305 °C) [10, 11]. Surprisingly, during step flow motion the step fluctuations are strongly suppressed, the more so the higher the temperature and the evaporation rate, and at high enough temperatures (> 1250 °C) the steps look smooth on a typical length of 1 μm within the experimental resolution (≈ 100–50 Å).

There is no convincing explanation for step smoothing. Recently, Uwaha and Saito [4] discussed this kinetical step smoothing by explicitly taking into account the Schwoebel effect. Bales and Zangwill [3] have shown that the Schwoebel effect produces an instability of Mullins–Sekerka type of the step shape during step flow growth: if the supersaturation is large enough compared with the step line stiffness, which has a stabilizing effect, fluctuations are strongly amplified and the straight shape becomes unstable. Uwaha and Saito [4] showed that during evaporation the straight shape of a step is always stable, and step fluctuations are decreased compared with the equilibrium case. However, they only considered an isolated step, and an unphysically large sticking asymmetry. We reconsider the whole area in the following.
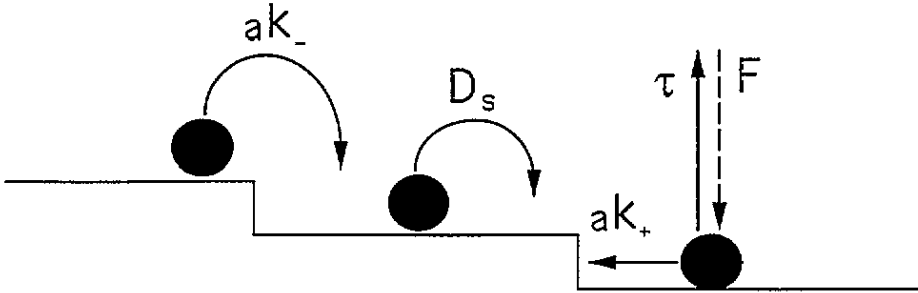

Figure 2. Microscopic rate parameters for adatoms dynamics in Burton, Cabrera and Frank's model of step motion. See the text for the definitions of the parameters.

3. Burton, Cabrera and Frank's theory of step flow growth and evaporation

The theory of Burton, Cabrera and Frank (BCF), originally devised for growth, can easily be adapted to the case of sublimation of a stepped surface. In this case one writes a diffusion equation for the adatom density $c(x, y, t)$ on a terrace delimited by two steps at distance ℓ,

$$\frac{\partial c}{\partial t} = D_s \nabla^2 c + F - \frac{c}{\tau} \tag{1}$$

where D_s is the adatom surface diffusion constant, F the atom deposition flux and $1/\tau$ is the desorption probability of an adatom per unit time and surface area (see figure 2 for a graphical definition of microscopic parameters). The important ingredients are now the boundary conditions at the steps. It is here that the asymmetry of the attachment–detachment kinetics plays its role. It is included by writing

$$D_s \left(\frac{\partial c}{\partial n} \right)_+ = k_+ \left[c - c_{eq}^0 + \Gamma \kappa_m \right]_+ \tag{2}$$

$$-D_s \left(\frac{\partial c}{\partial n} \right)_- = k_- \left[c - c_{eq}^0 + \Gamma \kappa_m \right]_- \tag{3}$$

where $\Gamma = c_{eq}^0 \Omega \beta \gamma$, c_{eq}^0 is the equilibrium adatom concentration, γ is the step stiffness, κ_m is the local step curvature and n is the local step normal. The coefficients k_+ and k_-, which refer to adatoms coming from the lower and upper terrace, respectively, determine the flux of adatoms onto the step. If k_- (respectively, k_+) vanishes, no adatom sticks from the upper (respectively, lower) terrace. In contrast, $k_\pm = \infty$ means instantaneous attachment of all adatoms impinging from the respective terraces. The latter condition warrants local

equilibrium at the step since it implies $[c - c_{eq}^0 + \Gamma\kappa_m]_\pm = 0$. At the high temperatures of interest here, it is quite natural to assume that each step is in equilibrium with the lower terrace, that is, $k_+ = \infty$. Very little is known on possible values for k_-, and one of our goals is to learn something about it. The special choice of completely asymmetric boundary conditions ($k_- = 0$, $k_+ = \infty$) leads to what is called the 'one-sided model' in the literature: each step is a perfect mirror for the adatoms coming from the upper terrace, and a perfect sink for those impinging from the lower side. We will investigate what happens if one departs from this extreme assumption by allowing the potential barrier at descending steps to be finite.

If one assumes that diffusion is sufficiently fast, that is $D_s\tau > \ell^2/\Omega^2$, the time dependence in (1) can be ignored, and a quasi-static solution can be found. In fact, a weaker condition can be found to be sufficient. If v is the step velocity, the quasi-static solution is a good approximation to the true one when v is smaller than the typical adatom velocity $\sqrt{D_s/\tau}$. For small terraces, $v/\Omega = \ell c_{eq}^0/\tau$. Thus, $v < \sqrt{D_s/\tau}$ implies $(\ell\Omega c_{eq}^0)^2 < D_s\tau$. The two conditions are essentially equivalent on Si(111) at high temperature (> 850 °C), where $c_{eq}^0 \approx 1$–0.1. They are well satisfied in this case at least up to 1300 °C.

Bales and Zangwill [3] investigated in the framework of BCF theory the onset of the shape instability of steps at mutual distance ℓ during step flow growth by atom deposition. If atoms are deposited at a rate F, either from the vapour or a beam, and an asymmetry in the attachment kinetics of adatoms at steps from the upper and from the lower side is included, then a fluctuation of sufficiently long wavelength will grow exponentially and the straight step configuration will become unstable. In order to evaluate the instability threshold quantitatively, the mth step is described as a function $x_m(y, t)$. The straight $t = 0$ configuration is thus $x_m(y, 0) = $ constant. At finite time one writes $x_m(y, t) = x_m(y, 0) + \delta x_m(y, t)$, where δx_m is a small-amplitude perturbation. Choosing $\delta x_m(y, t) = \epsilon \cos(qy) \exp(im\phi + \omega t) + CC$, one can compute the relaxation rate $\omega(q, \phi)$ as a function of the phase shift ϕ. The latter describes the phase relation of neighbouring steps in the train. A positive $\omega(q, \phi)$ for a given ϕ and some q means that the perturbation relative to that particular wavelength will grow unstably. For instance, Bales and Zangwill computed $\omega(q, \phi)$ assuming that all steps are fluctuating in phase, that is, for $\phi = 0$. As we show below, the in-phase mode is the most unstable one during growth, but it is not always so. We have therefore computed $\omega(q, \phi)$ for arbitrary phase shifts ϕ between adjacent fluctuating steps, and then determined which one is the most unstable mode in the various cases. We only give here the result, deferring all the rather cumbersome computational details to appendix 1. One finds that in the case of an arbitrary phase shift ω is a complex quantity, $\omega = \omega_r + i\omega_i$. As usual, the real part gives the relaxation rate, while the imaginary part gives the modulation of an oscillating contribution in time. As we show in appendix 1, the imaginary part is proportional to $\sin\phi$, so it vanishes for $\phi = 0$ or π. In fact, we will only consider these two values, since we will only be concerned with the most unstable in-phase and anti-phase modes.

Using Bales and Zangwill's notation (see (13) in their paper) we write

$$\omega(q, \phi) = g(q, \phi) - q^2 f(q, \phi) \tag{4}$$

where f which is given by

$$f(q, \phi) = D_s\Omega\Gamma\Lambda_q \frac{2[\cosh(\Lambda_q\ell) - \cos\phi] + \Lambda_q(d_+ + d_-)\sinh(\Lambda_q\ell)}{\Lambda_q(d_+ + d_-)\cosh(\Lambda_q\ell) + (d_+d_-\Lambda_q^2 + 1)\sinh(\Lambda_q\ell)} \tag{5}$$

is always positive and expresses the stabilizing effect of the line stiffness. The term $g(q, \phi)$ explicity reads

$$g(q, \phi) = \Omega \Delta F(d_- - d_+)N(q, \phi)/D(q) \tag{6}$$

where we defined

$$N(q, \phi) = \Lambda_q(d_+ + d_-)\big[\cos \phi + \Lambda_q x_s \sinh(\Lambda_q \ell) \sinh(\kappa \ell)$$
$$- \cosh(\kappa \ell) \cosh(\Lambda_q \ell)\big] + q^2 x_s^2 [\cosh(\kappa \ell) - 1] \sinh(\Lambda_q \ell) \tag{7}$$

$$D(q) = \big[(d_+ + d_-) \cosh(\kappa \ell) + (\kappa d_+ d_- + x_s) \sinh(\kappa \ell)\big]$$
$$\times \big[(d_+ + d_-)\Lambda_q \cosh(\Lambda_q \ell) + (d_+ d_- \Lambda_q^2 + 1) \sinh(\Lambda_q \ell)\big] . \tag{8}$$

Here Ω is the atomic area, $\Delta F = F - F_{eq} = F - c_{eq}^0/\tau$. The lengths $d_\pm = D_s/k_\pm$ are also introduced, which depend on the attachment coefficients k_+ and k_-, which refer to adatoms coming from the lower and upper terrace, respectively. The length $x_s = \sqrt{D_s \tau}$ is the surface diffusion length and $\kappa = 1/x_s$. Finally, $\Lambda_q = \sqrt{q^2 + \kappa^2}$.

3.1. On the role of the phase shift ϕ

Equation (4) displays separately the stabilizing effects (due to the step stiffness and to the step coupling through the diffusing adatoms), contained in f, and the destabilizing effect of the under- or supersaturation ΔF in the presence of a Schwoebel effect ($d_- - d_+ > 0$), contained in g. The latter vanishes as expected for symmetric kinetics at steps ($d_- = d_+$) and at equilibrium (i.e. when $F = F_{eq}$). In the case of growth ($\Delta F > 0$) it follows from (6) and (8) that $g(q, \phi)$, and therefore $\omega(q, \phi)$, have, as functions of ϕ, their maximum at $\phi = 0$ for any value of q. Therefore, the in-phase mode ($\phi = 0$) has the smallest relaxation rate and is thus the most unstable. This leads to the growth instability studied by Bales and Zangwill [3]. However, in the case of evaporation ($\Delta F < 0$) $g(q, \phi)$ and $\omega(q, \phi)$ have their maximum at $\phi = \pi$ for any q. Therefore the most unstable mode is in this case the anti-phase mode ($\phi = \pi$).

As a general rule, $\phi = 0$ gives the most unstable mode when $\Delta F(d_- - d_+) > 0$, and $\phi = \pi$ does it when $\Delta F(d_- - d_+) < 0$.

Finally, a comment on the lengths $d_\pm$. They are large if the corresponding kinetic coefficients $k_\pm$ are small compared to D_s/a, a being the lattice spacing. In particular, a very small d_- corresponds to a very low sticking coefficient from the upper terrace, and thus to a strong Schwoebel effect. Therefore d_- is a candidate as the relevant characteristic length to be compared with ℓ in order to judge of the importance of the Schwoebel effect. We will return to this point later.

4. Steps at equilibrium

As a first application we will re-derive from BCF theory the results found in [2] for steps at equilibrium. In that paper the creation time $\tau(L)$ of a perturbation of wavelength L was computed with a quite phenomenological approach. More details can be found in appendix 2. The time $\tau(L)$ should be equal at equilibrium to the reciprocal of the relaxation rate $\omega(\phi, q)$, equation (4), for $q = 1/L$, which allows the comparison. The derivation of ω given here is of course more rigorous, but it poses some problems: what value of the phase difference ϕ shall we choose? The most natural assumption is to consider any value as likely *a priori* as any other, and to average over ϕ with a uniform distribution over $(0, 2\pi)$.

Since in this case $\langle \cos \phi \rangle = 0$, we can also choose as typical the value $\phi = \pi/2$. This allows us to find the same results as in [2].

Having in mind a crystal under ultra-high vacuum at temperatures where evaporation is still negligible, we take $\tau \to \infty$ (which implies $\kappa \to 0$ and $\Lambda_q \to q$), and no deposition flux ($F = 0$). Therefore $g(q, \phi)$ vanishes identically, $f(q, \phi)$ has the interpretation of the equilibrium restoring force on a fluctuating step, and $\omega_{eq}(q, \phi) = -q^2 f(q, \phi)$ gives the reciprocal of the relaxation time of a step fluctuation of wavenumber q.

We treat for simplicity only the case of instantaneous sticking of adatoms to an ascending step. In a discretized model this should imply $d_+ = a$, that is the smallest possible value for d_+. In this continuum version we write instead $d_+ = 0$ if the sticking is instantaneous. We thus let $d_+ = 0$ in the following and write $d_- = d_S$ (S for Schwoebel) for typographical convenience. We also let $a = \Omega = 1$ everywhere . In the absence of desorption and deposition equation (5) reads

$$f(q, \phi) = D_s \Gamma q \frac{2[\cosh(q\ell) - \cos \phi] + q d_S \sinh(q\ell)}{q d_S \cosh(q\ell) + \sinh(q\ell)}. \tag{9}$$

4.1. An isolated step at equilibrium

Let us consider first the limit of an isolated step, $\ell \to \infty$. From formula (9) one finds

$$\omega_{eq}(q, \phi) \approx -D_s \Gamma q^3 \frac{2 + q d_S}{1 + q d_S}. \tag{10}$$

Clearly, the result does not depend on ϕ in this case. For both strong and weak Schwoebel effects we find in order of magnitude

$$\omega_{eq}(q, \phi) \approx -D_s \Gamma q^3 = -D_s c_{eq}^0 \beta \gamma q^3 \tag{11}$$

which is consistent with (11) (see [2]). Note that the diffusion constant enters via the product $D_s c_{eq}^0$, in much the same way as the desorption rate only appears as the ratio c_{eq}^0/τ. It is in fact a general rule that the consideration of macroscopic (with respect to the atomic spacing) phenomena only allows one to measure such 'macroscopic' (since they involve the particle density) quantities. The diffusion length x_s is a measure of the relative importance of the two mass transport terms, $D_s c_{eq}^0$ and c_{eq}^0/τ, since $x_s^2 = (D_s c_{eq}^0)/(c_{eq}^0/\tau)$.

4.2. A train of steps at equilibrium

We consider now the case of a train of steps at mutual distance ℓ. In this case, again neglecting deposition and evaporation, from equation (9) one immediately finds the leading term in q,

$$\omega_{eq}(q, \phi) \approx -D_s c_{eq}^0 \beta \gamma (1 - \cos \phi) \frac{q^2}{\ell + d_S}. \tag{12}$$

For $\phi = 0$ this term vanishes identically, and $\omega_{eq}(q, 0)$ is proportional to q^4 at small q. We will shortly show that the q^4-behaviour is characteristic of a very strong Schwoebel effect.

We consider first the case of a small potential barrier at descending steps (compared with temperature); that is, the weak Schwoebel effect.

4.3. A train of steps: the weak Schwoebel effect

In the weak Schwoebel effect d_S is smaller than the average step–step separation ℓ. Neglecting d_S in (12) and letting $\cos\phi = 0$, one finds that the relaxation time of a thermal fluctuation is proportional to ℓ, as in formula (12) of [2]. This dependence on ℓ of the relaxation time of (equilibrium) thermal fluctuation has also been observed on Si (111) at 900 °C [2]. At this temperature desorption is still negligible—the evaporation rate is approximately 10^{-4} s^{-1}. The sample was under ultra-high vacuum and no atoms were deposited. In this case equation (12) applies correctly. The physical implication is the following: a potential barrier is actually present at down steps, but if the terrace size ℓ is large enough, that is, if $\ell \gg d_S$, its effect is negligible. Thus, the crucial question we have to answer is: how strong could the Schwoebel effect reasonably be in the experimental situation? In [2] a relaxation time proportional to ℓ is found for terraces as narrow as 300 Å. If we assume that d_S is controlled by the barrier difference ΔW_S through the activated form

$$d_S \equiv \exp[\beta \Delta W_S] \tag{13}$$

we are able to provide an upper bound to ΔW_S. Letting $d_S \approx \ell \approx 300$ Å at $T = 1173$ K, we find $\Delta W_S \approx 0.47$ eV. The value seems very reasonable. Latyshev *et al* [10] claim that according to their measurements 'the difference in the values of energetic barriers for adatoms incorporation into the step ranges from 0.2 to 0.6 eV'. Since we assumed one of the barriers to vanish, this agrees with our estimate. Also, Zhang *et al* found from model potential calculations [19] that the potential barrier at the descending step, W_S, exceeds the potential barrier on the surface, W_{sd}, by approximately 0.5 eV. The latter is actually valid for step on a (001) surface, but the order of magnitude should be correct in general for semiconductors. Since $d_S = D_s/k_S$, we would find $\Delta W_S = W_S - W_{sd}$ if k_S were an activated quantity with activation energy equal to W_S. A natural expectation is that, in the higher-temperature range where evaporation takes place, d_S should decrease, and that the Schwoebel effect should become unimportant for still narrower terraces.

Note that both limiting cases (11) and (12) with $d_S \ll \ell$ can be summarized in the scaling form

$$\omega_{eq}(q,\phi) \approx -2D_s c_{eq}^0 \beta\gamma q^3 \left[\frac{1 - \cos\phi}{\sinh(q\ell)} + \tanh\left(\frac{q\ell}{2}\right) \right] \tag{14}$$

which results from equation (9) by letting $d_S \to 0$.

It is important to note that equation (12) cannot be found as limiting case of the formulae published by Bales and Zangwill [3]. We recall that they implicitly assume from the beginning $\phi = 0$, which correctly describes the growth instabilities but does not give the right behaviour at equilibrium or during evaporation. This clearly shows the need for explicitly treating the case of non-zero ϕ.

4.4. A train of steps: the strong Schwoebel effect

We give for completeness the result that one obtains from (9) if one assumes the Schwoebel effect to be strong. Letting $d_S \to \infty$ we see that the term containing $\cos\phi$ is negligible, and one finds

$$q^2 f(q) \approx D_s c_{eq}^0 \beta\gamma q^3 \tanh q\ell . \tag{15}$$

Equation (11) is recovered in the isolated step limit, as expected. In the case of a train of steps, we find instead for $q\ell < 1$

$$q^2 f(q) \approx D_s c_{eq}^0 \beta\gamma q^4 \ell . \tag{16}$$

This result agrees with equation (14) of [2]. Note that a q^4-dynamics is typical of one-dimensional diffusion [22]. In fact, coarse-graining on ℓ, much smaller than d_S in this case, one gets an arrays of fluctuating strings that do not exchange matter. Along each string there is then a coarse-grained density of diffusing adatoms (per unit length) $c_{eq}^0\ell$. On the other hand, dynamic renormalization is expected to produce an effective q^2-term (see for instance [8] and references therein).

We turn now to the question of major interest in this paper, that is, step motions during sublimation of a vicinal surface.

5. Step dynamics and step shape during evaporation

Let us go back to equation (5) and consider again weak ($d_S \to 0$) and strong ($d_S \to \infty$) Schwoebel effects, however keeping a non-vanishing evaporation rate c_{eq}^0/τ, and thus a non-zero diffusion length $x_s = 1/\kappa = \sqrt{D_s\tau}$. We find thus the formulae corresponding to equations (14) and (15), respectively:

$$q^2 f(q,\phi) = 2D_s\Gamma q^2 \Lambda_q \left[\frac{1-\cos\phi}{\sinh(\Lambda_q\ell)} + \tanh\left(\frac{\Lambda_q\ell}{2}\right)\right] \tag{17}$$

where we assumed $d_S \ll \ell$;

$$q^2 f(q,\phi) = D_s\Gamma q^2 \Lambda_q \tanh(\Lambda_q\ell) \tag{18}$$

where we assumed $\ell \ll d_S$.

However, it is now necessary to include into $\omega(q,\phi)$ also $g(q,\phi)$, which does not vanish, in general, out of equilibrium.

As we did with f, we write $g(q,\phi)$ for weak and strong Schwoebel effects, respectively:

$$g(q,\phi) \approx d_S\Delta F q^2 x_s \tanh\left(\frac{\kappa\ell}{2}\right) \tag{19}$$

for $d_S \to 0$, and

$$g(q,\phi) \approx \Delta F \frac{1}{\cosh(\kappa\ell)\cosh(\Lambda_q\ell)} \big[\cos\phi - \cosh(\kappa\ell)\cosh(\Lambda_q\ell) \tag{20}$$
$$+ \Lambda_q x_s \sinh(\kappa\ell)\sinh(\Lambda_q\ell)\big]$$

for $d_S \to \infty$. Collecting (17) and (19) we find the relaxation rate for weak Schwoebel effect:

$$\omega(q,\phi) \approx d_S\Delta F q^2 x_s \tanh\left(\frac{\kappa\ell}{2}\right) \tag{21}$$
$$-2D_s\Gamma q^2 \Lambda_q \left[\frac{1-\cos\phi}{\sinh(\Lambda_q\ell)} + \tanh\left(\frac{\Lambda_q\ell}{2}\right)\right].$$

From (18) and (21) we find the relaxation rate for the strong Schwoebel effect:

$$\omega(q,\phi) \approx \Delta F \frac{1}{\cosh(\kappa\ell)\cosh(\Lambda_q\ell)} \big[\cos\phi - \cosh(\kappa\ell)\cosh(\Lambda_q\ell) \tag{22}$$
$$+ \Lambda_q x_s \sinh(\kappa\ell)\sinh(\Lambda_q\ell)\big] - D_s\Gamma q^2 \Lambda_q \tanh(\Lambda_q\ell).$$

5.1. Step-pairing instability

If we look at (22) and (23), we realize that the relaxation rate ω vanishes at $q = 0$ for in-phase fluctuations, $\phi = 0$, only. This happens because when the fluctuations are not in phase, a uniform translation of the whole step train is not a neutral 'Goldstone mode'. The $q = 0$ contribution, which appears to be positive—destabilizing—in the presence of any non-zero Schwoebel effect, acts on homogeneous fluctuations, and it is therefore the term that is responsible for step bunching [16], that is, for the formation of close groups of steps separated by wide terraces. We showed in section 3 that the most unstable mode corresponds to $\phi = \pi$, and it acts so as to pair steps. When the crystal evaporates, and adatoms mostly attach to and detach from steps from the lower terrace, a step delimiting a narrow lower terrace moves slower than a step bordering a larger one, so the second quickly reaches the first, forming a step pair. Since a step pair is slower than a single step, it is rapidly reached by a third step, and so on. Repulsive elastic interactions obviously oppose the instability. In discussing the formation of bumps of finite wavelength in a step; that is, in discussing the stability of the step's shape at finite q, we may ignore the contribution from $q = 0$ if the characteristic time of the step-pairing instability, $1/\omega(q = 0, \phi = \pi)$, turns out to be small compared with the time $\tau_{ev} = \tau/c_{eq}^0$ needed to evaporate one layer. It is straightforward to see how $\omega(0, \pi)$ varies as a function of the strength of the Schwoebel effect. In fact,

$$\omega(0, \pi) = -2\Delta F \frac{d_S^2}{[d_S \cosh(\kappa\ell) + x_s \sinh(\kappa\ell)]^2}. \tag{23}$$

When there is no deposition flux, $F = 0$ and $\Delta F = -c_{eq}^0/\tau$. Letting now $\kappa\ell \ll 1$ and varying d_S in comparison to ℓ, we find the two limits

$$\omega(0, \pi) \approx 2\frac{c_{eq}^0}{\tau}\left(\frac{d_S}{\ell}\right)^2 \tag{24}$$

for $d_S \ll \ell$, and

$$\omega(0, \pi) \approx 4\frac{c_{eq}^0}{\tau} \tag{25}$$

for $d_S \gg \ell$. If we take the condition

$$\omega(0, \pi)\tau/c_{eq}^0 \geqslant 1 \tag{26}$$

as a criterion for an appreciable step bunching during UHV evaporation, this condition is seen to be fulfilled only for $d_S \geqslant \ell$: a strong Schwoebel effect or short distance between steps. This is seen well in computer simulations by Uwaha and Saito [6] which assume an infinite Schwoebel effect during evaporation.

Note that we have not explicitly included the effects of a DC electric current. However, we can discuss its role qualitatively, leaving a quantitative treatment for future work. Stoyanov [23] has shown that the current-induced facetting of a vicinal surface can be described assuming that the electric current exerts a force on the adatoms. This force acts so as to accumulate the adatoms, according to its sign, near the descending or the ascending side of each step; if the force is such as to increase the adatom concentration next to the descending side, and to lower it next to the ascending one, it increases the emission rate of adatoms from the step to the lower terrace. In other words, an increase in k_+ and a decrease of k_- result. The current effectively increases the Schwoebel effect. Step bunching occurring through a sequence of step pairings can then be expected. It is easy to realize

that an opposite current decreases the Schwoebel effect. Our discussion in terms of the Schwoebel barriers includes thus at least qualitatively the effect of the electric current.

Note, finally, that in the case of silicon (111) the conditions $\kappa\ell \ll 1$ and $q\ell \ll 1$ are satisfied. For completeness, we give the form of the relaxation rate in this limit, for any value of the Schwoebel length d_S and for $\Delta F = -c_{eq}^0/\tau$:

$$\omega(q,\phi) \approx \omega(0,\phi) - c_{eq}^0 D_s \left\{ \frac{(1-\cos\phi)}{(\ell+d_S)^3} \left[2\beta\gamma(\ell+d_S)^2 \right. \right. \tag{27}$$

$$\left. -\frac{1}{3}\beta\gamma(\ell+3d_S)(\ell+d_S)\Lambda_q^2\ell^2 + \frac{1}{6}(\ell+3d_S)d_S^2\kappa^2\ell^2 \right]$$

$$\left. +\beta\gamma\Lambda_q^2\ell + \frac{1}{2}\frac{d_S}{\ell+d_S}\kappa^2\ell^2 \right\} q^2 .$$

We can go on now to investigate the morphological stability of steps in various physical cases. As before, we address first the isolated step case. This is not very important for experiments, but allows comparison with published theoretical results. The case of a step train follows.

5.2. An isolated step: the weak Schwoebel effect

For an isolated step the notion of phase shift becomes meaningless. We can thus write simply $\omega(q)$ for the relaxation rate. In the weak Schwoebel effect $d_S \ll x_s$ in this case. From (22) one gets in the $\ell \to \infty$ limit

$$\omega(q) \approx -D_s c_{eq}^0 \left[\beta\gamma\Lambda_q + d_S\kappa \right] q^2 . \tag{28}$$

At small q, that is, for $\kappa \geqslant q$, one finds

$$\omega(q) \approx -D_s c_{eq}^0 \kappa \left[\beta\gamma + d_S \right] q^2 . \tag{29}$$

As shown by Uwaha and Saito [4] and Salditt and Spohn [9], it is possible to deduce from the previous formula the amplitude of step fluctuations, which turns out to be given by

$$\langle |\delta x_q|^2 \rangle = \frac{D_s c_{eq}^0 \kappa}{-\omega(q)} . \tag{30}$$

An approximate, but simple derivation of this formula is given in appendix 2. Following [4], it may be convenient to introduce a quantity ν_q, defined by

$$\langle |\delta x_q|^2 \rangle = \frac{k_B T}{\nu_q} . \tag{31}$$

At equilibrium we have $\langle |\delta x_q|^2 \rangle = k_B T/(\gamma q^2)$, where γ is the step stiffness. We can thus define a non-equilibrium effective step stiffness $\tilde{\gamma} \equiv \nu_q/q^2$. Since for $\kappa \geqslant q$ one has $\Lambda_q \approx \kappa$, we deduce from (29), (30) and (31)

$$\nu_q = \left[\beta\gamma + d_S \right] q^2 \tag{32}$$

and the effective step stiffness is $\tilde{\gamma} = \gamma + k_B T d_S$. Typically $\beta\gamma$ is of order 1 in atomic units. Since we are only supposing here $d_S \ll x_s$, $\tilde{\gamma}$ may be much larger than γ. This has been called 'dynamic step stiffening'.

5.3. An isolated step: the strong Schwoebel effect

This case has been considered by Uwaha and Saito [4]. From (23), taking $d_S \gg x_s$ and again $\kappa \geqslant q$, one finds

$$\omega(q) \approx -D_s c_{eq}^0 \left[\beta\gamma + \frac{1}{2} x_s \right] \kappa q^2 \tag{33}$$

or, according to (29) and (30),

$$\nu_q = \left[\beta\gamma + \frac{1}{2} x_s \right] q^2 . \tag{34}$$

This is Uwaha and Saito's result [4]. Note that d_S and x_s exchanged their roles compared to those in (32). Since x_s may be very large even at high T, the step stiffness is renormalized to an effective large value $\beta\tilde{\gamma} \approx x_s/2$.

We go on now to consider the more interesting case of a train of steps that are at mutual distance ℓ.

5.4. A train of steps: the weak Schwoebel effect

It is now more convenient to start from equation (28). We assume first that the Schwoebel effect is small, that is, $d_S \ll \ell$, as we expect it to be on Si(111) around 1000 °C. In this limit (28) becomes

$$\omega(q, \phi) \approx \omega(0, \phi) - D_s c_{eq}^0 \left[2(1 - \cos\phi)\beta\gamma + \frac{1}{3}\beta\gamma(2 + \cos\phi)\Lambda_q^2 \ell^2 + \frac{1}{2} d_S \kappa^2 \ell^2 \right] \frac{q^2}{\ell} . \tag{35}$$

This result is, to our knowledge, new.

We can estimate the quantities in brackets for Si(111) at 1273 K, and typical terrace sizes $\ell \approx 10^3$ au. In [2] we found an activation energy for the mass transport coefficient $c_{eq}^0 D_s$ of 1.1 eV on Si(111). For the activation energy of the mass evaporation rate, c_{eq}^0/τ, the value of 4.3 eV is well established [12, 10]. The diffusion length $x_s = \sqrt{D_s \tau}$ is then as usual taken as a simple activated quantity, with an activation energy of approximately 1.6 eV. At 1000 °C we thus find $1/\kappa = x_s \approx 10^6$ in units of the atomic spacing. According to our previous estimate we also have $d_S \approx 10^2$ at this temperature. Also, in a typical reflection electron microscopy experiment, fluctuations of wavelengths of the order of 10^4 are observed [11]. Hence, we find that only the first term in the bracket of equation (35) survives. Since the constant $\omega(0, \phi)$ is also negligible, we find

$$\omega(q, \phi) \approx -D_s c_{eq}^0 2(1 - \cos\phi)\beta\gamma \frac{q^2}{\ell} \tag{36}$$

as we found at equilibrium (compare equation (12)). It can be shown (see appendix 2) that step fluctuations are also of the same order of magnitude as (equilibrium) thermal fluctuations.

5.5. A train of steps: the strong Schwoebel effect

Let us consider now equation (28) for the strong Schwoebel effect, $d_S \ll \ell$. We readily find

$$\omega(q, \phi) \approx \omega(0, \phi) - c_{eq}^0 D_s \left[\beta\gamma \ell \Lambda_q^2 + \frac{1}{2}(2 - \cos\phi)\kappa^2 \ell^2 \right] q^2 . \tag{37}$$

Uwaha and Saito addressed the same problem (see (5) and (6) of [5]), but presumably they only considered in-phase fluctuation, as Bales and Zangwill did. In fact, we can recover

their result only if we let $\phi = 0$. We stress that the choice $\cos \phi = 1$ gives the correct answer on the stability of the step morphology only during deposition (more exactly, for $\Delta F > 0$). If adatom desorption is prevailing (i.e. if $\Delta F < 0$), the most unstable fluctuational mode is found for $\phi = \pi$. In this case, and for a strong Schwoebel effect, the $q = 0$ term is as large as the evaporation rate, and a step pairing instability occurs as soon as step flow starts. This is quite clearly seen in Uwaha and Saito's simulations (see figure 2.a of [5]). On the other hand, as we discussed at end of subsection 4.1, if the effect of a DC current is neglected (or if AC current heating is assumed), step-pairing does not occur on Si(111) if the miscut is small enough, or, equivalently, if the average step–step distance is large enough. We take this as evidence of a weak Schwoebel effect; that is, we conclude that $d_S \ll \ell$. The step-bunching instability that takes place between 1050 and 1250 °C when a DC electric current in the step-up direction is used to heat the sample [10, 23] may be physically justified in terms of the Schwoebel effect. The applied external potential would act so as effectively to increase the potential barrier for adatoms (if adatoms are positively charged, as Stoyanov assumes) at down-steps, finally leading to a increase of the Schwoebel effect for increasing T (increasing current).
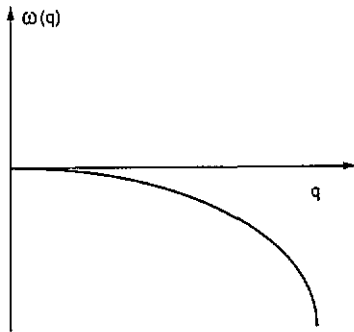

Figure 3. A sketch of the relaxation rate $\omega(q)$ for an isolated step. Note that the same qualitative form would be obtained for in-phase ($\phi = 0$) fluctuations of a train of steps.
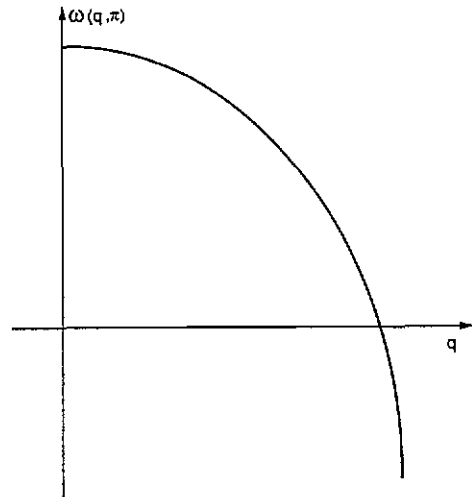
Figure 4. A sketch of the relaxation rate $\omega(q, \pi)$ for anti-phase ($\phi = \pi$) fluctuations of a train of steps.

6. On the shape of steps during evaporation

The main conclusion of this work is that a strong Schwoebel effect has consequences for step dynamics that are qualitatively quite different in the two cases of an isolated step and of a train of steps. In the former case, evaporation always has a smoothing action. The relaxation rate $\omega(q)$, qualitatively sketched in figure 3, is negative (and stabilizing) for any q. A strong Schwoebel effect increases the smoothing action, and an evaporating isolated step is stiffer for stronger asymmetry of the adatom kinetics at steps.

In the case of a train of steps, which is realized on any vicinal surface, any non-vanishing Schwoebel effect makes the anti-phase ($\phi = \pi$) fluctuation relaxation rate, $\omega(q, \pi)$, positive

at small q, with a finite value at $q = 0$ (see figure 4). This implies (i) that a step-pairing instability occurs, and (ii) that in a finite range, long-wavelength anti-phase fluctuations are also unstable. The destabilizing action is stronger for a stronger Schwoebel effect.

Therefore, we conclude that evaporation does not seem to introduce any special smoothing effect on the shape of steps. However, we are able to provide an argument for showing that a smooth step should remain smooth if it evaporates in the presence of a *weak* Schwoebel effect. Our argument states that, whereas at equilibrium the only relevant rate is adatom surface diffusion—and so equilibrium step fluctuations depend mainly on the mass diffusion constant $D_s c_{eq}^0$—during sublimation another timescale becomes important: the reciprocal of the desorption rate $1/\tau$ of adatoms from the surface. If the time needed to sublimate one layer, τ/c_{eq}^0, is smaller than the time needed to build a meander of given size and wavelength, then this meander will not appear. This mechanism should prevent long-wavelength and large-amplitude fluctuations from appearing, and would thus justify the persistence of straight steps during step flow evaporation, if they were originally straight. To be quantitative, let us take formula (35), which we found to be appropriate to the experimental situation under examination, and evaluate the time needed to create a fluctuation of wavelength L, for a typical phase shift, $\cos\phi = 0$. Then, let us compare it to $\tau_{ev} = \tau/c_{eq}^0$:

$$\frac{\tau_\ell(L)}{\tau_{ev}} \approx \frac{\kappa^2 L^2 \ell}{\beta\gamma}. \tag{38}$$

Equation (38) implies that only fluctuations of wavelengths smaller than L_c, where

$$L_c \approx \frac{x_s}{\sqrt{\beta\gamma\ell}} \tag{39}$$

will appear in a typical REM experiment. In turn, this implies that only amplitudes of the order of or smaller than h_c, where (see appendix 2)

$$h_c^2 \approx \frac{L_c}{\beta\gamma} \approx \frac{x_s\beta\gamma}{\sqrt{\ell}} \tag{40}$$

will be seen. Using the experimental values [2, 12] $\ell \approx 10^4$, $\beta\gamma \approx 1$, $x_s \approx 10^5$ at 1305 °C, we can estimate the order of magnitude of the largest fluctuation amplitude that one should see:

$$h_c \approx 10^2. \tag{41}$$

The resolution of REM allows one to see fluctuations of amplitude larger than ≈ 50 Å [24]. The 'critical' amplitude h_c lies near the limit of the instrumental resolution (note that several numerical factors have been omitted in the estimation of h_c). Summarizing our argument, larger amplitudes than h_c are kinetically hindered (the corresponding fluctuations have no time to appear), and smaller amplitudes lie below the instrumental resolution. The steps thus look to be smooth.

7. Conclusions

We have computed the relaxation rate of small sinusoidal perturbations in the shape of steps on a sublimating vicinal surface. A finite asymmetry in the kinetics coefficients of adatom attachment and detachment at step sticking sites (the Schwoebel effect) has been included. The latter has been recently held to be responsible of the observed phenomenon of kinetic step smoothing during the sublimation of the crystal face. We have discussed the plausible values of this asymmetry at high temperature, where sublimation takes place, with special

reference to Si(111) where obervations have been made. We have concluded that, at least in this case, the Schwoebel effect may not be sufficiently strong to explain step smoothing. This conclusion especially follows from observations of step fluctuations at lower temperature, which are not compatible with a strong Schwoebel effect. We propose that step smoothing is due to the finite time needed to create a fluctuation of given wavelength along a step: if this time is longer than the time needed to sublimate one surface layer, that type of fluctuation cannot take place. This mechanism is operative independently of any asymmetry of step kinetics coefficients, is the stronger for higher evaporation rates and larger terrace sizes. Our argument is very naive and only explains why straight steps remain straight, but it has the advantage of not invoking an enormously strong Schwoebel effect, of which there is no evidence, at least for steps on a silicon (111) face.

Acknowledgments

Appendix A.

In this appendix we give the derivation of the relaxation rate in the case of a train of steps with arbitrary phase shift ϕ. We shall denote by δx_m the step excursion about the straight step position labelled by the integer m. The adatom density $c(r)$ is replaced by u, where $u(r) = c(r) - F\tau$. Equation (1) becomes

$$\nabla^2 u - \frac{u}{x_s^2} = 0 . \tag{A1.1}$$

On both sides of the step we have the boundary conditions

$$D_s \left[\frac{\partial u}{\partial n} \right]_+ = k_+ [u + \tau \Delta F - \Gamma \kappa_m]_+ \tag{A1.2}$$

$$-D_s \left[\frac{\partial u}{\partial n} \right]_- = k_- [u + \tau \Delta F - \Gamma \kappa_m]_- \tag{A1.3}$$

where the '+' and '−' signs refer to the lower and upper terraces respectively, $\partial u / \partial n$ stands for the normal derivative, with the normal pointing in the positive x direction, k_+ and k_- are the sticking coefficients from the lower and the upper terraces respectively, and κ_m is the step curvature, defined as positive for a convex profile.

Once the concentration gradients on both sides of the step are known we can evaluate the step velocity by means of mass conservation law at the step

$$v_n = D \left\{ \left[\frac{\partial u}{\partial n} \right]_+ - \left[\frac{\partial u}{\partial n} \right]_- \right\} . \tag{A1.4}$$

The set (A1.1)–(A1.4) completely describes the step dynamics.

This set admits a straight-step solution moving at a constant speed V along the x-direction. The zeroth-order solution is a train of equidistant steps separated by the distance ℓ. The general solution for the density is given by

$$u_0 = A \cosh(\kappa x) + B \sinh(\kappa x). \tag{A1.5}$$

The two constants of integration A and B are easily obtained by making use of the two boundary conditions (A1.2) (A1.3). The result is

$$A = -k_+\tau\Delta F\frac{k_-\sinh(\kappa\ell) + \kappa D_s[\cosh(\kappa\ell) + k_-/k_+]}{[(\kappa D_s)^2 + k_+k_-]\sinh(\kappa\ell) + \kappa D_s(k_+ + k_-)\cosh(\kappa\ell)}$$

$$B = k_+\tau\Delta F\frac{\kappa D_s\sinh(\kappa\ell) + k_-[\cosh(\kappa\ell) - 1]}{[(\kappa D_s)^2 + k_+k_-]\sinh(\kappa\ell) + \kappa D_s(k_+ + k_-)\cosh(\kappa\ell)}. \tag{A1.6}$$

Making use of (A1.4), we obtain the step velocity

$$V = x_s\Delta F\frac{2[\cosh(\kappa\ell) - 1] + (d_+ + d_-)\kappa\sinh(\kappa\ell)}{[1 + d_+d_-\kappa^2]\sinh(\kappa\ell) + (d_+ + d_-)\kappa\cosh(\kappa\ell)}. \tag{A1.7}$$

The linear stability of the solution is analysed by looking for perturbations of the step profile in the form

$$\delta x_m = A_m e^{iqy+\omega t} + \text{CC} \tag{A1.8}$$

where A_m stands for a complex amplitude of the mth step, ω is the amplification (or damping) rate that we whish to determine, and q is the perturbation wavenumber. The adatom density variation in response to step perturbations is written as

$$u_{1m} = [\alpha_m\sinh(\Lambda_q x) + \beta_m\cosh(\Lambda_q x)]e^{iqy+\omega t} + \text{CC} \tag{A1.9}$$

where $\Lambda_q = \sqrt{q^2 + \kappa^2}$. The adatom density in equation (A1.9) above is the one on the lower terrace, just ahead of the step. In order to compute the constants α_m and β_m we have to use (A1.2) and (A1.3). Care should be taken when using these boundary conditions. Equation (A1.2) should be written at $x = \delta x_m$. Since u_0 (the straight-step solution) is of order zero in the perturbation, we must take into account terms like $\delta x_m \partial u_0/\partial x(x = 0)$. On the other hand the constant α_m and β_m must be evaluated within the terrace under consideration ($\delta x_m < x < \ell + \delta x_{m+1}$). This means that the second condition (A1.3) is to be used at $x = \ell + \delta x_{m+1}$ (here also we have a contribution from u_0, as mentioned above). If one takes care of these points the calculation is straightforward and yields the following expressions:

$$\alpha_m = -(\mathcal{D}_q x_s)^{-1}\{\mathcal{B}_q A_{m+1} + k_+^{-1}\mathcal{A}_q A_m[D_s\Lambda_q\sinh(\Lambda_q\ell) + k_-\cosh(\Lambda_q\ell)]\}$$

$$\beta_m = k_+^{-1}[D_s\alpha_m\Lambda_q + \kappa\mathcal{A}_q A_m] \tag{A1.10}$$

where $\mathcal{A}_q$, $\mathcal{B}_q$ and $\mathcal{D}_q$ are given by

$$\mathcal{A}_q = k_+\Gamma x_s q^2 - D_s\tau\Delta F\Psi_q\left\{\left(1 + \frac{d_-}{d_+}\right)\sinh(\kappa\ell) + \kappa d_-\cosh(\kappa\ell)\right.$$

$$\left. + \frac{1}{\kappa d_+}[\cosh(\kappa\ell) - 1] + \kappa d_+\right\}$$

$$\mathcal{B}_q = -k_-\Gamma x_s q^2 + D_s\tau\Delta F\Psi_q\left\{\frac{1}{\kappa d_-} - \kappa d_- - \left[\frac{1}{\kappa d_-} + \kappa d_+\right]\cosh(\kappa\ell)\right.$$

$$\left. - \left[1 + \frac{d_+}{d_-}\right]\sinh(\kappa\ell)\right\}$$

$$\mathcal{D}_q = D_s\left\{\Lambda_q\left(1 + \frac{d_-}{d_+}\right)\cosh(\Lambda_q\ell) + \sinh(\Lambda_q\ell)[d_- + 1/d_+\Lambda_q^2]\right\} \tag{A1.11}$$

where we also used

$$\Psi_q = \{[\kappa d_+d_- + x_s]\sinh(\kappa\ell) + (d_+ + d_-)\cosh(\kappa\ell)\}^{-1}. \tag{A1.12}$$

Note that both amplitudes A_m and A_{m+1} enter in the determination of u_m. In order to find the relaxation rate, we make use of (A1.4). For that purpose we need to evaluate the normal derivative of u at $x = \delta x_m - 0^+$. This can be done by finding the adatom density u_{m-1} in the domain $[\delta x_{m-1} - \ell < x < \delta x_m]$. It is easy to see that u_{m-1} is given by an expression analogous to that for u_m where in α_m and β_m we substitute for the amplitudes of the deformations A_m and A_{m+1} with A_{m-1} and A_m, respectively. Having determined the expression for the density on both sides of the step considered, we are in a position to use equation (A1.4). The calculation involves tedious algebra, and the result can be finally written in the following form:

$$\omega A_m = -q^2 \Gamma D \Lambda_q \frac{[2\cosh(\Lambda_q \ell) + \Lambda_q(d_+ + d_-)\sinh(\Lambda_q \ell)]A_m - A_{m-1} - A_{m+1}}{\Lambda_q(d_+ + d_-)\cosh(\Lambda_q \ell) + (1 + d_+ d_- \Lambda_q^2)\sinh(\Lambda_q \ell)}$$
$$+ \Delta F\{(d_- - d_+)\{\Lambda_q(d_+ + d_-)[x_s \Lambda_q \sinh(\Lambda_q \ell)\sinh(\kappa \ell)$$
$$- \cosh(\Lambda_q \ell)\cosh(\kappa \ell)] + \sinh(\Lambda_q \ell)[\cosh(\kappa \ell) - 1]x_s^2 q^2\}A_m$$
$$+ \Lambda_q[d_-^2 A_{m+1} - d_+^2 A_{m-1}]\cosh(\kappa \ell) - \Lambda_q[x_s(d_+ + d_-)\sinh(\kappa \ell)$$
$$+ (d_+ d_- + x_s^2)\cosh(\kappa \ell) - x_s^2](A_{m-1} - A_{m+1})\}/D(q) . \tag{A1.13}$$

where $D(q)$ is given, as in the body of the paper, by

$$D(q) = [\Lambda_q(d_+ + d_-)\cosh(\Lambda_q \ell) + (1 + d_+ d_- \Lambda_q^2)\sinh(\Lambda_q \ell)] \tag{A1.14}$$
$$\times [(d_+ + d_-)\cosh(\kappa \ell) + (\kappa d_+ d_- + x_s)\sinh(\kappa \ell)].$$

Equation (A1.13) is what we are looking for. It relates the dynamics of a step to its neighbours, when each step is moving in an asynchronized way with respect to the others. The general solution equation (A1.13) is given by $A_m = C e^{im\phi}$ (for periodic boundary conditions) where ϕ is an arbitrary real number measuring the phase shift between two consecutive steps, and C is a real constant. Inserting this expression into equation (A1.13) we obtain the relaxation rate as a function of the phase ϕ. Writting $\omega = \omega_r + i\omega_i$, we have

$$\omega_r = -q^2 \Gamma D_s \Lambda_q \frac{2[\cosh(\Lambda_q \ell) - \cos(\phi)] + \Lambda_q(d_+ + d_-)\sinh(\Lambda_q \ell)}{\Lambda_q(d_+ + d_-)\cosh(\Lambda_q \ell) + (1 + d_+ d_- \Lambda_q^2)\sinh(\Lambda_q \ell)}$$
$$+ \Delta F(d_- - d_+)\{\Lambda_q(d_+ + d_-)[x_s \Lambda_q \sinh(\Lambda_q \ell)\sinh(\kappa \ell)$$
$$- \cosh(\Lambda_q \ell)\cosh(\kappa \ell) + \cos(\phi)] + \sinh(\Lambda_q \ell)[\cosh(\kappa \ell) - 1]x_s^2 q^2\}/D(q)$$
$$\omega_i = \Lambda_q \Delta F \sin(\phi)\{2x_s(d_+ + d_-)\sinh(\kappa \ell) + 2(d_+ d_- + x_s^2)\cosh(\kappa \ell)$$
$$+ d_-^2 + d_+^2 - 2x_s^2\}/D(q). \tag{A1.15}$$

It can be checked that for collective fluctuations ($\phi = 0$) we recover the result of Bales and Zangwill. Letting $A_{m-1} = A_{m+1} = 0$ we find the relaxation rate of a step fluctuating between two straight neighbours, which corresponds to $\cos\phi = 0$. Note that here we have an imaginary part (which vanishes for $\phi = 0$ or $\phi = \pi$ only). This means that propagative effects enter in the way by which perturbations are transported from one step to another.

Appendix B.

The amplitude of step fluctuations has been calculated in various cases by Uwaha and Saito [4] and by Salditt and Spohn [9]. It may be convenient for the reader to have all cases gathered together in the following simplified evaluation. The principle is the same as in [2].

Instead of the fluctuation amplitude, equation (31), we shall evaluate the average width $h(L)$ of a 'bump' of length $L \approx 1/q$ along a step (figure A1). An elementary calculation
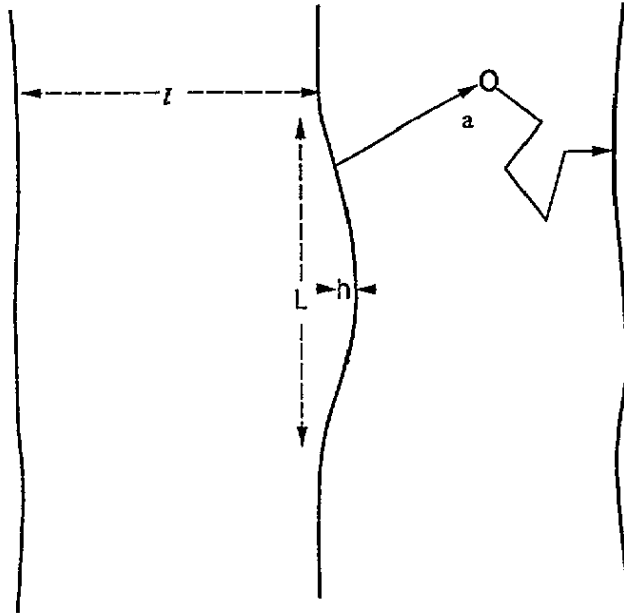
Figure A1. A schematic description of a step fluctuation ('bump') of length L and width h, in a train of steps with average separation ℓ. Adatom a is diffusing from a step to the neighbouring one, where it will stick if the Schwoebel effect is weak enough ($d_S \ll \ell$, small asymmetry or large terrace).

or dimensional analysis show that $h(L)$ is related to (31) through

$$h^2(L) \approx \frac{1}{L} \langle |\delta x_{1/L}|^2 \rangle . \tag{B1.1}$$

In order to evaluate $h^2(L)$, we shall compute the mean square width at time t, $h^2(L, t)$, neglecting the restoring force. The latter is approximately $h/\omega(1/l)$, where $\omega(q)$ is the quantity computed throughout this article in various cases. Then, the steady-state value $h^2(L)$ can be evaluated by requiring the time t to be of the order of the relaxation time $1/\omega(1/L)$, so that

$$h^2(L) \approx h^2(L, 1/\omega(1/L)) . \tag{B1.2}$$

In order to evaluate $h^2(L, t)$, we write that the fluctuation of the area of the bump, $L^2 h^2(L)$, is equal to the fluctuation of the total number $N(t)$ of atoms sticking to the bump during time t. If the various sticking events may be considered as independent, one has from elementary probability theory that the fluctuation is of the order of the square root of N, or, equivalently,

$$L^2 h^2(L) \approx N(t) . \tag{B1.3}$$

The adatoms sticking to a portion of step of length L are those contained in a surface area of size $L \times L_s$. The length L_s is, in general, a function of L, ℓ and κ, as we shall see later. The number of these adatoms is equal to $c_{eq}^0 L L_s$, and they spend, on average, inside that area a time $\tau^* \approx D_s/\inf(L^2, L_s^2)$. Sticking events whose time separation is larger then τ^* can be considered as independent. On the other hand, the number of atoms that stick to

the step segment during a time τ^* is of order $c_{eq}^0 L L_s$, the total number of atoms inside the area $L \times L_s$. Finally, $N(t)$ should be proportional to t for $t \gg \tau^*$. Therefore,

$$N(t) \approx c_{eq}^0 L L_s \frac{t}{\tau^*} . \tag{B1.4}$$

All these formulae are derived in [2] in more detail. There, we used them to compute in a simple way $\omega(q)$ at equilibrium. Here, since we know $\omega(q)$ from the (less simple) calculations of appendix 1, we use this knowledge to evaluate the fluctuation (B1.1).

(i) *An evaporating isolated step when* $q \ll \kappa$. This is the case treated in subsections 4.2 and 4.3. The values of the parameters in (B1.4) are $L_s \approx 1/\kappa$ and $1/\tau^* \approx D_s \kappa^2$. Inserting them into (B1.4) yields

$$N(t) \approx c_{eq}^0 D_s \kappa L t . \tag{B1.5}$$

Letting $t \approx 1/\omega(1/L)$ and inserting into (B1.3) yields

$$h^2(L) \approx \frac{D_s c_{eq}^0 \kappa}{L\omega(1/L)} . \tag{B1.6}$$

Using $\omega(q)$ given in equation (29), one obtains for a weak Schwoebel effect

$$h^2(1/q) \approx \frac{1}{(\beta\gamma + d_S)q} . \tag{B1.7}$$

Equation (32) is then obtained from equations (B1.1) and (31).

If the Schwoebel effect is strong, then equation (34) is obtained from (B1.6) through (B1.1), (31) and (33).

(ii) *An evaporating vicinal surface with a weak Schwoebel effect.* This is the situation considered in subsection 4.4. As discussed in [2], the values of the parameters in (B1.4) are $L_s = \ell$ and $1/\tau^* \approx D_s/\ell^2$. Inserting them into (B1.4) yields $N(t) \approx D_s c_{eq}^0 L t/\ell$. Then equation (B1.3) gives

$$h^2(L) \approx \frac{D_s c_{eq}^0}{\ell L\omega(1/L)} . \tag{B1.8}$$

When using $\omega(q, \phi)$ in equation (36), $\cos\phi$ can be replaced by its average value 0. Therefore one obtains

$$h^2(L) \approx \frac{L}{\beta\gamma} \tag{B1.9}$$

which is the equilibrium value.

(ii) *An evaporating vicinal surface with a strong Schwoebel effect.* This is the situation considered in subsection 4.5. As discussed in [2], the values of the parameters in (B1.4) are $L_s = \ell$ and $1/\tau^* \approx D_s/L^2$. Inserting them into (B1.4) yields $N(t) \approx D_s c_{eq}^0 \ell t/L$. Then equation (B1.3) gives

$$h^2(L) \approx \frac{D_s c_{eq}^0 \ell}{L^3 \omega(1/L)} . \tag{B1.10}$$

When using $\omega(q, \phi)$ given in equation (37), $\cos\phi$ can be replaced by its average value 0. We get, to order of magnitude,

$$h^2(L) \approx \frac{\ell L}{\kappa^2 L^4 + \ell \kappa^2 L^2 (\beta\gamma + \ell) + \beta\gamma\ell} . \tag{B1.11}$$

Inserting experimental values appropriate to silicon (111) at 1300 °C ($\kappa \approx 10^{-5}$, $\ell \approx 10^4$, $\beta\gamma \approx 1$) and considering long ($L \approx 10^4$) and short ($L \approx 10^2$) wavelengths, we find, respectively,

$$h^2(L) \approx \frac{L}{\beta\gamma} \tag{B1.12}$$

which is the equilibrium fluctuation amplitude for short wavelengths, and

$$h^2(L) \approx \frac{\ell}{\kappa^2 L^3} \tag{B1.13}$$

for long wavelengths, where the $\omega(0)$-term dominates and step bunching is expected.

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