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## Fractal terraces in MBE growth

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**Abstract.** — MBE growth on high symmetry surfaces depends to some extent on the shape of the terraces which form the embryos of each new atomic layer. The conditions for formation of compact or fractal terraces are discussed in two limits: large terraces, to which a continuous description applies, and small terraces, which require that the discrete nature of matter be taken into account. In both cases it is found that terraces are fractal if and only if diffusion of adatoms along steps is much slower than on the flat surface at the same temperature. Precise conditions are given which should be accessible to experiment.

### 1. Introduction.

Progress in the understanding of fractal phenomena has been a major contribution of Rammal and his coworkers. In the present work we address the possibly fractal shape of terraces on a crystal surface growing by molecular beam epitaxy (MBE).

An important problem in this field is the typical size  $\ell$  of the islands on the growing surface. Indeed, the measurement of  $\ell$  can give information on the surface diffusion constant  $D_s$ , and even allows a quantitative determination of  $D_s$  itself, at least in simple cases [1]. Domains of size  $\ell$  may also form in the surface layer if, for instance, the adsorbate has a variety of possible equivalent superstructures.

In most theoretical investigations [2, 3, 4], the island shape has been assumed to be compact, *i.e.* not fractal. However, fractal islands have been observed by scanning tunneling microscopy (STM) in the deposition of Pt on Pt at 205 K [5] and of Au on Ru (0001) [6] at room temperature. Fractal islands seem to be observed only at (relatively) low temperatures. Si on Si (001) has been investigated also at low temperature by Mo *et al.* [1], but the situation is complicated because both surface diffusion and sticking-to-step are highly anisotropic.

The mechanism underlying island fractality is the so-called diffusion limited aggregation (DLA) [7, 8, 9]. DLA is a growth process where atoms diffuse from far away and stick to the

first growing island they find, at the very place where they touch it for the first time. Because of fluctuations, islands develop tips, most incoming atoms reach islands at tips, and thus the tips become longer and the fractal shape appears.

In MBE growth this scheme is oversimplified because atoms can also be deposited right on top of an island. This favours a compact shape, since atoms diffuse to the edge of the island both from outside and inside. For small islands, however, the flux from inside is negligible.

A theory describing the shape of growing islands on a high symmetry crystal surface will be outlined in the present article. Analogies and differences with existing theories will be discussed.

## 2. Magnitude of $\ell$ for fractal islands.

The effect of terrace edge fractality on the typical length  $\ell$  has been discussed by Villain *et al.* [10] and will only briefly be summarized here.

Evaporation will be neglected since the temperatures of interest are low. The basic picture is that atoms falling on the surface mostly land on terraces between steps. These mobile "adatoms" diffuse (with a diffusion constant  $D_s$ ) until they reach a step and are incorporated there. Sometimes, however, they meet another diffusing adatom before reaching a step, and they may therefore form a pair. Pairs are supposedly immobile and stable at the low temperatures we are considering.

The density  $\rho_1$  of diffusing adatoms per absorption site may be easily evaluated. Adatoms have an average lifetime  $\tau_1$  before being incorporated into a step, and  $\rho_1$  is equal to the product of  $\tau_1$  times the rate  $F$  of incoming atoms per site. That is,  $\rho_1 = F\tau_1$ . During its lifetime  $\tau_1$  an adatom diffuses a distance of the order  $\sqrt{D_s\tau_1}$ . By definition, this length has to be identified with the typical distance between steps,  $\ell$ . Thus,  $\ell^2 \approx D_s\tau_1$  and

$$\rho_1 \approx F\ell^2/D_s. \quad (1)$$

The sign " $\approx$ " means "of the order of magnitude of". We can now evaluate the rate of pair formation. Diffusing on a two dimensional surface, an adatom visits approximately (*i.e.* disregarding logarithmic corrections)  $D_s\tau_1 \approx \ell^2$  distinct sites. The unit of length is the interatomic distance. Each of these  $\ell^2$  sites has, on average, a probability  $\rho_1$  of being occupied, and each adatom has therefore a probability  $\rho_1\ell^2$  of forming a pair. The rate  $1/\tau_{\text{nuc}}$  of pair formation (*nucleation*) per site is obtained by multiplying by the rate  $F$  of atoms falling on each site

$$1/\tau_{\text{nuc}} \approx F\rho_1\ell^2 \approx F^2\ell^4/D_s. \quad (2)$$

Now, since  $\ell$  is, among other things, the typical island size before coalescence, there should be one and only one pair formation in an area  $\ell^2$  during the whole life of an island. The corresponding lifetime is equal to  $1/F$  times the island density, which is  $1/\ell^{d_f}$  for fractal islands with fractal dimension  $d_f$ . DLA in 2 dimensions gives  $d_f = 1.7$ . Therefore

$$1/\tau_{\text{nuc}} \approx F/\ell^{d_f} \quad (3)$$

Equations 2 and 3 yield

$$\ell \approx \left( \frac{D_s}{F} \right)^{\frac{1}{4+d_f}} \quad (4)$$

For non-fractal terraces,  $d_f = 2$  and [3]

$$\ell \approx \left( \frac{D_s}{F} \right)^{1/6} \quad (5)$$

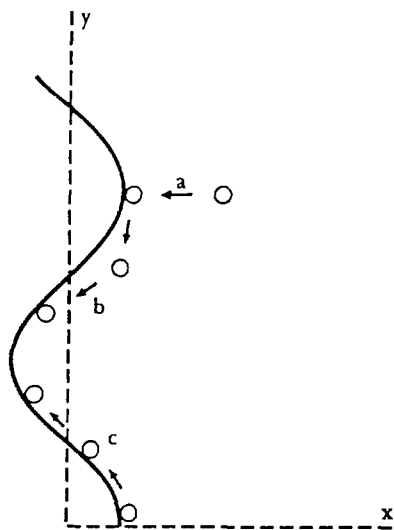


Fig.1. — An adatom approaching a terrace (a) sticks preferentially at a tip, and can then be remitted (b) and diffused into a gulf. It may also diffuse straightforwardly to a gulf along the step edge (c).

### 3. Stabilizing effects.

We will now discuss the stability of non-fractal shapes during growth. Compact, non-fractal forms are stabilized by the rearrangement of atoms within a terrace, which is a negligible effect only at very low temperature. Indeed, compact shapes are energetically favoured because they minimize the surface tension energy. Thus, atoms bound at the edge of an island (hereafter called *gradatoms*) will diffuse along the same edge and tend to repair fractality. This will be called edge- or step-diffusion rearrangement.

The word “gradatom” (from the Italian *gradino*, the French *gradin* or the Latin *gradus*) is preferred to “stepatom” to avoid the initial “s” which we prefer to keep for “surface”.

A similar healing effect is due to atoms temporarily ejected from tips which diffuse to a deeper place, or to atoms which do not stick at first hit to the island and diffuse then to a site inside some deep gulf (Fig. 1). This will be called off-cluster rearrangement.

### 4. Small islands at low temperature.

Fractal islands actually observed during heteroepitaxy of Au on Ru(0001) [6] show dendritic arms with a typical width  $r_c$  of some ten nanometers. The very simple argument that follows suggests that this width is mainly controlled by the diffusion constant  $D_g$  of gradatoms. We assume low temperature, and that two gradatoms meeting along the edge of a terrace form the immobile and stable nucleus of a new layer for the lateral growth of the terrace.

Imagine for instance a small island growing on (001) face. At first it will keep its most stable form, that is, a square (Fig. 2a), even though the adatoms mainly stick to the corners. If we assume that they all stick to the corners, for a linear dimension  $r$  sufficiently large the island will take a cross-like shape (Fig. 2b). This shape is observed in Hwang *et al.* experiment (but with a substrate of hexagonal symmetry!). For a smaller size  $r < r_c$  gradatoms have the

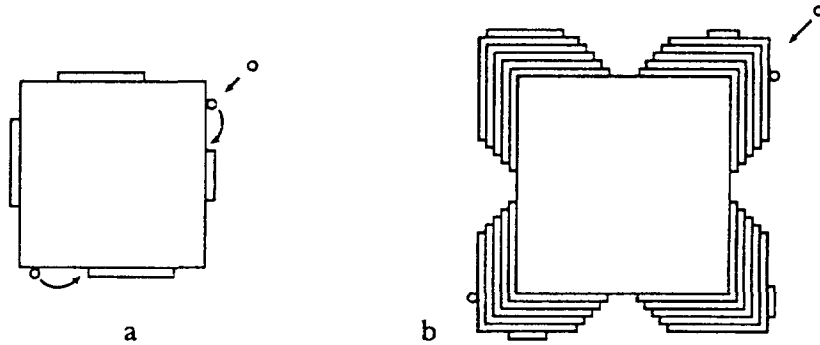


Fig.2. — a) Stable form of a small terrace at low temperature. b) Dendritic instability under growth conditions.

time to diffuse to the center of an edge and to form there a stable nucleus. Before reaching a newly formed layer, gradatoms have to cover a distance of the order of the linear size  $r$  of the island. This takes a time  $\tau \approx r^2/D_g$ . The density of gradatoms on the side of an island is  $\rho_1^g = \tilde{F}\tau$ , where  $\tilde{F}$  is the rate of incoming adatoms per unit site on the island edge; thus  $\tilde{F} = F\ell^2/r$ . Therefore,  $\rho_1^g = F\ell^2 r/D_g$ . The island undergoes stable growth if the gradatoms are able to cover a distance of order  $r$  without meeting another gradatom. This implies  $r\rho_1^g < 1$ . This condition is not sufficient, because it only requires that one particular gradatom be little likely to form a pair before reaching an adsorption site (kink). In fact, we must ask the same requirement from a number of gradatoms of the order of  $r$ . Since for each gradatom the probability of pair formation is  $r\rho_1^g$  for  $r$  gradatoms it is  $r^2\rho_1^g$ . Therefore the critical linear size  $r_c$  beyond which the island growth is unstable is given by

$$r_c^3 \ell^2 = D_g/F. \quad (6)$$

$\ell$  is directly accessible to measure by scanning tunneling microscopy, for instance. Assuming for  $\ell$  equation (5), one finds

$$r_c \approx \left( \frac{D_g^3}{F^2 D_s} \right)^{\frac{1}{5}} \quad (7)$$

Equation (6) is meaningful only if  $r_c < \ell$ , that is if  $r_c^3 \ell^2 < \ell^5$ , and according to equation (5)

$$F > D_g^6/D_s^5. \quad (8)$$

Since  $F$  is known and both  $r_c$  and  $\ell$  are directly measurable with STM, equation (6) allows an experimental determination of the diffusion constant of gradatoms  $D_g$ . In the case of Au on Ru [6], the pictures taken at 300 K suggest  $r_c \approx 10$  nm,  $\ell \approx 100$  nm, with  $F^{-1} = 30$  s, which imply  $D_g \approx 10^6$  s $^{-1}$ . Assuming an activated form

$$D_g = D_g^0 \exp(-\beta W_g) \quad (9)$$

and a typical value of the prefactor  $D_g^0 \approx 10^{12}$  s $^{-1}$ , our estimate yields an activation energy  $W_g$  of the order of 0.35 eV, which is not unreasonable. Equation (8) implies that the surface diffusion constant has a lower bound of approximately  $10^7$  s $^{-1}$  at room temperature, which in turn gives an activation energy  $W_{sd} \approx 0.29$  eV for surface diffusion.

The simple argument above gives only an estimate of the typical length beyond which an island is no longer able to retain its polygonal shape. What happens when the size exceeds  $r_c$  can only be studied by numerical simulations. This has been done by Xiao *et al.* [11]. They assumed off-cluster diffusion due to the weakness of the sticking coefficient of adatoms on islands. Since this type of sticking coefficient is assumed to increase with temperature, they found that island fractality decreases with temperature, in contrast with experiments on homoepitaxial Pt [5].

### 5. Classical linear stability analysis.

In the previous section, the appearance of a fractal shape has been attributed to the fact that a small tip on an island tends to become longer because incoming adatoms land preferentially on that tip. This instability is not typical of a random process only, and its competition with stabilizing effects can be studied within a deterministic growth scheme. In the case of a planar surface this was done by Mullins and Sekerka [12]. We shall not go beyond their analysis, but we shall adapt it to our purpose. The argument (given more in detail in Appendix 1) can be summarized as follows. Away from a step, the density  $\rho$  of adatoms obeys the equation  $\nabla^2 \rho + F = 0$ . The boundary condition along a step in the case of perfect sticking is  $\rho = 0$ . The growth rate on each point of the surface is proportional to the current  $\mathbf{j} = -D\nabla\rho$ . From these equations, it can be deduced that a sinusoidal perturbation  $\delta x \sim \cos(ky)$  on a straight step parallel to the  $x$  direction generates a destabilizing term  $\delta \dot{x}_{\text{dest}} \approx \text{Const} \times k \delta x$  due to atoms diffusing toward the step. This term is proportional to the wavevector  $k$  of the perturbation as in the analysis of Mullins and Sekerka. The constant is proportional to the flux  $F$ , but also depends on the average distance  $\ell$  between islands, and on the radius  $R$  of the cluster of interest. More precisely, the island collects in the time  $t$  the atoms falling in the area  $\ell^2$  during that time. Therefore, the number of atoms reaching a given island per unit time is of order  $F\ell^2$ , and the flux per unit step length is obtained dividing by  $R$ . Finally

$$\delta \dot{x}_{\text{dest}} \approx \frac{F\ell^2}{R} \times k \delta x \quad (10)$$

The stabilizing term due to off-cluster rearrangement is [12, 13]

$$\delta \dot{x}_{\text{stab}} \approx -\text{Const} \times k^3 \delta x. \quad (11)$$

This formula is rederived in Appendix 1. Instability occurs if the wavelength  $1/k$  of the perturbation is larger than  $1/\sqrt{\ell F}$ , multiplied by some temperature dependent constant. Of course,  $1/k$  has to be smaller than  $\ell$ . Thus the instability arises if  $\ell^3 > \text{Const}/F$ . Replacing  $\ell$  by equation (5), fractal islands are seen to be favoured by a strong deposition flux  $F$ . The case of step-diffusion will now be investigated in more detail. The stabilizing term  $\delta \dot{x}_{\text{stab}}$  satisfies a continuity equation

$$\delta \dot{x}_{\text{stab}} = -\frac{\partial j}{\partial y} \quad (12)$$

The current  $j$  along the step is related to the gradient of the chemical potential  $\mu$  by the relation

$$j = -\Lambda_g \frac{\partial \mu}{\partial y} \quad (13)$$

The chemical potential  $\mu$  is proportional to the curvature,

$$\mu \simeq -C \frac{\partial^2 x}{\partial y^2}. \quad (14)$$

Thus,

$$\delta \dot{x}_{\text{stab}} = -C \Lambda_g k^4 \delta x. \quad (15)$$

The kinetic coefficient  $\Lambda_g$  which appears in equation (15) is given by a Kubo formula, which may be seen to be (Appendix 2)

$$\Lambda_g = \frac{1}{NT} \sum_m \int_0^\infty dt \langle v_m(t) v_m(0) \rangle \quad (16)$$

where  $m$  labels the adatoms and the  $v_m$ 's are their velocities. The correlation function is easily computed (Appendix 2) and one obtains

$$\Lambda_g = \frac{1}{T \tau_k} \quad (17)$$

where  $1/\tau_k$  is the rate of emission of gradatoms from a given kink. The constant  $C$  depends on the orientation of the step. For the easy orientation, it can be shown that  $C = \frac{T}{2} \exp(\beta W_0)$  where  $W_0$  is the energy of a kink. Therefore

$$\Lambda_g C \approx \frac{1}{\tau_k^0} \exp\{\beta(W_0 - W_k)\}. \quad (18)$$

It is more relevant to consider the uneasy orientation. Indeed, if the island has the shape of a rounded polygon at equilibrium, the tips are expected to develop at the corners since the latter are preexisting tips. Then, in a square lattice,  $C = 4T$  and

$$\Lambda_g C \approx \frac{1}{\tau_k^0} \exp\{-\beta W_k\} \quad (19)$$

In any case this is small at low temperature.

A straight step is unstable if

$$\frac{F \ell^2}{R k^3} > \Lambda_g C. \quad (20)$$

Since  $R < \ell$ , the previous result implies  $F \ell^4 > \Lambda_g C$ .

At low temperature,  $\ell \approx (D_s/F)^{1/6}$  and instability can occur only if

$$F/D_s > (\Lambda_g C/D_s)^3 \quad (21)$$

Bales and Zangwill [14] and Uwaha and Saito [15] have also investigated the stability of a straight step on a growing crystal. They took into account evaporation, which modifies pretty much the above analysis. Also, they have assumed the presence of the so-called Schwoebel effect. This effect consists in the impossibility (or difficulty) for adatoms coming from the top of a terrace to go down the step because of a potential barrier. The existence of such a barrier in certain cases, postulated by Schwoebel [16], is supported by microscopic calculations [17]. The Schwoebel effect is a necessary condition for the instability of an infinite step, but is not essential for small islands since, anyway, the number of atoms coming from inside the island is much smaller than the number of those coming from outside. On the other hand, evaporation of adatoms from the surface can be neglected at those low temperatures where fractal islands are expected according to calculations and have been actually observed.

As seen above, most of the analytic treatments address the stability of infinitely long straight steps. Of course, small islands have not infinitely long straight edges. The case of a circular

island can be treated. But islands are not circular either at low temperature, even at equilibrium. The equilibrium shape at temperatures of interest is something like a square with severely rounded corners. The stability of such a shape is a nonlinear problem. A numerical treatment has been given by Ben Amar [18].

## 6. Conclusion.

We have demonstrated that islands on a surface growing by MBE can be fractal at low temperature, in agreement with experiment. According to our formulae, fractality disappears at low growth rates. This does not seem to be confirmed by experiments [6]. This may be an effect of a low sticking coefficient.

Fractality of terraces is predicted to increase the average terrace size  $\ell$ . This may be expected to have effects on the quality of crystals, but these effects depend on the crystal structure. The influence of the Schwoebel effect on the crystal quality is also expected to be important, but this is left for further investigation.

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## Appendix 1.

### The Mullins-Sekerka instability in the case of step-diffusion rearrangement.

The usual Mullins-Sekerka instability takes into account off-cluster rearrangements. Here the appropriate formulae for step-diffusion will be derived.

We want to specify conditions for the fractal growth of an island, in the intermediate temperature regime where a continuous description can be properly employed. As a first case we will consider the boundary edge of the island as an infinite straight step parallel to the  $y$  axis (Fig. 1). Since atoms have a small probability of falling directly on top of a small island, we assume that adatoms diffuse on the surface to the step only from one side, the right one for definiteness. The step thus grows to the right (positive  $x$ ).

As we discuss in the body of the paper, the step in a diffusion field is unstable to fluctuations, since new adatoms will preferentially hit the protruding parts of the boundary and will amplify the fluctuation itself. This is the assumed mechanism leading to a fractal structure.

We replace this stochastic behaviour by a deterministic process, an average current of adatoms onto the boundary edge. We introduce also a diffusion current along the step as a stabilizing effect, and compute the resulting step profile to first order in the amplitude of the fluctuation.

Assume a sinusoidal perturbation,

$$\delta x = \eta \cos(ky). \quad (\text{A.1.1})$$

The density of adatoms obeys the diffusion equation with a constant source (the deposition flux  $F$ ); we still consider the temperature sufficiently low as to allow neglecting evaporation from the surface. To simplify we require stationarity:

$$D_s \nabla^2 \rho + F = 0. \quad (\text{A.1.2})$$



Approximating the island boundary with a straight step is certainly not satisfying when the wavelength  $1/k$  of the perturbed profile becomes of the order of the linear size  $R$  of the island, but we expect the results to be correct in order of magnitude. The solution of equation (A.1.2) for  $\eta = 0$  is

$$\rho_0(x) = -\frac{F}{2D_s}x^2 + A + Bx, \quad (\text{A.1.3})$$

where  $A$  and  $B$  are constants. Since the  $y$  axis has been chosen along the average step direction (Fig. 1), the term in  $x^2$  may be neglected in the linear stability analysis. For that reason the linear stability analysis yields the same results as in the case  $F = 0$  already discussed in literature ([18, 19, 20]). Even the nonlinear version should give analogous results for  $F = 0$  and for small  $F$  in the case of a small island.

Imperfect sticking of adatoms to the step can be taken into account if we write Eq.(A.1.2) in a discretized form

$$D_s(\rho_{n+1} + \rho_{n-1} - 2\rho_n) + F = 0 \quad (1 < n < \ell - 1) \quad (\text{A.1.4})$$

and supplement it with appropriate conditions near the step. For  $n = 1$ , the diffusion constant  $D_s$  will be assumed to have a value  $D' \leq D_s$ . A small value of  $D'$  corresponds to a small sticking coefficient. Also, adatoms can be released from the step, at a rate  $1/\tau_g$ . The resulting equation is

$$D_s(\rho_2 - \rho_1) - D'\rho_1 + 1/\tau_g + F = 0. \quad (\text{A.1.5})$$

We shall first discuss the case of perfect sticking. This means that the sticking coefficient is equal to 1 ( $D' = D_s$ ) and that no adatom is released from the step ( $\tau_g = \infty$ ). Then insertion of (A.1.3) into equation (A.1.4) yields  $A = 0$ . This means that  $\rho$  should vanish along the step. This should also be true for  $\eta \neq 0$ . It turns out that a solution of equation (A.1.2) for  $\eta \neq 0$  is

$$\rho = -\frac{F}{2D_s}(x^2 - \ell x) - \rho_1 \cos(ky) \exp(kx). \quad (\text{A.1.6})$$

The first term is the particular form of equation (A.1.3) which vanishes on both steps at  $x = 0$  and  $x = \ell$ . The second term should vanish when  $x$  has the value (A.1.1). Therefore, for small  $\eta$ ,

$$\rho_1 = \eta \frac{\ell F}{2D_s} \quad (\text{A.1.7})$$

The velocity of propagation of the step, disregarding any effect of line tension, is in each point equal to  $D_s \mathbf{n}(\mathbf{n} \cdot \nabla \rho)$ , where  $\mathbf{n}$  is the unit vector normal to the step.

The two components of the gradient of the density are

$$\begin{aligned} \frac{\partial \rho}{\partial x} &= -\frac{F}{D_s} \left( x - \frac{\ell}{2} \right) + k\rho_1 \cos(ky) \exp(kx) \\ &\simeq -\frac{F}{D_s} \left( \frac{2D_s \rho_1}{\ell F} \cos(ky) - \frac{\ell}{2} \right) + k\rho_1 \cos(ky) \end{aligned} \quad (\text{A.1.8})$$

$$\frac{\partial \rho}{\partial y} = -k\rho_1 \sin(ky) \exp(kx) \simeq -k\rho_1 \sin(ky). \quad (\text{A.1.9})$$

To first order in the amplitude of the fluctuation the normal vector  $\mathbf{n}$  reads

$$\mathbf{n} = \left( 1, \frac{2D_s k \rho_1}{\ell F} \sin(ky) \right) / \sqrt{1 + \left( \frac{2D_s k \rho_1}{\ell F} \sin(ky) \right)^2} \quad (\text{A.1.10})$$

and we finally find

$$v_x = \delta \dot{x} = -F\ell/2 + D_s \rho_1 (k - 1/\ell) \cos(ky). \quad (\text{A.1.11})$$

In a comoving frame the first term vanishes. Since we are interested in the case  $k\ell \gg 1$ , in the comoving frame we have

$$\delta \dot{x} = k\ell F/2 \delta x. \quad (\text{A.1.12})$$

The fluctuation is therefore exponentially amplified, since  $k\ell F$  is positive. This formula is valid between two infinite steps. For a terrace of radius  $R$ , it is reasonable to replace  $\ell$  by  $\ell^2/R$  as explained in the text and equation (10) is obtained.

We now discuss the case of imperfect sticking. The quadratic term in equation (A.1.3) will be neglected. Equation (A.1.5) implies

$$(D_s - D')B - D'A + 1/\tau_g = 0. \quad (\text{A.1.13})$$

Therefore the value  $A$  of the adatom density on the step is given by

$$A = \frac{1}{D'\tau_g} + \left( \frac{D_s}{D'} - 1 \right) B. \quad (\text{A.1.14})$$

In the case of two parallel steps,  $B$  is of order  $F\ell/2D_s$ , as in equation (A.1.6). The main difference with the case of perfect sticking is that  $1/\tau_g$  now depends on the curvature, since adatoms are more easily released at tips, where  $x'' = \partial^2 x / \partial y^2$  is negative:

$$A \simeq A_0 - A_1 x''. \quad (\text{A.1.15})$$

The solution of the diffusion problem has still the form (A.1.6). The boundary condition is  $\rho(x)$  equal to (A.1.15) when  $x$  is given by (A.1.1). Then a correction proportional to  $k^2$  should be added to (A.1.7). This yields a stabilizing term proportional to  $-k^3 \delta x$  in  $\delta \dot{x}$ , as in the original calculation of Mullins and Sekerka.

## Appendix 2.

### Calculation of the diffusion coefficient $\Lambda$ .

$\Lambda$  is defined by

$$\mathbf{j} = -\Lambda \nabla \mu \quad (\text{A.2.1})$$

where  $\mu$  is the chemical potential. Let us assume a free energy of the form

$$\mathcal{F} = \frac{A}{2} \int d^d r \rho^2(\mathbf{r}) - \int d^d r \mu(\mathbf{r}) \rho(\mathbf{r}). \quad (\text{A.2.2})$$

For small variations of  $\mu$  one has

$$A \delta \rho(\mathbf{r}) = \delta \mu(\mathbf{r}) \quad (\text{A.2.3})$$

which implies

$$\mathbf{j}(\mathbf{r}) = -\Lambda A \nabla \rho(\mathbf{r}) \quad (\text{A.2.4})$$

and therefore

$$\dot{\rho}(\mathbf{r}) = -\text{div } \mathbf{j}(\mathbf{r}) = \Lambda A \nabla^2 \rho(\mathbf{r}). \quad (\text{A.2.5})$$

Transforming to Fourier components this becomes

$$\dot{\rho}_k = -\Lambda A k^2 \rho_k \quad (\text{A.2.6})$$

which can be readily integrated, with solution

$$\rho_k(t) = \rho_k(0) \exp(-\Lambda A k^2 t). \quad (\text{A.2.7})$$

Taking the thermal average according to  $\mathcal{F}$  we get

$$\langle \rho_k(t) \rho_{-k}(0) \rangle = \langle \rho_k \rho_{-k} \rangle \exp(-\Lambda A k^2 t). \quad (\text{A.2.8})$$

Taking the time derivative of the left-hand side we end up with the following equation,

$$\langle \dot{\rho}_k(t) \rho_{-k}(0) \rangle = -\Lambda A k^2 \langle \rho_k \rho_{-k} \rangle \exp(-\Lambda A k^2 t). \quad (\text{A.2.9})$$

Making use of the trivial identity

$$\langle \dot{\rho}_k(t) \rho_{-k}(0) \rangle = - \int_0^t dt' \langle \dot{\rho}_k(t') \dot{\rho}_{-k}(0) \rangle \quad (\text{A.2.10})$$

and using Equation (A.2.5), we finally find

$$\begin{aligned} \langle \dot{\rho}_k(t) \rho_{-k}(0) \rangle &= - \sum_{\alpha=1}^d k_\alpha^2 \int_0^t dt' \langle j_{\alpha k}(t') j_{\alpha, -k}(0) \rangle = \\ &= - \frac{k^2}{d} \int_0^t dt' \langle \mathbf{j}_k(t') \cdot \mathbf{j}_{-k}(0) \rangle. \end{aligned} \quad (\text{A.2.11})$$

Summarizing,

$$\Lambda A \langle \rho_k \rho_{-k} \rangle \exp(-\Lambda A k^2 t) = \frac{1}{d} \int_0^t dt' \langle \mathbf{j}_k(t') \cdot \mathbf{j}_{-k}(0) \rangle. \quad (\text{A.2.12})$$

The latter relation is true if  $t$  is large enough to provide for an exponential decrease of the integral. If we also assume that  $t \ll \Lambda A / k^2$ , we have

$$\Lambda A \langle \rho_k \rho_{-k} \rangle = \frac{1}{d} \int_0^t dt' \langle \mathbf{j}_k(t') \cdot \mathbf{j}_{-k}(0) \rangle. \quad (\text{A.2.13})$$

Since the free energy is quadratic in the density, the correlation function is simply

$$\langle \rho_k \rho_{-k} \rangle = \frac{T}{A} \quad (\text{A.2.14})$$

and Eq.(A.2.13) becomes

$$\Lambda = \frac{1}{Td} \int_0^t dt' \langle \mathbf{j}_k(t') \cdot \mathbf{j}_{-k}(0) \rangle. \quad (\text{A.2.15})$$

This general formula will now be applied to the special case of diffusion on a step. We take the current of gradatoms along a step edge,

$$j_k = \frac{1}{\sqrt{N}} \sum_m v_m \exp(ikx_m), \quad (\text{A.2.16})$$

where  $m$  labels the gradatoms and  $N$  is the total length (assumed to be large) of a step (assumed to be straight). Then equation (16) is obtained by inserting equation (A.2.16) in equation (A.2.15).

The average thermal distance between kinks is  $\ell_0 = \exp(W_0/T)$ . A gradatom covers such distance in a time  $\tau_0 = \ell_0^2/D_g$ . Let  $t = \tau_0$ . The average number of gradatoms emitted by a kink per unit time is  $1/\tau_k$ . A fraction  $1/\ell_0$  of these gradatoms reach a kink. Therefore the number of gradatoms which are effective on a length  $\ell_0$  is given as  $\tau_0/(\tau_k \ell_0)$ . Since their velocity is  $v = \ell_0/\tau_0$ , we find

$$\Lambda = \frac{1}{\ell_0 T} \tau_0 \frac{\tau_0}{\tau_k \ell_0} v^2 = \frac{1}{T \tau_k} \quad (\text{A.2.17})$$

which is equation (17).

Quite surprisingly, the result is independent of both  $D_g$  and  $W_g$ .

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