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Kinetic aspects of thermodynamical instabilities

Aspects cinétiques des instabilités thermodynamiques

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Abstract

The instability of a coherent (i.e., without misfit dislocations), epitaxial film with respect to surface shape modulation is, in the absence of growth, an activated process if the surface has a singular orientation, e.g., (001). If the misfit is small, the activation energy is very large and practically forbids the instability. It was recently pointed out by Tersoff that the nature of the instability is deeply modified by growth. In the case of a small misfit, Tersoff used the theory of capillary effects elaborated by Nozières and Gallet in 1987 to argue that the *growing* surface becomes *linearly* unstable as in the case of a non-singular surface. In the following pages, the effect of time is addressed in the case of singular, initially planar surface. It is claimed that the instability is neither linear, nor thermally activated, but of a third type. In fact, it appears after a well-defined time. **To cite this article:** J. Villain, C. R. Physique 4 (2003).

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Résumé

L'instabilité d'un film épitaxial cohérent (c'est-à-dire sans dislocations de désaccord) par rapport à une modulation de la surface est, en l'absence de croissance, un processus activé si la surface a une orientation singulière, par exemple (001). Si le désaccord paramétrique est faible, l'énergie d'activation est élevée et interdit pratiquement l'instabilité. Tersoff a récemment remarqué que la nature de l'instabilité est profondément modifiée si la couche épitaxiale croît. Dans le cas d'un désaccord faible, il a suggéré qu'une surface singulière devient *linéairement* instable lors de la croissance. Dans cette note, nous montrons que l'instabilité n'est ni linéaire, ni thermiquement activée, mais d'un troisième type : elle apparaît au bout d'un temps bien déterminé. **Pour citer cet article :** J. Villain, C. R. Physique 4 (2003).

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Version française abrégée

La structure d'un adsorbant cristallin A de constante réticulaire $a + \delta a$ déposé sur un substrat cristallin S de constante réticulaire a est un problème d'intérêt à la fois fondamental et technologique [1,2]. Si le désaccord paramétrique $\varepsilon_0 = \delta a/a$ est inférieur à quelques %, on peut obtenir une structure qui a la topologie d'un cristal parfait, sans dislocations. C'est l'épitaxie cohérente. Un dépôt épitaxial cohérent d'épaisseur uniforme et suffisamment grande est en principe instable si $\varepsilon_0 \neq 0$ parce que son énergie élastique est positive et de l'ordre de $M\varepsilon_0^2 V_a$ par atome, où M est une constante élastique et V_a le volume atomique. L'instabilité se traduit généralement par des dislocations de désaccord. Cependant, l'énergie élastique peut aussi être

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abaissée si l'adsorbat forme des amas ou si sa surface est modulée au lieu de demeurer plane. C'est l'instabilité d'Asaro–Tiller–Grinfeld [1–7]. La discussion de l'instabilité est l'objet du présent travail. Elle fait intervenir l'énergie élastique (déstabilisante) qui s'annule avec ε_0 , et l'énergie de surface (ou « capillaire »), qui résulte en gros des liaisons cassées pour faire la surface. La nature de l'instabilité *en l'absence de croissance et d'évaporation* dépend de l'orientation de la surface. Une surface de haute symétrie, par exemple (001), a généralement une énergie libre de marche $\tilde{\gamma}_s > 0$, de sorte qu'une désorientation d'angle θ produit un excès d'énergie libre par unité d'aire non analytique $\delta f_{\text{cal}} \sim \tilde{\gamma}_s |\text{tg } \theta|$. Une telle orientation est dite *singulière*. Au contraire, pour une surface non-singulière, δf_{cal} est définie par la formule (1) où apparaît la rigidité de surface γ finie. Pour une orientation non singulière, la surface initialement plane forme immédiatement des ondulations [1–3,6,7] dont les composantes de Fourier de grande longueur d'onde croissent exponentiellement avec le temps. En d'autres termes leur amplitude h_q évolue aux temps courts selon l'équation linéaire $dh_q(t)/dt = \alpha_q h_q(t)$, et nous dirons que l'instabilité est linéaire. Pour une orientation singulière, l'instabilité exige de surmonter une barrière de potentiel, et nous dirons qu'il y a une instabilité activée.

Il a été suggéré récemment par Tersoff [9] que lors de la *croissance* d'une couche épitaxiale limitée par une surface singulière, l'instabilité est linéaire. Nous confirmons ce point de vue dans un cas particulier : celui où un effort anisotrope est appliqué brusquement à un système dont la croissance a atteint son état stationnaire, cinétiquement rugueux. Dans le cas de la croissance en phase vapeur sous faible sursaturation, nous obtenons alors la formule linéaire (8) qui coïncide, à un facteur peu significatif près, avec le résultat de Tersoff. Quand la sursaturation devient forte, il peut être nécessaire d'introduire des phénomènes non-linéaires ignorés par Tersoff.

Un cas différent est celui d'une surface singulière initialement bien plane. L'énergie libre de marche est alors non nulle et ne s'annule qu'à temps t infini. Aux temps courts, elle rend l'instabilité linéaire impossible. Au fur et à mesure que le temps s'écoule, la rugosité cinétique se développe, l'énergie libre de marche $\tilde{\gamma}_s(t)$ diminue et la hauteur $H(t)$ des bosses augmente. L'instabilité se produit au bout d'un certain temps t_1 donné approximativement par la condition $\tilde{\gamma}_s(t_1)/H(t_1) \approx M\varepsilon_0^2 a_s$, où ε_0 est le désaccord de maille, a_s la hauteur des marches atomique, M une constante élastique et $\tilde{\gamma}_s(t)$ et $H(t)$ peuvent être en principe calculés pour $\varepsilon_0 = 0$. Des conjectures sur le comportement de $\tilde{\gamma}_s(t)$ sont présentées dans la Section 4. On a en somme un troisième type d'instabilité, qui se produit au bout d'un certain temps bien déterminé.

1. Coherent epitaxy

The structure of a crystalline adsorbate A of lattice constant $a + \delta a$ on a crystalline substrate S of lattice constant a is a problem of current fundamental and technological interest [1–5]. If the misfit $\varepsilon_0 = \delta a/a$ is less than a few percents, one can obtain a structure which has the topology of an ideal crystal, i.e., avoid misfit dislocations. This is called coherent epitaxy. A coherent, epitaxial deposit of large, uniform thickness is in principle unstable if $\varepsilon_0 \neq 0$ because it involves a positive elastic energy of the order of $M\varepsilon_0^2 V_a$ per atom, where M is an elastic constant and V_a the atomic volume. In the absence of metastability, the usual fate of the surface is to form misfit dislocations. However, the elastic energy can also be lowered if the adsorbate forms clusters or if its surface is modulated instead of remaining planar [1–7]. The discussion of this instability is the purpose of the present note. It requires to take into account the (destabilizing) elastic energy which vanishes with ε_0 , and the stabilizing surface energy (or capillary energy), which roughly results from breaking chemical bonds to make the surface.

The nature of the instability depends on the orientation of the surface. In the absence of growth and evaporation, as will be seen, it can be 'linear', i.e., appear in a linear stability analysis, or it can be 'activated'.

The case discussed by Asaro and Tiller [6] and by Grinfeld [7] is that of a non-singular surface, when the capillary energy $\mathcal{F}_{\text{cap}}(x, y)$ is analytic. If the surface height $z(x, y)$ is a slowly varying function of the other two coordinates x and y , then $\mathcal{F}_{\text{cap}}(x, y) = \mathcal{F}_{\text{cap}}^0 + \delta \mathcal{F}_{\text{cap}}(x, y)$ where $\mathcal{F}_{\text{cap}}^0$ is the free energy of the planar surface $z = \text{Const}$ and

$$\delta \mathcal{F}_{\text{cap}} = \frac{\gamma}{2} \int d^2 r [(\partial_x z)^2 + (\partial_y z)^2] = \int d^2 r f_{\text{cap}} \quad (1)$$

where $f_{\text{cap}} = \gamma[z_x^2 + z_y^2]/2$ is the surface free energy density and $z_\alpha = \partial_\alpha z = \partial z/\partial \alpha$. In that case, a linear stability analysis, summarized in Section 2, is sufficient to demonstrate the instability of the planar surface [1]. The surface is 'linearly unstable'.

The other case is that of a high symmetry surface such as (001). Then, (1) does not apply [1]. Indeed the formation of bumps requires the formation of steps of atomic height a_s , which have a positive free energy $\tilde{\gamma}_s$ per unit length. The formation of a bump of height h and volume $\approx hR^2$ requires a capillary energy of order

$$\delta \mathcal{F}_{\text{cap}} \approx \frac{\tilde{\gamma}_s h R}{a_s} \quad (2)$$

which, for $h \ll R$, is much larger than the value of the order of γh^2 predicted by (1). The elastic energy gain is of the order of $-M\varepsilon_0^2 h^2 R$ [1], where M is an elastic constant. For small h , this negative energy is dominated by the positive contribution (2),

so that bump formation requires overcoming an activation free energy [8] which is proportional to ε_0^{-4} and very large for small misfit ε_0 . This instability will be called ‘activated’. It develops more slowly than the linear instability. If (2) holds, the surface is called ‘singular’.

For a non-singular surface, (2) should vanish, so that $\tilde{\gamma}_s = 0$. This implies that arbitrarily large closed steps are present. This property is characteristic of a *rough* surface.¹ A rough surface is non-singular and a smooth surface is singular. This is well known at equilibrium. Now, a surface which is smooth and singular at equilibrium becomes rough when it grows. It is thus reasonable to expect that it also becomes non-singular and undergoes a linear instability if $\varepsilon_0 \neq 0$. This is Tersoff’s suggestion [9]. However, in usual situations, the surface is initially planar and singular, and kinetic roughness develops only progressively with time. This effect of time will be discussed in Section 4, where it will be argued that the instability of a growing, singular surface belongs to a third type. In Sections 2 and 3, Tersoff’s theory will be discussed in the steady, kinetically rough state.

As in Tersoff’s paper, attention is focussed on the case of a weak misfit and a modulation size larger than the typical distance L_s between steps in the steady regime. Gravity will be neglected. The notations γ , ε_0 , M and L_s have the same meaning as in Tersoff’s paper [9] as well as a_s , the step velocity v_s , while the local growth rate will be called v and its value for a planar surface will be called v_0 . A particularly important point concerns the step free energy per unit length $\tilde{\gamma}_s$. It is defined as follows for a square sample of size $L \times L$. Let $\mathcal{F}_1(t)$ be the free energy with boundary conditions $z(L, y, t) - z(0, y, t) = a_s$, and $\mathcal{F}_0(t)$ the free energy with boundary conditions $z(L, y, t) - z(0, y, t) = 0$. Then $\tilde{\gamma}_s(t) = [\mathcal{F}_1(t) - \mathcal{F}_0(t)]/L$. For an initially planar surface, $\tilde{\gamma}_s$ is initially equal to the step free energy at thermal equilibrium, $\tilde{\gamma}_s(0) = \gamma_s$, which will be assumed positive. At $t = \infty$, the surface is kinematically rough and one expects $\tilde{\gamma}_s = 0$ in the steady state. More precise predictions about this point will be conjectured in Section 4.

2. The Asaro–Tiller–Grinfeld instability: a reminder

It is of interest to recall the mechanism of the instability for a crystal in the presence of its vapour when the interface is non-singular [1,2,6,7]. The surface height $z(x, y, t) = v_0t + \delta z(x, y, t)$ satisfies the equation

$$\frac{\partial z}{\partial t} = v_0 - k[\delta\mu_{\text{cap}} + \delta\mu_{\text{el}} + \delta\mu_{\text{int}}], \quad (3)$$

where $v_0 = 0$ in the case of a vanishing supersaturation addressed by Asaro and Tiller [6] and by Grinfeld [7]. The quantities $\delta\mu_{\text{cap}}$, $\delta\mu_{\text{el}}$, $\delta\mu_{\text{int}}$ (which depend on x, y, t) are respectively the changes in surface chemical potential, elastic chemical potential and interface chemical potential when the surface modulation $\delta z(x, y, t)$ is created. They vanish when the surface is a plane ($\delta z(x, y, t) = 0$). The kinetic coefficient k is positive.

The interface chemical potential may be assumed to be constant if, as will be assumed, the adsorbate is thicker than some lower thickness which is probably atomic (‘wetting layer’ thickness). Thus $\delta\mu_{\text{int}} = 0$.

The capillary chemical potential excess $\delta\mu_{\text{cap}}(x, y)$ is given by the Herring–Mullins formula

$$\frac{1}{V_a} \delta\mu_{\text{cap}} = -\partial_x \left(\frac{\partial f_{\text{cap}}}{\partial z_x} \right) - \partial_y \left(\frac{\partial f_{\text{cap}}}{\partial z_y} \right) = -\gamma [\partial_{xx}^2 z + \partial_{yy}^2 z]. \quad (4)$$

The elastic chemical potential excess will be assumed to be

$$\delta\mu_{\text{el}}(x, y, t) = V_a \varepsilon_0^2 \int dx' dy' \Gamma(x - x', y - y') \delta z(x', y', t), \quad (5)$$

where $\Gamma(x, y)$ is an elastic Green function. Formula (5) is exact if the substrate and the adsorbate have identical elastic constants. Then, the Green function $\Gamma(x, y)$ is proportional to $M(x^2 + y^2)^{-3/2}$. In the general case when the substrate and the adsorbate usually have different elastic constants, (5) is approximate.

The linear instability analysis consists in assuming $\delta z(x, y, t) = h(t) \cos(qx)$. Then (5) yields [1,2,6,7] $\delta\mu_{\text{el}}(x, y, t) = M V_a \varepsilon_0^2 |q| h(t) \cos(qx)$. Formulae (3)–(5) then yield $dh/dt \approx k V_a (M \varepsilon_0^2 |q| - \gamma q^2) h$ and an instability appears for $|q| < M \varepsilon_0^2 / \gamma$.

3. Growing surface: steady case

The actual structure of the surface is a complicated array of steps. To simplify the description, it is convenient to define $z(x, y, t)$ as the average surface height on the time interval $[t - t_c/2, t + t_c/2]$, where t_c is the time to grow a single atomic

¹ This corresponds to the definition used in statistical mechanics, for instance when discussing the roughening transition. The word ‘rough’ is sometimes used with a different meaning.

layer. Thus, $z(x, y, t)$ may plausibly be assumed to be a continuous, analytic function which, in the presence of a vapour of supersaturation $\Delta\mu$, satisfies (3). From the expression [9,10] of $v_0(\Delta\mu)$,

$$v_0 \approx \text{Const}(\Delta\mu)^{4/3} \exp\left(-\frac{\pi V_a \gamma_s^2}{3k_B T a_s \Delta\mu}\right) \quad (6)$$

the value of $k = \partial v_0 / \partial(\Delta\mu)$ is deduced, namely (for small $\Delta\mu$)

$$k \approx \frac{\pi \gamma_s^2 V_a v_0}{3k_B T a_s \Delta\mu^2}. \quad (7)$$

The elastic chemical potential excess $\delta\mu_{\text{el}}$ is given by (5). To derive $\delta\mu_{\text{cap}}$ from (4), one can write that the capillary free energy is γ_s times the total step length, so that $f_{\text{cap}} \approx \gamma_s n(\theta)$, where n is the step density (per unit length). Since $n(\theta) \approx (1/a_s) \tan\theta$ for $\theta \gg L_s/a_s$ and $n(\theta) \approx (1/L_s)$ for $\theta \ll L_s/a_s$, a plausible interpolation [10] is

$$n(\theta) \approx \frac{1}{L_s} \left[1 + \left(\frac{L_s}{a_s} \right)^2 \tan^2 \theta \right]^{1/2}$$

or, for small θ , $f_{\text{cap}}(\theta) \approx \gamma_s n(0) + \gamma_s L_s \tan^2 \theta / (2a_s^2)$, which reads $f_{\text{cap}} \approx \text{Const} + \gamma_s L_s [(\partial_x z)^2 + (\partial_y z)^2] / (2a_s^2)$. This is the surface energy density of a *non-singular* surface of surface stiffness $\gamma_{\text{kin}} = \gamma_s L_s / a_s^2$. Replacing γ by this expression in (4), inserting the result into (3), replacing in (3) k by its expression (7) and $\delta\mu_{\text{el}}$ by $M V_a \varepsilon_0^2 |q| h(t) \cos(qx)$ as in Section 2, one obtains for a sinusoidal modulation $z(x, y, t) = v_0 t + h(t) \cos(qx)$

$$\frac{\partial h}{\partial t} = \frac{\pi \gamma_s^2 V_a^2 v_0}{3a_s k_B T \Delta\mu^2} \left[M \varepsilon_0^2 |q| - \frac{\gamma_s L_s}{a_s^2} q^2 \right] h. \quad (8)$$

This is formula (9) of [9], apart from the dimensionless factor $(\pi \gamma_s V_a)^2 / (a_s k_B T \Delta\mu)^2$ ignored by Tersoff. Formula (8) holds for small $\Delta\mu$. When $\Delta\mu$ is too large, the term proportional to $|q|$ should be corrected (see Tersoff's formula (13)) and nonlinear terms should be added. This point has been disregarded by Tersoff and will be discussed now for $\varepsilon_0 = 0$, when the dynamics are described by the 'KPZ' equation [11]

$$\frac{\partial z}{\partial t} = v_0 + v \nabla^2 z + \frac{1}{2} \lambda (\nabla z)^2 + \eta(x, y, t). \quad (9)$$

The random quantity $\eta(x, y, t)$, usually called 'noise', expresses the stochastic properties of deposition, diffusion evaporation and nucleation. It will be assumed to be 0, as it was in (8). Comparison of (9) with (8) for small h yields

$$v = \frac{\pi \gamma_s^3 L_s V_a^2 v_0}{3a_s^3 k_B T \Delta\mu^2}. \quad (10)$$

The parameter λ can be obtained, assuming a vanishing curvature, if the interpolation procedure which was previously used for $n(x, y, t)$ is applied to the growth velocity $v = \partial z / \partial t$. For $\tan\theta \gg a_s / L_s$, $v(\theta) \simeq v_s \tan\theta$, while $v(0)$ has a value v_0 which is quite small at low supersaturation [9,10]. A reasonable interpolation is $v(\theta) = v_0 [1 + (v_s / v_0)^2 \tan^2 \theta]^{1/2}$ or, if θ is small, $v(\theta) = v_0 + (1/2)(v_s^2 / v_0)(\nabla z)^2$. Comparison with (9) yields

$$\lambda = v_0 / v_s^2. \quad (11)$$

The non-linear λ -term is a weak perturbation to (8) if $\lambda h \ll v$. One might think that it is always so if h is small enough. However, it does not make much sense to assume $h < a_s$. A plausible condition of validity of the linear approximation is $\lambda a_s \ll v$. This condition is indeed satisfied for weak supersaturation, as seen from (10) and (11).

Thus, Tersoff's essential results have been reproduced (apart from a minor correction). However, the theory of this section holds only in special cases, for instance if the crystal grows steadily for $\varepsilon_0 = 0$ and then an anisotropic stress $\varepsilon_0 \neq 0$ is applied by a mechanical procedure. This looks compatible with Balibar's experiments [13]. In the next section, another situation is addressed.

4. Time

In many usual cases, especially in epitaxial growth, the anisotropic stress $\varepsilon_0 \neq 0$ (which results from the substrate) is already applied at the time $t = 0$ at which the growth begins, and at this time the surface is almost exactly a plane $z = \text{Const}$. This is the situation addressed in this section.

In that case, the continuity assumptions and interpolations made in the previous section do not hold at $t = 0$, since the surface is singular, $\tilde{\gamma}_s(0) = \gamma_s \neq 0$. They do not hold either at short times, because $\tilde{\gamma}_s(t)$ vanishes only for $t = \infty$ as speculated in Section 1 and in a more precise way below in this section.

An equation which should yield a correct qualitative description of the evolution is the following

$$\frac{\partial z}{\partial t} = v_0 + \nu \nabla^2 z + \frac{1}{2} \lambda (\tilde{\nabla} z)^2 - Q \sin(2\pi z) - k \delta \mu_{\text{el}}(x, y, t) + \eta(x, y, t), \quad (12)$$

where Q is a constant which mimicks the effect of the lattice. The other terms already appear in (9) or (3). For $\varepsilon_0 = 0$, $\delta \mu_{\text{el}} = 0$ and (12) reduces to the ‘HKP’ equation introduced by Hwa et al. [12]. However, the time-independent renormalisation group equations derived by Hwa et al. hold only in the steady regime.

A few phenomenological conjectures will be given below.

For short times, the bumps which arise from kinetic roughness are small, the step free energy is large and the capillary energy (2) dominates the elastic energy which can be neglected. The effect of the noise η is now essential, since it produces an increase of the kinetic roughness, i.e., an increase of the maximum bump radius $\xi(t)$ and the typical bump height $H(t)$ increase. This has been observed in many simulations (mainly in the case of MBE [2]) and might presumably be deduced from the HKP equation. Thus the elastic free energy $-M\xi(t)H^2(t)\varepsilon_0^2$ of a typical bump [2,8] increases in absolute value and the capillary free energy $\tilde{\gamma}_s(t)H(t)\xi(t)/a_s$ increases less rapidly or decreases. When both energies become comparable, the elastic term of the HKP equation can no longer be neglected, and the instability takes place. This occurs at a time t_1 defined by $\tilde{\gamma}_s(t_1)/H(t_1) \approx M\varepsilon_0^2 a_s$. The left-hand side of this equation is a decreasing function of t , which does not depend on the misfit ε_0 , and it vanishes at $t = \infty$. Therefore t_1 becomes very large for small ε_0 . It also becomes large for a weak supersaturation because kinetic roughness increases less rapidly, so the decrease of $\tilde{\gamma}_s(t)$ and the increase of $H(t)$ are slower.

Another conjecture concerns $\tilde{\gamma}_s(t)$. Although it is dangerous to apply the Gibbs–Boltzmann formula to a non-equilibrium situation, it will be assumed that the number $n(\xi)$ of steps of radius ξ per unit area is dominated by a factor $\exp[-\beta\gamma_s\xi]$. On the other hand, $n(\xi)$ is presumably not very different from the value it has in the steady, rough state, which may be assumed to be a power of ξ . This argument yields $\exp[-\beta\gamma_s\xi] \sim \xi^{-\lambda}$ and $\gamma_s(t) \sim \lambda \ln \xi(t)/\xi(t)$, which does vanish, as expected, at long time.

5. Conclusion

To conclude, the instability of coherent epitaxial films with respect to surface modulation is a *thermodynamical* one since it decreases the free energy. However its nature is deeply modified by kinetic factors when the crystal grows. In the present note, the growing surface is assumed to have a high symmetry orientation, so that it is smooth and ‘singular’ at equilibrium. Two extreme cases can be distinguished.

The first one is when a mechanical, anisotropic stress is applied as the growing surface is in its steady state. Then the surface is linearly unstable, as stated by Tersoff [9]. In Section 3 of the present note, Tersoff’s calculation has been reproduced, a more precise form of his result has been given, and the effect of nonlinear terms neglected in his article has been shown to be small in the case of weak supersaturation.

The second case is that of an initially smooth surface. It corresponds to epitaxy. Then, the short time features of the instability are those of an activated one. In practice, this means that no surface modulation appears at short time if the misfit is weak, in contrast with a linear instability. Kinetic roughness, which increases with time, is essential to trigger the thermodynamic instability which does appear at a particular time t_1 . Conjectures on t_1 have been presented in Section 4. Such an instability is neither linear nor activated, but belongs to a new type. This is also true for molecular beam epitaxy although the quantitative form of the equations is completely different.

An experimental check of these predictions is not easy for the following reasons: (i) the instability is generally observed for rather large misfits. Its observation at low misfit would require thick deposits; (ii) a singular orientation is a limiting case which is never realized because a real surface always contains a few steps.

Moreover, the limit of very low growth rate, which has been investigated here and by Tersoff, is not realistic. In the case of a faster growth, the equations would be somewhat more complicated and the nonlinear term of (9) might become important. Despite these remarks, the present observations are of interest because a good understanding of limiting cases is useful to understand real problems.

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