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LETTER TO THE EDITOR

Morphological instability at the early stages of heteroepitaxial growth on vicinal surfaces

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Abstract
We study the strain-induced morphological instability at the submonolayer coverage stage of heteroepitaxial growth on a vicinal substrate with regularly spaced steps. We study the regime in which diffusion along the film edge is the dominant mechanism of transport of matter. We perform a linear stability analysis and determine for which conditions of coverage a flat front is unstable and for which conditions it is stable. We discuss the effect of step energy. Our results predict that when the thin film covers less than one-half of the terraces the flat front is unstable. For very small coverage, the front will spontaneously break into a regular array of islands. We obtain expressions for the aspect ratio, the size and the spacing of the islands forming this array. This opens the possibility of inducing the spontaneous formation of an array of two-dimensional quantum structures with the desired size and spacing by controlling the cutting angle of the vicinal surface and the fraction of the surface covered by the material.

Two-dimensional (2D) structures at the very early stages of epitaxial growth are crucial to understand the fundamental mechanisms of film growth. Instabilities, that were once considered a problem in growing smooth films, are now of fundamental importance. Technologically, understanding the spontaneous ordering of nano-structures opens the possibility of tailoring arrays of quantum wires and quantum dots which might form the basis of novel electronic and optoelectronic devices. Nowadays the whole field of thin-film deposition and spontaneous ordering of nanostructures on crystal surfaces has become a subject of intense experimental and theoretical study [1–3]. As Li *et al* [4] pointed out, on a vicinal substrate one may expect approximate 2D analogues of all the 3D equilibrium growth modes. For homoepitaxy on a vicinal substrate one would expect ideal step flow growth, i.e. stripes

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of the deposited material advancing with a smooth front [5]. In heteroepitaxy, one would expect the formation of a stressed domain structure. Strain-induced self-organized formation of 3D nano-structures has been studied in the last few years [6]. In a recent letter Li *et al* [4] demonstrate by both theory and experiment that a similar approach for nanofabrication can be applied to the growth of strained 2D layers. By comparing the energy of an array of stripes with the energy of an array of rectangular islands aligned along the steps they showed that stripes of width $w = \theta W$ growing on terraces of width W that have a smooth front are energetically unfavourable when the coverage θ is small. To do this, they have taken into account the elastic energy as well as the step energy. They have also shown that for large coverage an array of rectangular islands with less energy than an array of stripes does not exist.

The total free energy approach determines which shape is energetically favourable. However, it does not say anything about the dynamical stability of the considered shape. Moreover, there might be different configurations, some stable some unstable, with exactly the same total energy. Therefore the total energy criteria would not discriminate among them. On the other hand, a stability analysis that describes the relaxation of a structure towards equilibrium could tell us whether a structure is or is not dynamically stable. It gives information on how a perturbation grows or decays, the wavelength that dominates the pattern and the dynamics towards the equilibrium or steady state. A linear stability analysis is, of course, not as general as a full stability analysis, since it is only valid at very short times, but it is able to predict whether a structure is stable or not, and in the case when it is unstable, it gives the wavelength that dominates the pattern at onset. Therefore, a linear stability analysis should be considered as complementary to a total energy analysis. The former approach is used in this letter.

In this letter we present a study of the strain-induced morphological instability at the submonolayer coverage stage of heteroepitaxial growth on a vicinal substrate with regularly spaced steps. The growth process could be roughly described as follows. Particles from the external flux fall into the terraces, diffuse rapidly on them and are incorporated into the growing film. We consider the case of very slow external flux in such a way that at any time the film could be considered to be in mechanical equilibrium and to have a fixed number of particles. It should be clear that none of the experimental growing conditions will meet these requirements. It might sound contradictory to talk about growth and to analyse a film with a given number of particles, but it is a question of timescales. For fast flows, an analysis such as that in [7] should be considered. Moreover, to freeze the growth process in order to study instabilities is a regular assumption in the area of pattern formation [8]. The probability of particles separating from the film, which implies lowering the coordination number, is neglected in the present analysis. Also, since diffusion in terraces is very fast, its characteristic time may be considered negligible with respect to the characteristic time of diffusion along the step edges. We therefore consider that diffusion along the step edges is the dominant mechanism of transport of matter once mechanical equilibrium has been reached. We perform a linear stability analysis and determine for which conditions of coverage a flat front is unstable and for which conditions it is stable. We obtain that the flat front is unstable for any coverage below $\theta = 0.5$ whereas it is stable for any coverage above $\theta = 0.5$. We find that elasticity by itself gives no wavelength selection. We then consider the step energy (or line tension) which, due to its stabilizing effect, gives rise to wavelength selection when the flat front is unstable. For very small coverage, the instability will cause the breaking of the stripe into an array of islands aligned along the steps. For systems in which the linear regime is observable, the fastest-growing wavelength of our analysis will determine the size and spacing of such an array. Our results give the explicit dependence of the islands' size and spacing on the terrace width and the coverage. Since these are parameters that can be controlled experimentally, our

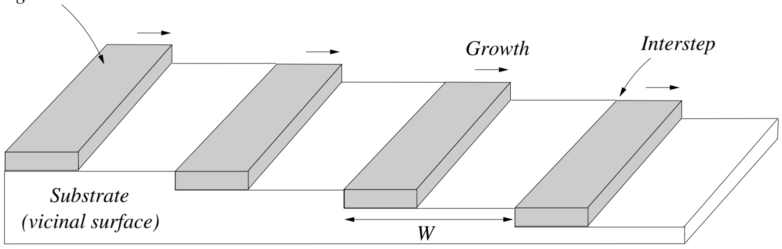

Figure 1. Step flow on a vicinal surface.

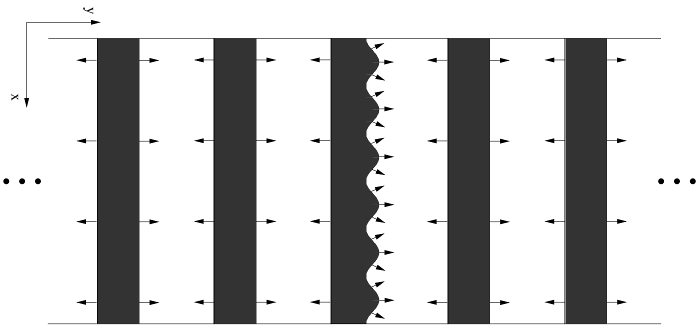

Figure 2. Schematic representation of the force distribution along the array of stripes. The vicinal surface is viewed from the top.

results offer a way to find out whether the linear regime determines the island structure for a given system. When it turns out to be such, our results give a way of tailoring the spontaneous array of islands with the desired size and spacing.

We consider an array of straight stripes growing on a vicinal surface as shown in figure 1. The lattice mismatch between the growing material and the substrate introduces an elastic-force monopole along the stripe periphery proportional to the misfit strain and the height of the step that forms the edges of the stripe [3]. Each stripe covers a fraction θ of the terrace width W. For one of them[4] we set an arbitrarily perturbed contour which can be expanded in plane waves. It turns out that for a linear analysis all modes decouple and it is enough to study one mode, so we study a perturbation of the form

$$y(x,t) = y_0 + \delta(t)\cos(kx) \tag{1}$$

where $\delta(t)$ is the time-dependent amplitude of the Fourier mode with wavenumber k and y_0 is the average position of the perturbed growing front. In the linear regime the growth or decay of a perturbation is exponential $\delta(t) = \delta_0 \exp \omega t$.

Since our system is in mechanical equilibrium we use elasticity of continuum media. This approach has been used in [4]. A distribution of force per unit length $\vec{F} = F_0\hat{n}$ acts all along the periphery of the array of stripes shown in figure 2. The displacement field $\vec{u}(x)$ over the perturbed stripe edge due to such a force distribution can be computed as [9]

$$\vec{u}(x) = \frac{1+\sigma}{\pi E} F_0 \int \left[(1-\sigma)\frac{\hat{n}}{r} + \sigma\frac{\vec{r}}{r^3}\hat{n}\cdot\vec{r}\right] \mathrm{d}S \tag{2}$$

[4] In principle, one should perturb all fronts, but in order to obtain analytical results we treat the simpler problem of perturbing a single front. This is also because perturbing several fronts at a time involves other problems, for instance the question of whether the perturbations on different fronts are in phase or out of phase.

where $\vec{r}$ is the vector difference between the point at which the force acts and the point at which the displacement is evaluated. The integration is performed along the edges of all the stripes, not only the perturbed one. The outward normal to a stripe's edge is given by $\hat{n}$. E and σ are the Young's modulus and Poisson's ratio of the substrate respectively. A cut-off length a of the order of the lattice constant is introduced in the integration. The local free energy per unit length at a point over the perturbed stripe edge is given by

$$\mu(x) = -\tfrac{1}{2}\vec{F}(x) \cdot \vec{u}(x). \tag{3}$$

The mechanism of transport of matter that we consider is the diffusion of particles along the perturbed stripe edge. A gradient in the local free energy along the edge induces a local force. Particles move in the direction that minimizes this gradient. Assuming linear response we have a flux along the stripe periphery proportional to the gradient in the free energy along the edge, i.e. $J_s \propto -\partial\mu/\partial S$. When J_s is constant, matter moves along the stripe periphery without a net effect on the stripe shape. On the other hand, when at a point on the stripe edge there is a gradient of J_s along the interface, matter accumulates, giving a contribution to the local normal velocity proportional to

$$v_n \propto -\frac{\partial J_s}{\partial S}. \tag{4}$$

The normal velocity at any point on the stripe edge is then given by

$$v_n = D\frac{\partial^2}{\partial S^2}\mu(x) \tag{5}$$

where $\mu(x)$ is the local free energy, D is a diffusion coefficient in which we have grouped all proportionality constants and $\partial/\partial S$ means differentiation along the arc length.

The perturbation given by (1) is a solution of the interface dynamics only if ω and k are related by a dispersion relation $\omega(k)$. If there are values of k for which ω is positive, an arbitrary perturbation containing such values of k is amplified and the straight stripe is unstable. If ω is negative for all k, an arbitrary perturbation is damped and the straight stripe is stable [8].

In the linear approximation the outward normal to the perturbed stripe is given by $\hat{n}(x) = \delta k \sin(kx)\hat{i} + \hat{j}$ and the differential arc length is given by $\mathrm{d}S = x$.

We obtain the following local free energy:

$$\mu_e(x) = -2E_s\left\{(1-\sigma)\ln\left[\frac{W}{2\pi a}\mathrm{sen}(\pi\theta)\right] + (1-\sigma)\frac{\pi\delta}{W}\cot(\pi\theta)\cos(kx) - \sigma\right\} \tag{6}$$

where $E_s = \frac{1+\sigma}{2\pi E}F_0^2$ is the unit strain energy introduced in [10].

We compute v_n from both our equation of motion (5) and the time derivative of $y(x)$, equate the two results and obtain an expression for the growth rate of the perturbation ω. This is given by

$$\omega_{\text{elastic}} = \frac{2E_s(1-\sigma)D\pi}{W}\cot(\pi\theta)k^2. \tag{7}$$

Figure 3 shows the plot for this dispersion relation. As is clear from equation (7), we obtain that for any coverage below $\theta = 0.5$, ω is positive, all modes grow and the flat front of the growing stripe is unstable. Moreover the smaller the wavelength of the perturbation, the faster the perturbation grows. Also, for any coverage above $\theta = 0.5$ ω is negative, meaning that all modes decay and that the straight stripe is stable. For a given mode, the larger the coverage, the faster the perturbation decays. The coverage $\theta = 0.5$ has neutral stability and gives the critical coverage. Our results agree with those obtained via the total energy criteria in the sense that stripes of width $w = \theta W$ having a smooth front are unstable when the coverage is small. So far, our stability analysis predicts no wavelength selection. That is, a finite mode k that

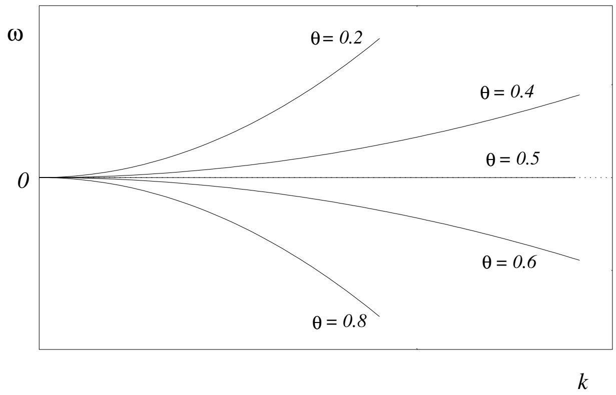

Figure 3. Elastic effect on the dispersion relation for the growth and decay of a perturbation. The upper curves are for coverages $\theta < 0.5$. For these coverages all modes are unstable and any perturbation to the straight stripe will grow. The lower curves are for coverages $\theta > 0.5$. For these coverages the straight stripe is stable. The curve $\omega = 0$ denotes neutral stability and corresponds to $\theta = 0.5$, which is the critical coverage.

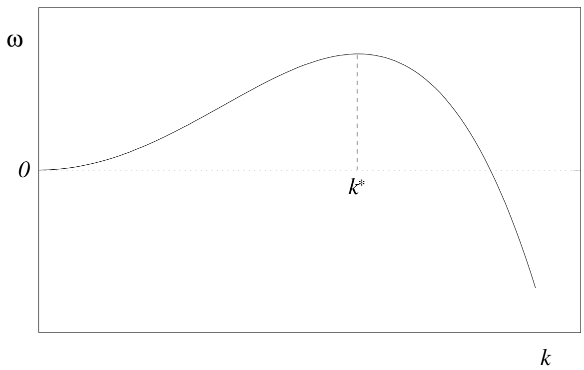

Figure 4. Dispersion relation showing the stabilizing effect of step energy for $\theta < 0.5$. The fastest-growing mode is given by k^*.

grows faster than the others and determines the wavelength of the pattern does not exist when considering solely elastic effects.

In order to have a wavelength selected, it is necessary to account for the effect of the step energy. The local free energy per particle at the step is given by [2]

$$\mu_{\text{step}}(x) = a_p^2 \gamma(x) \kappa(x) \tag{8}$$

where $\kappa(x)$ is the local curvature of the perturbed stripe edge, $\gamma(x)$ is a line tension, which in general is anisotropic, and a_p^2 is the area occupied by an atom. Here we take an isotropic γ to illustrate the overall effect of a line tension. The contribution ω_{step} to the dispersion relation is

$$\omega_{\text{step}} = -D'\gamma k^4. \tag{9}$$

The total dispersion relation is then $\omega = \omega_{\text{elastic}} - D'\gamma k^4$. Note that D' is again a diffusion coefficient. It has different units from D because the local free energy at (8) is an energy per particle, while the local elastic energy (6) is an energy per unit length. The minus sign indicates that the line tension has a stabilizing effect. For $\theta > 0.5$ the straight stripes become even more stable. This means that any perturbation will decay faster due to the step energy effect. Figure 4 illustrates the effect of the line tension on the dispersion relation for $\theta < 0.5$. For the figure we have taken arbitrary values for the parameters involved. As usual, the step energy is unable to stabilize long-wavelength modes but stabilizes short-wavelength ones. Therefore the dispersion relation has a maximum at a mode that we call k^*. This is the fastest-growing mode and determines the wavelength of the pattern at short times. It is given by

$$k^* = \left(\frac{(1-\sigma)E_s D\pi \cot(\pi\theta)}{W\gamma D'}\right)^{1/2}. \tag{10}$$

For small coverages, the growing amplitude of the perturbation will soon reach the interstep (defined in figure 1) and the undulated stripe will break into domains separated by a distance $d = 2\pi/k^*$ given by

$$d = 2W^{1/2}\left(\frac{\gamma D'\pi}{(1-\sigma)E_s D\cot(\pi\theta)}\right)^{1/2}. \tag{11}$$

Each of the domains will have an area $A = \theta W d$, given by

$$A = 2\theta W^{3/2}\left(\frac{\gamma D'\pi}{(1-\sigma)E_s D\cot(\pi\theta)}\right)^{1/2} \tag{12}$$

and an aspect ratio $c = \theta W/d$ given by

$$c = \frac{1}{2}\theta W^{1/2}\left(\frac{(1-\sigma)E_s D\cot(\pi\theta)}{\gamma D'\pi}\right)^{1/2}. \tag{13}$$

These expressions provide a way to control the domain's size and spacing. Experimentally it is possible to control the terrace width, through the cutting angle of the vicinal surface in which the material grows, and the coverage. Our results predict that for small coverages the separation between islands goes as $W^{1/2}$, the area of the islands as $W^{3/2}$ and the aspect ratio as $W^{1/2}$.

For larger coverages the undulated stripe could take much longer to break into separate domains (if it ever breaks) and the linear stability analysis results may no longer be valid since they hold only at short times. Nevertheless our results for the most unstable wavelength should be valid at the onset of the instability. To test this experimentally it would be convenient to start with high coverages for which the straight stripes are stable and allow for evaporation.

Our results agree with those obtained via the energy criteria [4] in the sense that stripes of width $w = \theta W$ having a smooth front are unstable when the coverage is small. However, the two approaches present some differences. For instance, using a total energy criterion the islands' size and spacing can be predicted even in the absence of step energy. According to our results this does not happen. Also in our approach the separation between islands tends to zero for very small coverages while in the total energy criterion the separation between islands diverges at small coverages. However, direct comparison of the results of the two approaches should be made with care, since they address different questions in different regimes. The total free energy approach determines which shape is energetically favourable. However, it does not say anything about the dynamical stability of the considered shape. Note that to linear order in δ all the contours considered in this paper have exactly the same total energy for given coverage and step energy. Hence, a total energy criterion would have predicted that they are all equally likely.

We have studied the linear stability of a planar front at the early stages of heteroepitaxial growth on a vicinal surface. We have determined whether a small perturbation around the planar front grows or decays at the early times of the dynamics. Our approach cannot say anything about the late-time regime and nonlinear effects or about situations in which several layers of material are deposited, since for such situations there is a third dimension in which matter could rearrange. The linear stability analysis cannot predict the shape of the pattern at late times. However the fastest-growing mode of our analysis determines the wavelength of the pattern and therefore, for small coverages, the size, aspect ratio and separation of an array of islands aligned along the steps. This opens the possibility of inducing the spontaneous formation of an array of quantum structures with the desired size and spacing by controlling the cutting angle of the vicinal surface and the fraction of the surface covered by the material.

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References

[1] Shchukin V A and Bimberg D 1999 *Rev. Mod. Phys.* **71** 1125
Thomas E L 1999 *Science* **286** 1307
Bowden N, Brittain S and Whitesides G M 1998 *Nature* **393** 146
Murray C B, Kagan C R and Bawendi M G 1995 *Science* **270** 1335
Shchukin V A, Ledentsov N N, Kop'ev P S and Bimberg D 1995 *Phys. Rev. Lett.* **75** 2968
Tersoff J, Teichert C and Lagally M G 1996 *Phys. Rev. Lett.* **76** 1675
Liu F, Davenport S E, Evans H M and Lagally M G 1998 *Phys. Rev. Lett.* **82** 2528
Venezuela P, Tersoff J, Floro J A, Chason E, Follstaedt D M, Liu F and Lagally M G 1998 *Nature* **397** 678
[2] Pimpinelli A and Villain J 1998 *Physics of Crystal Growth* (Cambridge: Cambridge University Press)
[3] Tersoff J, Phang Y H, Zhang Z and Lagally M G 1995 *Phys. Rev. Lett.* **74** 3632
Liu F, Tersoff J and Lagally M G 1998 *Phys. Rev. Lett.* **80** 1268
[4] Adam Li, Liu F, Petrovykh D Y, Lin J-L, Viernow J, Himpsel F J and Lagally M G 2000 *Phys. Rev. Lett.* **85** 5380
[5] Villain J 1991 *J. Physique I* **1** 19
[6] Liu F and Lagally M G 1997 *Surf. Sci.* **386** 169
Müller J and Grant M 1999 *Phys. Rev. Lett.* **82** 1736
[7] Bales G S and Zangwill A 1990 *Phys. Rev.* B **41** 5500
[8] Langer J S 1980 *Rev. Mod. Phys.* **52** 1
Langer J S 1987 *Chance and Matter (Les Houches Session XLVI, 1986)* ed J Soulettie, J Vannimenus and R Stora (Amsterdam: North-Holland)
Pelcé P 1988 *Dynamics of Curved Fronts* (San Diego, CA: Academic)
[9] Landau L D and Lifshitz E M 1986 *Theory of Elasticity* (Oxford: Pergamon)
[10] Liu A, Liu F and Lagally M G 2000 *Phys. Rev. Lett.* **85** 1922