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Steps and facets at the surface of soft crystals

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Abstract. We consider the shape of crystals which are soft in the sense that their elastic modulus μ is small compared to their surface tension γ, more precisely $\mu a < \gamma$ where a is the lattice spacing. We show that their surface steps penetrate inside the crystal as edge dislocations. As a consequence, these steps are broad with a small energy which we calculate. We also calculate the elastic interaction between steps a distance d apart, which is a $1/d^2$ repulsion. We finally calculate the roughening temperatures of successive facets in order to compare with the remarkable shapes of lyotropic crystals recently observed by Pieranski *et al.* [1,2]. Good agreement is found.

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1 Introduction

An elastic material is characterized by an elastic modulus μ which is an energy per unit volume. Any interface, whether with a vapour or a liquid phase, has a surface energy γ per unit area. The ratio γ/μ is thus a characteristic length which should be compared to other lengths in the problem, for instance the lattice spacing a. More specifically, let us define

$$d_0 = \frac{(1-\sigma)\,\gamma}{\mu} \tag{1}$$

where σ is Poisson's modulus. Three situations may occur: (i) For usual materials in contact with their vapour, d_0 is smaller than a. Indeed, according to Frank and Stroh [3], a usual value for γ/μ is $0.1a$ and Poisson's modulus is usually equal to 1/3. As a consequence, we will see that steps are edge dislocations bound to the surface. The familiar "terrace, ledge, kink" ("TLK") model [4,5] holds. (ii) At a solid-melt interface γ is smaller because the solid density is not very different from that of the liquid. Then $d_0 \ll a$: elastic strains due to surface perturbations like steps are only small corrections. (iii) It may happen that d_0 is larger than a: then it pays to spend elastic energy in order to gain surface energy. As we shall see below, steps are dislocations which are buried a depth h below the surface. A new length scale appears in the problem. The steps are broad and their energy is strongly reduced by an elastic relaxation. That should always occur if the material is soft enough. A spectacular illustration of this situation is given by the lyotropic crystals recently studied by Pieranski *et al.* [1,2]. These crystals are water solutions of non-ionic surfactant molecules which arrange in a loose cubic structure with a of order 50 Å. The structure is soft ($\mu = 10^7$ erg/cm^3), but the surface tension is only a little less than that of pure water ($\gamma = 25\,$dyn/cm). As a result, the ratio d_0/a is much larger than in usual crystals. This is a typical example of the anomalous case of "soft crystals" which we consider in this article [6].

Pieranski *et al.* [1,2] have observed that the facetting of their lyotropic crystals is remarkable: up to 60 different facets are present on the equilibrium shape. Our first motivation was to understand the origin of the large number of facets on these crystals. As we shall see, it is the large size of the lattice spacing a. Pieranski *et al.* also invoked the large value of the step-step repulsion, but, as we shall see, it is compensated by the small value of the step energy in the calculation of the roughening temperatures. In Section 2, we calculate the unusual structure and the energy of steps in such soft crystals. In Section 3, we calculate the interaction energy of two steps a distance d apart, and we show that it is a $1/d^2$ repulsion whose origin is surface capillary forces within the step profile. In Section 4 we calculate the net macroscopic surface energy of a vicinal surface, with regularly spaced steps, which is the starting point for a discussion of shapes. Section 5 is devoted to the roughening transition of such vicinal surfaces. In the last section, we use our results to calculate how many facets should be observed on the crystals of Pieranski *et al.* [1,2] and we compare with their experimental results.

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2 Structure and energy of a single step

For simplicity we consider a simple cubic crystal with lattice spacing a. If no relaxation occurred, a single step on a (100) facet would expose an extra surface a (per unit length of the step). In this case, the step energy β would be γa. Such a step is a remnant of an edge dislocation along the y axis with a Burgers vector along the z axis as it moves towards the surface. One way to reduce its energy is to move it inside the crystal as an edge dislocation which is now a depth h below the free surface $z = 0$. A similar phenomenon was considered by Lejcek *et al.* [7] in the different context of smectic A liquid crystals. The dislocation introduces an extra lattice plane on half the area and produces a rounded step at the surface. Burying the dislocation rounds off the step and decreases the surface energy. The price to pay is the dislocation elastic energy. As we shall see, this price is low for soft crystals, but it would be too high for usual crystals.

In an infinite medium, the displacement $u_i^{(0)}$ and stress $\sigma_{ij}^{(0)}$ created by the edge dislocation are known [9], as well as the total elastic energy per unit length

$$\beta^{(0)} = \frac{\mu a^2}{4\pi\left(1-\sigma\right)} \ln \frac{R_{\max}}{r_0} \tag{2}$$

where r_0 is an appropriately defined core radius of the order of the lattice spacing a. $R_{\max}$ is the size of the system which will be replaced by h in the presence of a free surface (images suppress divergences at infinity). Qualitatively we can guess the final result easily: the step width at the surface is of order h and the profile lengthening is thus $\approx a^2/h$. Balancing $\beta^{(0)}$ against the capillary energy $\gamma a^2/h$ yields at once $h \approx \gamma(1-\sigma)/\mu = d_0$: steps are buried in soft crystals! But a quantitative discussion requires more effort! In order to calculate this elasticity problem, we shall proceed in four successive steps. As a zeroth order approximation, we will start with an infinite medium, and our notations for stresses and displacements will have a superscript (0). Suppressing the upper half space modifies the strain field in the lower part, thereby adding an extra displacement $u^{(1)}$ at the surface $z = 0$. The energy stored below is modified, and we must subtract the energy originally stored above. In a final step we must add a correction u^{cap} due to the existence of surface tension.

Since we first ignore the effect of surface tension, the normal stresses $\sigma_{zz}^{(0)}$ and $\sigma_{zx}^{(0)}$ have to be cancelled at the crystal surface. If we dealt with screw dislocations, we would treat the elasticity of a half space by introducing image dislocations. However, this method does not work with edge dislocations: adding a symmetric edge dislocation with opposite Burgers vector does cancel out the stress σ_{zz}, but it doubles σ_{zx}! We are thus forced to solve the elastic problem directly. In an infinite medium, the upper half exerts a force $F_i^{(0)} = \sigma_{iz}^{(0)}$ on the lower half. Cancelling this force is the same as applying an opposite force $-F_i^{(0)}$, resulting in an extra displacement $u_i^{(1)}$ and stress $\sigma_{ij}^{(1)}$ on top of the original $u_i^{(0)}$, $\sigma_{ij}^{(0)}$. This extra displacement *at the surface* is

$$u_i^{(1)}\left(x\right) = \int_{-\infty}^{+\infty} \chi_{ij}\left(x-x'\right)\left(-F_j\left(x'\right)\right)\mathrm{d}x' \tag{3}$$

where $\chi_{ij}\left(x-x'\right)$ is the two dimensional surface response function, given for instance in Landau-Lifschitz [9] (see Eq. (47) in the Appendix). The net normal elastic displacement that controls the surface profile is

$$u_z^{\mathrm{el}} = u_z^{(0)} + u_z^{(1)}. \tag{4}$$

In order to calculate the elastic energy, two corrections are needed

(i) Add the energy lowering upon relaxation of the forces $F_i^{(0)}$

$$\begin{aligned}\delta\beta^{(1)} &= \int_{-\infty}^{+\infty}\mathrm{d}x\int_0^1 u_i^{(1)}\mathrm{d}\lambda\sigma_{zi}^{(0)}\left(1-\lambda\right)\\ &= -\frac{1}{2}\int_{-\infty}^{+\infty}\mathrm{d}x\,\mathrm{d}x'\chi_{ij}\left(x-x'\right)\sigma_{zi}^{(0)}\left(x\right)\sigma_{zj}^{(0)}\left(x'\right)\end{aligned} \tag{5}$$

(ii) Subtract the part of $\beta^{(0)}$ that was stored above the surface. The bottom exerts on the top a force $-F_i^{(0)}$, which by itself would create a displacement

$$u_i^{(2)}\left(x\right) = \int_{-\infty}^{+\infty} \overline{\chi}_{ij}\left(x-x'\right)\left(-F_j^{(0)}\left(x'\right)\right)\mathrm{d}x'. \tag{6}$$

This displacement is calculated in a way similar to the previous one ($u^{(1)}$). As explained by Landau [9], it involves a response function $\overline{\chi}_{ij}$ which is the transposed of χ_{ij}, because the medium now sits above instead of below. One has to substract an energy

$$\delta\beta^{(2)} = -\frac{1}{2}\int_{-\infty}^{+\infty}\mathrm{d}x\,u_i^{(2)}\left(x\right)\left[-F_i^{(0)}\left(x\right)\right]. \tag{7}$$

The cross terms cancel out between χ and $\overline{\chi}$ and we are left with a simple result for the elastic energy at this stage:

$$\beta^{\mathrm{el}} = \beta^{(0)} - \int_{-\infty}^{+\infty}\mathrm{d}x\,\mathrm{d}x'\sum_{i=x,z}\left[\chi_{ii}\left(x-x'\right)\sigma_{zi}^{(0)}\left(x\right)\sigma_{zi}^{(0)}\left(x'\right)\right]. \tag{8}$$

As explained in the appendix, we know χ_{ij}, so that we can solve this elastic problem analytically. Note that we could also use the method of images if we added counterforces $2F_x^{(0)}$ along the surface only. Comparing the two approaches is a useful check of a rather cumbersome algebra.

The calculation is most easily carried out in Fourier space, according to the definitions

$$\begin{cases} f\left(x\right) = \displaystyle\int_{-\infty}^{+\infty} f\left(k\right)\mathrm{e}^{\mathrm{i}kx}\mathrm{d}k \\ f\left(k\right) = \displaystyle\frac{1}{2\pi}\int_{-\infty}^{+\infty} f\left(x\right)\mathrm{e}^{-\mathrm{i}kx}\mathrm{d}x \end{cases}. \tag{9}$$

For a periodic force $u_i(k) = \chi_{ij}(k) F_j(k)$. It is easily verified that

$$\chi_{ij}(x) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \chi_{ij}(k) \, \mathrm{e}^{\mathrm{i}kx} \mathrm{d}k. \tag{10}$$

The elastic energy is

$$\beta^{\mathrm{el}} = \beta^{(0)} - 2\pi \int_{-\infty}^{+\infty} \mathrm{d}k \left[\left| \chi_{zz}(k) \, \sigma_{zz}^{(0)}(k) \right|^2 + \chi_{xx}(k) \left| \sigma_{zx}^{(0)}(k) \right|^2 \right]. \tag{11}$$

We only need carry integrals!

But this is not the end of the story. We have not yet included elastic deformations due to the existence of the surface tension, that is to capillarity! The normal stress σ_{zz} at the surface is not zero. The surface is under mechanical equilibrium because the surface stress is compensated by the Laplace force $\varPhi_z$ which is exerted by the surface on the inside. It is a consequence of the curvature in the step profile:

$$\varPhi_z = \gamma \frac{\mathrm{d}^2 u_z}{\mathrm{d}x^2} \cdot \tag{12}$$

That force creates an extra displacement u^{cap} which in turn modifies the profile. We thus write in reciprocal space

$$\begin{aligned} \varPhi_z(k) &= -\gamma k^2 \left(u_z^{\mathrm{el}} + u_z^{\mathrm{cap}} \right) \\ u_z^{\mathrm{cap}}(k) &= \chi_{zz}(k) \, \varPhi_z(k) \end{aligned}$$

whose solution is

$$\begin{aligned} u_z^{\mathrm{cap}}(k) &= -\frac{\alpha_k}{1+\alpha_k} \, u_z^{\mathrm{el}} \\ \alpha_k &= \gamma k^2 \chi_{zz}(k) \, . \end{aligned}$$

Because the displacement u^{cap} modifies $\varPhi_z(k)$, the usual counterforce argument does not apply and it is simpler to write the net capillary energy as the sum of its bare value and the elastic energy stored in producing u^{cap}

$$\beta^{\mathrm{cap}} = 2\pi \int_{-\infty}^{+\infty} \mathrm{d}k \left\{ \frac{\gamma k^2}{2} \left| u_z^{\mathrm{el}}(k) + u_z^{\mathrm{cap}}(k) \right|^2 + \frac{\left| u_z^{\mathrm{cap}}(k) \right|^2}{2\chi_{zz}(k)} \right\} \tag{13}$$

(minimizing with respect to u^{cap} reproduces the expression of u^{cap}). We thus find

$$\beta^{\mathrm{cap}} = 2\pi \int_{-\infty}^{+\infty} \mathrm{d}k \frac{\gamma k^2}{2(1+\alpha_k)} \left| u_z^{\mathrm{el}} \right|^2. \tag{14}$$

As expected, the elastic relaxation reduces the original capillary energy.

Our program is now clear: first we calculate $\sigma^{(0)}(k)$ and $\chi(k)$, then we minimize $\beta = \beta^{\mathrm{el}} + \beta^{\mathrm{cap}}$ with respect to the depth h.

Finally we calculate the step energy, which is the total energy of the dislocation. The algebra is done in Appendix A. It leads to

$$\beta = \frac{\mu a^2}{4\pi(1-\sigma)} \int_0^{1/r_0} \frac{\mathrm{d}k}{k} \left\{ 1 - \mathrm{e}^{-2kh} \times \left[1 + 2kh + 2k^2h^2 - 2kd_0 \frac{(1+kh)^2}{1+kd_0} \right] \right\} \tag{15}$$

where $d_0 = (1-\sigma)\gamma/\mu$ is our characteristic length. The term 1 is the logarithm in $\beta^{(0)}$, the first three terms in the square bracket are the contribution of counterforces and the last one the effect of capillary forces. The denominator $(1+kd_0)$ reflects the self consistent calculation of u^{cap}. Note that capillarity is hidden in the single length d_0. We rewrite the curly bracket as

$$1 - \mathrm{e}^{-2kh} - 2kh \, \mathrm{e}^{-2kh} \left[1 - \frac{d_0}{h} \right] \left[1 + kh \frac{1 - d_0/h}{1+kd_0} \right] \cdot \tag{16}$$

It is easily verified that this quantity is stationary with respect to h when $h = d_0$. That result is an exact version of our original estimate. Note that minimization is the same *for every* k: as a consequence it will persist for two steps or for a regular array. Altogether the total energy takes an incredibly simple form

$$\begin{aligned} \beta &= \frac{\mu a^2}{4\pi(1-\sigma)} \int_0^{1/r_0} \frac{\mathrm{d}k}{k} \left[1 - \mathrm{e}^{-2kd_0} \right] \qquad (17) \\ &= \frac{\mu a^2}{4\pi(1-\sigma)} \left[\varGamma + \ln\left(\frac{2d_0}{r_0} \right) \right] \qquad (18) \end{aligned}$$

where $\varGamma \approx 0.577$ is Euler's constant. All complications have disappeared! In view of the messy intermediate algebra there is probably a reason for that, but we do not know it.

Equation (18) is our first conclusion and it is non trivial: it shows that the energy scale for steps is not γa, but μa^2, down by a factor $\mu a/\gamma \approx a/d_0$. This will affect the roughening transition temperature of vicinal surfaces drastically. Step burying is physically important.

3 The interaction between steps

The interest of Fourier space calculations is the fact that they are immediately extendable to several steps. For a pair of steps a distance d apart, the strain and stress tensors are simply multiplied by a factor $(1+\mathrm{e}^{\mathrm{i}kd})$. The resulting energy integral for two steps is modified by a factor:

$$|1 + \mathrm{e}^{\mathrm{i}kd}|^2 = 2(1 + \cos kd) \, . \tag{19}$$

Note that the depth $h = d_0$ is unchanged, as minimization was achieved for *each* k. The interaction between steps is contained in the term $2\cos kd$. Using the above result for the energy of an isolated step we find that the energy for the two steps becomes:

$$\begin{aligned}2\beta(d) &= \frac{\mu a^2}{2\pi\,(1-\sigma)} \int_0^{1/r_0} \frac{\mathrm{d}k}{k}\left[1-\mathrm{e}^{-2kd_0}\right]\left[1+\cos kd\right] \\ &= \frac{\mu a^2}{2\pi\,(1-\sigma)}\left[\varGamma + \ln\left(\frac{2d_0}{r_0}\right) + \frac{1}{2}\ln\left(1+\frac{4d_0^2}{d^2}\right)\right]\cdot\end{aligned} \tag{20}$$

The interaction energy is contained in the last term and for large d it is:

$$\epsilon_{12} = \frac{\mu a^2 d_0^2}{\pi\,(1-\sigma)\,d^2}\,. \tag{21}$$

It is interesting to compare this global repulsion with the successive results one would get at various steps of an analysis using image dislocations.

(i) Using only image dislocation without compensating forces $2F^{(0)}$ leads to the interaction energy of four dislocations arranged in a rectangle with height $2h$ and base d. We want half the total elastic energy ϵ of these four dislocations since we integrate the elastic energy over a half space. The starting point is the interaction energy of a pair of parallel edge dislocations, with Burgers vectors b_1 and b_2 pointing in the same direction, but either parallel or antiparallel. That interaction is given in the literature [10,11]

$$\epsilon_{12} = \frac{\mu b_1 b_2}{2\pi\,(1-\sigma)}\left[\ln\frac{R_{\max}}{R} - \sin^2\theta\right] \tag{22}$$

where R and θ are the polar coordinates of 2 with respect to 1 (polar axis along b), and $R_{\max}$ an upper cutoff which will disappear for dislocation dipoles. The first term in the bracket yields a radial force which is repulsive for $b_1 = b_2$ but attractive if $b_1 = -b_2$. The dislocation 1 is repelled by its neighbour (dislocation 2), but it is attracted by the image 2' of this neighbour. We also recover the known result that a double dislocation with Burgers' vector $2b$ tends to split into two single dislocations b. The second term in the bracket of equation (22) yields a torque. For our rectangular configuration, and burgers vectors of amplitude a, we find

$$\begin{aligned}\epsilon = \frac{1}{2}\,\frac{\mu a^2}{4\pi(1-\sigma)}&\left[4\ln\frac{R_{\max}}{r_0} - 4\ln\frac{R_{\max}}{2h}\right. \\ &\left.+4\left(\ln\frac{R_{\max}}{d}-1\right) - 4\left(\ln\frac{R_{\max}\cos\alpha}{d} - \cos^2\alpha\right)\right].\end{aligned}$$

In the bracket the first term is the energy of individual dislocations, the second the interaction of genuine dislocations 1 and 2 with their respective images $1'$ and $2'$, the third $(1,2)$ and $(1',2')$, the last one $(1,2')$ and $(1',2)$. The angle α is the tilt of cross links, $\tan(\alpha) = 2h/d$. That result holds for any value of h/d as long as h and d are much larger than a. The elastic interaction between the steps corresponds to the last two terms in the bracket

$$\epsilon_i = -\frac{\mu a^2}{2\pi\,(1-\sigma)}\left[\sin^2\alpha + \ln(\cos\alpha)\right] \tag{23}$$

For small α, *i.e.* large distance d, the bracket is $\alpha^2/2$: the interaction is *attractive*, proportional to $1/d^2$ as befits a step interaction. It turns into a repulsion for larger α, the change corresponding to the minimum of ϵ_i at $\alpha = \pi/4$, *i.e.* $d = 2h$. If true, such a behavior would lead to the prediction of a sharp angular matching at the edge of a flat facet [8].

(ii) The next approximation is the inclusion of counterforces at the surface. One finds that the $1/d^2$ attraction is *exactly* cancelled out. The net step interaction is controlled by the next order in the expansion: it is a $1/d^4$ *attraction.*

(iii) But capillarity ruins the above result. As any relaxation mechanism it induces a $1/d^2$ *repulsion* that supersedes the $1/d^4$ term. We conclude that step interaction is controlled by the elastic strain field which is produced by the surface tension. Just as in the classic picture by Marchenko and Parshin [16] it is repulsive and $\sim 1/d^2$. Our picture does not depart from standard wisdom. Interestingly, equation (21) can be written as

$$\epsilon_{12} = \frac{(1-\sigma)\gamma^2 a^2}{\pi\mu d^2} \tag{24}$$

which looks like Marchenko's result with a force doublet γa on the step; however, the force doublet is μa^2 distributed on a length d_0.

4 The energy of a vicinal surface

In practice we are not interested in a single pair of steps, but in a regular array with period d, describing a vicinal surface with average tilt $\overline{\phi}$ such that $\tan\overline{\phi} = a/d$. Due to that tilt the Burgers vector of dislocations is not exactly perpendicular to the real surface, which strictly speaking makes the elastic calculation far more complicated. We can ignore such a complication if we assume again that the lengths h and d are both much larger than the atomic spacing a: the angle $\overline{\phi}$ is then small and we retain only the lowest order in a/d and a/h while making no assumption on h/d. Once more, the depth h is the same as for a single step.

We want to calculate the total energy per step β and per unit area $U = \beta/d$. We can proceed in the same way we did for two dislocations. Given the Fourier transform of the stress tensor of a single step $\sigma^{(0)}(k)$, we can generate the periodic superposition as follows:

$$\sigma(k) = \sum_{n=-N}^{+N} \mathrm{e}^{\mathrm{i}\,kdn}\sigma^{(0)}(k)\,, \tag{25}$$

where $2N+1$ is the number of steps. If the total energy of a single step is

$$\beta_1 = \int_{-\infty}^{+\infty} b(k)\mathrm{d}k \tag{26}$$

the total energy per step in a periodic arrangement becomes:

$$\beta = \frac{1}{2N+1} \sum_{n,m=-N}^{+N} \int \mathrm{d}k\, \mathrm{e}^{\mathrm{i}\, kd(n-m)} b(k)$$
$$= \frac{2\pi}{d} \sum_{\nu=-\infty}^{+\infty} b\left(\frac{2\pi\nu}{d}\right) \cdot \quad (27)$$

Using the expression for $b(k)$ given in equation (17) we obtain:

$$\beta(d) = \frac{\mu a^2}{4\pi(1-\sigma)} \left[\frac{2\pi d_0}{d} + \sum_{\nu=1}^{d/(2\pi r_0)} \frac{(1-\mathrm{e}^{-4\pi\nu d_0/d})}{\nu} \right] \cdot \quad (28)$$

The energy per unit area is $U = \beta(d)/d$. The single term $\nu = 0$ yields the capillary energy $\gamma a^2/2d^2$ associated to the surface lengthening due to an average tilt $\overline{\phi}$.

The summation can be done analytically for d_0/r_0 large and it gives:

$$\beta(d) = \frac{\mu a^2}{4\pi(1-\sigma)}$$
$$\times \left[\frac{2\pi d_0}{d} + \Gamma + \ln\left(\frac{d}{2\pi r_0}\right) + \ln\left(1-\mathrm{e}^{-4\pi d_0/d}\right) \right],$$

where we used $\sum_{\nu=1}^{\nu_m} 1/\nu = \Gamma + \ln \nu_m + O(1/\nu_m)$ and $\sum_{\nu=1}^{\infty} \mathrm{e}^{-\nu\lambda}/\nu = -\ln(1-\mathrm{e}^{-\lambda})$. We rearrange the expression in order to separate the core dependence. We finally find:

$$\beta(d) = \frac{\mu a^2}{4\pi(1-\sigma)}$$
$$\times \left[\Gamma + \ln\left(\frac{2d_0}{r_0}\right) + \frac{2\pi d_0}{d} + \ln\left(\frac{1-\mathrm{e}^{-4\pi d_0/d}}{4\pi d_0/d}\right) \right] \cdot \quad (29)$$

As mentioned above the relevant scale for the d dependence of β is d_0. The expansion of $\beta(d)$ for large d begins with a positive $1/d^2$ term, indicating repulsion of steps:

$$\beta(d) = \frac{\mu a^2}{4\pi(1-\sigma)} \left[\Gamma + \ln\left(\frac{2d_0}{r_0}\right) + \frac{2\pi^2 d_0^2}{3d^2} \right] \cdot \quad (30)$$

The step-step interaction coefficient is correctly $\sum_{n=1}^{\infty} 1/n^2 = \pi^2/6$ times the two-step interaction energy given in (21). For small d the leading term is simply the capillary energy needed to lengthen the surface in order to tilt it $\sim 1/d$.

5 Roughening temperatures

It is now well known [8,12] that, as their temperature decreases, crystals are facetted in an increasing number of different directions. In any rational direction, there is a roughening transition at a temperature T_{R}. Below T_{R}, the surface is "smooth", *i.e.* facetted or flat. It is localized by the influence of the crystal lattice. Above T_{R}, the surface is "rough" like a liquid surface and rounded on the equilibrium shape of the crystal. The roughening temperature is given by the universal relation [15,12]

$$k_{\mathrm{B}} T_{\mathrm{R}} = \frac{2}{\pi}\, \gamma\, a^2 \quad (31)$$

where a the periodicity of the surface energy along the normal to the interface, in practice the step height. As for γ, it is the value of the surface stiffness at T_{R}. In the case of interest here, where the lattice periodic potential is a small perturbation ("weak coupling"), one can neglect the critical variation of γ near T_{R} and use a temperature independent value for γ.

We are concerned here with facets of a vicinal surface $(1,0,p)$, which correspond to an arrangement of "primary" $(1,0,0)$ steps commensurate with the *in plane* periodicity of the underlying crystal. (If the crystal was periodic in one direction only, it would be a stack of structurless plates, and there would be no $(1,0,p)$ facet.) This problem is discussed in the accompanying paper [17]. The use of the universal formula (31) needs a little care:

(i) a becomes $a(p) = a/p$. Indeed, for a cubic crystal, one goes from one (1,0,p) plane to the next by translating primary steps by a lattice spacing a.

(ii) γ is anisotropic and it should be replaced by the "surface stiffness"

$$\gamma_{ij} = \gamma\delta_{ij} + \frac{\partial^2\gamma}{\partial\theta_i\partial\theta_j} \cdot \quad (32)$$

After diagonalization of γ_{ij}, equation (31) is replaced by

$$k_{\mathrm{B}} T_{\mathrm{R}}(p) = \frac{2}{\pi} \sqrt{\gamma_x \gamma_y}\, a(p)^2 \,. \quad (33)$$

The quantity γ_x is controlled by step compression and it is given by:

$$\gamma_x = \frac{1}{a^2} \frac{\mathrm{d}^2 U}{\mathrm{d}n^2} \cdot \quad (34)$$

As for γ_y, it appears in the energy cost for an undulation of steps which move in the x direction by an amount $u = u_0 \exp(\mathrm{i}k_y y)$. Whenever a step sweeps by a given point the interface is raised by a, from which we infer a height change $\delta z = n a_0\, u$. If each step rotates by an angle $\zeta = \partial u/\partial y$, there is both *lengthening* and *compression* of the steps. Since one has to replace the energy element $\beta(n)\mathrm{d}x$ by $\beta(n/\cos\zeta).(\mathrm{d}x/\cos\zeta)$, the energy change is of order ζ^2, and writes

$$\delta U = \frac{\zeta^2}{2}\left(n\beta + n^2\frac{\mathrm{d}\beta}{\mathrm{d}n}\right) = \frac{n\zeta^2}{2}\frac{\mathrm{d}U}{\mathrm{d}n} = \frac{1}{2}\gamma_y k_y^2 \delta z^2 \quad (35)$$

from which we infer

$$\gamma_y = \frac{1}{n\, a^2} \frac{\mathrm{d}U}{\mathrm{d}n} \cdot \quad (36)$$

Alltogether, we find

$$\sqrt{\gamma_x \gamma_y}\, a^2 \;=\; \sqrt{\frac{U'U''}{n}}\,. \tag{37}$$

Since the interaction at small angle, *i.e.* small n, is proportional to $1/d^2$, the asymptotic form of $U(n \to 0)$ is $n\beta + \delta n^3$ and the effective stiffness writes $\sqrt{6\beta\delta}/a^2$ in this limit. It is independent of the tilt angle or step density. It only involves the product of the step energy β by the step-step interaction coefficient δ that can be inferred from (30).

We are now equipped to infer T_{R} from the preceding section. Note that we do not need to expand in powers of n, we can calculate T_{R} for arbitrary values of $n\,d_0$, the only assumption being $d \gg a$ as before. The d dependence of the factor $\sqrt{U'U''/n}$ is not strong and can be conveniently studied by defining the dimensionless $\overline{T}_{\mathrm{R}}$:

$$T_{\mathrm{R}} = \frac{2}{\pi}\gamma a(p)^2 \overline{T}_{\mathrm{R}}\,. \tag{38}$$

The $d \to \infty$ limit of T_{R} involves only the step energy and the step-step interaction:

$$\overline{T}_{\mathrm{R}}(d \to \infty) = \frac{1}{2}\sqrt{\ln\left(\frac{2d_0}{r_0}\right) + \mathit{\Gamma}} \tag{39}$$

It thus depends on the core radius r_0. The next order in the development in $1/d$ is quadratic and it can change sign as a function of d_0/r_0 due to the fact that the $1/d^4$ term in the interaction energy of two steps is negative (*cf.* Eq. (20)). In the limit where $d \to 0$, $\overline{T}_{\mathrm{R}} = 1$. The full expression coming from (29) interpolates between these two limits and it is shown in Figure 1 for different values of the ratio r_0/d_0. It indicates the departure from a simple quadratic variation of T_{R} with the step density $n = 1/d$.

The crucial result is that T_{R} is of the order $\gamma\, a^2$, despite the fact that elasticity played a central role. The lower energy scale μa^3 is compensated by the lower density scale $1/d_0$: since the effective stiffness has dimension U/n, elasticity drops out. That might be seen as trivial! But a naive argument bared in the usual TLK model would predict a step energy $\beta \sim \gamma\, a$ and an elastic interaction between steps $\delta \sim (\gamma\, a)^2/\mu d^2$. The resulting effective γ is then $\gamma(\gamma/\mu\, a)^{1/2}$: an exact calculation does reduce the scale of T_{R}. As a consequence of the small value of the elastic modulus μ, the step-step interaction is large, but in the expression for the roughening temperature, it appears multiplied by the step energy which is small. We have shown that the product of these two quantities is nearly independent of μ.

6 Comparison with the experimental shape of lyotropic crystals

Let us finally consider the number of facets observed by Pieranski *et al.* [1,2]. These crystals show 6 beautiful staircases of facets around large (1,1,2) facets which we consider as the main ones (see Fig. 5 in Ref. [2]). These crystals are cubic with Ia3d symmetry, which explains why the main facets are not (1,0,0). The last facet, *i.e.* the one with the highest Miller indices, is (9,8,15) (see Fig. 7 in Ref. [2]) at 293 K, the temperature of the experiment. It corresponds to the smallest tilt angle with respect to the (1,1,2) facet, namely 4 degrees. Pieranski *et al.* explain that the distance between equivalent planes in a $[h,k,l]$ direction is given by

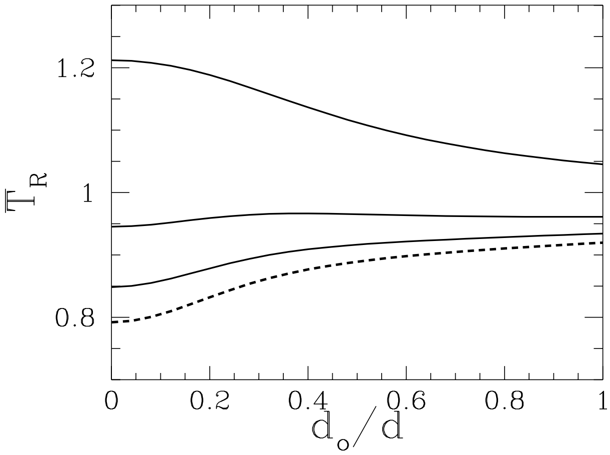

Fig. 1. $\overline{T}_{\mathrm{R}}$ for different values of the parameter d_0/r_0 as a function of $\overline{n} = d_0/d$. The parameter d_0/r_0 is 4, 10, 100. starting from the bottom. The dashed line corresponds to a $d_0/r_0 = 4$ which is a reasonable value for the experiment of reference [1].

$$d_{hkl} \;=\; \frac{118}{\sqrt{h^2+k^2+l^2}}\ \text{Å}. \tag{40}$$

This means that the "primary" step height is $a = 48.2$ Å on (1,1,2) facets. As for the "secondary" steps on vicinal facets, one finds respective heights of 7.02, 6.13, 5.44 and 4.89 Å for (8,7,13), (9,8,15), (10,9,17) and (11,10,19) facets.

The large value of a is responsible for the large number of facets. Indeed, according to equation (31), the roughening temperature of (1,1,2) facets is very high, 26 800 K. We now understand that room temperature (293 K) is very cold compared to the energy scale involved in the roughening of the main (1,1,2) facets. If we ignored the correction $\overline{T}_{\mathrm{R}}$ in equation (38), we would predict that the last facet at 293 K corresponds to a step height slightly larger than $a/\sqrt{26800/293} = 5.04$ Å. This would allow (10,9,17) facets to exist at 293 K.

In the case of their crystals, we find a length $d_0 = 3.46a = 167$ Å. According to our calculation (see Fig. 1) the correction $\overline{T}_{\mathrm{R}}$ is about 0.84 for tilt angles of order 4 degrees, *i.e.* $d_0/d = 0.24$, so that we predict that the last facet should have a step height larger than a minimum value 5.50 Å. This is just enough to avoid the existence of (10,9,17) facets, and allow that of (9,8,15) facets, as seen by Pieranski *et al.* Equivalently, we could say that our predictions for the roughening temperatures are 364 K for (9,8,15) surfaces which have thus to be facetted at

293 K, but 287 K for (10,9,17) surfaces which should thus be rough at 293 K.

Given the number of approximations in our theory, the agreement with the experiment is surprisingly good. The calculation is made for a simple cubic lattice where the Burgers vectors are normal to the main (001) faces. Since Pieranski *et al.* studied lyotropic crystals which have a Ia3d symmetry and tilted Burgers vectors, our model is clearly oversimplified. We believe that two things should remain if one introduced such details in a more accurate calculation:
1- The highest order facet has to correspond to a step height of order one tenth of the main one because $T_{\mathrm{R}0}$ is about 100 times 293 K.
2- The correction $\overline{T}_{\mathrm{R}}$ being smaller than one, one observes a slightly smaller number of facets than what a simple quadratic law would predict.

Eventually, it is interesting to explain why we did not consider entropic interactions between steps. The origin of this other type of interaction is the no-crossing condition of steps [8]. Its amplitude has been calculated [14] and measured [13]. It is

$$\epsilon^{\mathrm{ent}} = \frac{\pi^2}{6}\frac{(k_{\mathrm{B}}T)^2}{\beta}\frac{1}{d^2}\,\cdot \tag{41}$$

Given the low value of the step energy that we have found, one could think that the entropic interaction is a significant effect. In the case of soft crystals, the ratio of the entropic interaction to the elastic interaction is independent of the shear modulus μ. It would be large near the first roughening temperature, but it is very small at temperatures two orders of magnitude lower.

We are very grateful to Pawel Pieranski and Paul Sotta, who introduced us to the physics of their crystals and communicated their last results before publication.

Appendix A: Solution of the 2D elastic problem

In this appendix we provide details on the solution of the two dimensional elastic problem (every quantity depends only on the depth z and the coordinate x). We consider a half space isotropic elastic medium $z < 0$. The basic ingredient is the response function relating the displacement $u_i(x)$ at the free surface to the force $F_j(x')$ applied elsewhere on the same surface

$$u_i(x) = \int_{-\infty}^{+\infty} \chi_{ij}(x - x')\, F_j(x')\,\mathrm{d}x'. \tag{42}$$

The problem is solved *via* Airy functions (see Ref. [9])

$$\sigma_{xx} = \frac{\partial^2 A}{\partial z^2}\,,\ \sigma_{zz} = \frac{\partial^2 A}{\partial x^2}\,,\ \sigma_{xz} = -\frac{\partial^2 A}{\partial x \partial z}\,\cdot \tag{43}$$

For a single Fourier component the biharmonic function $A(x,z)$ is

$$A(x,z) = (a + bz)\exp\left[|k|\,z + \mathrm{i}kx\right]. \tag{44}$$

The constants a and b are fixed by the surface conditions $\sigma_{zi}(0) = F_i$, yielding

$$a = -F_z/k^2, \qquad b = \mathrm{i}\,F_x/k + F_z/|k|\,. \tag{45}$$

From σ_{ij} we infer the strain u_{ij}, then the displacement u_x integrating u_{xx}. We thus know $\partial u_x/\partial z$, from which we infer $\partial u_z/\partial x$: u_z follow upon integration over x. The final result in momentum space is

$$u_i(k) = \chi_{ij}(k)\,F_j(k) \tag{46}$$

$$\chi_{zz} = \chi_{xx} = \frac{1-\sigma}{|k|\,\mu}, \qquad \chi_{xz} = \frac{1-2\sigma}{2\mathrm{i}k\mu}\,\cdot \tag{47}$$

That same result could be obtained from the response to a point force given in reference [9], integrating first on the dummy coordinate y and then Fourier transforming.

The displacement and stress fields due to an edge dislocation along the y axis in an infinite medium are well known

$$u_z^{(0)} = \frac{b}{2\pi}\left[\arctan\frac{x}{\overline{z}} + \frac{1}{2(1-\sigma)}\frac{x\overline{z}}{x^2+\overline{z}^2}\right]$$

$$u_x^{(0)} = -\frac{b}{2\pi}\left[\frac{1-2\sigma}{4(1-\sigma)}\log\left(x^2+\overline{z}^2\right) + \frac{1}{2(1-\sigma)}\frac{\overline{z}^2}{x^2+\overline{z}^2}\right]$$

$$\sigma_{zz}^{(0)} = -\frac{\mu b}{2\pi(1-\sigma)}\,\frac{x\left(3\overline{z}^2+x^2\right)}{\left(x^2+\overline{z}^2\right)^2}$$

$$\sigma_{xz} = \frac{\mu b}{2\pi(1-\sigma)}\,\frac{\overline{z}\left(\overline{z}^2-x^2\right)}{\left(x^2+\overline{z}^2\right)^2}$$

$\overline{z}$ is the distance from the core. If the dislocation is buried at a depth h below the origin, $\overline{z} = z + h$. The corresponding stress and displacement at $z = 0$ are in Fourier space

$$\sigma_{zx}^{(0)}(k) = \frac{\mu b}{4\pi(1-\sigma)}\,|k|\,h\exp\left[-|k|\,h\right]$$

$$\sigma_{zz}^{(0)}(k) = \mathrm{i}\frac{\mu b\,\mathrm{sgn}(k)}{4\pi(1-\sigma)}\left[1+|k|\,h\right]\exp\left[-|k|\,h\right]$$

$$u_z^{(0)}(k) = -\frac{\mathrm{i}b}{4\pi k}\left[1+\frac{|k|\,h}{2(1-\sigma)}\right]\exp\left[-|k|\,h\right].$$

From that we infer the extra displacement $u_z^{(1)}(k)$ due to relaxation of counterforces, in which we identify the contributions due to $F_z^{(0)}$ and to $F_x^{(0)}$

$$u_z^{(1x)}(k) = -\chi_{zx}(k)\,\sigma_{zx}^{(0)}(k) = \frac{b(1-2\sigma)}{8\pi\mathrm{i}k(1-\sigma)}\,|k|\,h\mathrm{e}^{-|k|h}$$

$$u_z^{(1z)}(k) = -\chi_{zz}(k)\,\sigma_{zz}^{(0)}(k) = -\frac{\mathrm{i}b}{4\pi k}\left[1+|k|\,h\right]\mathrm{e}^{-|k|h}.$$

The net surface profile as controlled by elasticity is

$$u_z^{\mathrm{el}} = u_z^{(0)} + u_z^{(1x)} + u_z^{(1z)} \tag{48}$$

It is easily verified that $u_z^{\mathrm{el}} = 2u_z^{(1z)}$: the shape of the surface is controlled by by $F_z^{(0)}$ only. Such a very simple result is not an accident: instead of using a single dislocation with two counterforces $F_z^{(0)}$ and $F_x^{(0)}$, we could have used an image picture with a doubled tangential counterforce $2F_x^{(0)}$. The displacement would then read $u_z^{\rm el} = 2u_z^{(0)} + 2u_z^{(1x)}$; identification of the two expressions of $u_z^{\rm el}$ yields the above result! (As mentioned in the text explicit comparison of the approaches at every stage of the calculation is an useful trick for hunting sign mistakes.)

In addition to the surface profile, we need the net elastic energy which we write as

$$\begin{aligned} U^{\rm el} &= U^{(0)} \\ &- 2\pi \int_{-\infty}^{+\infty} {\rm d}k \ \left[\left| \chi_{zz}(k)\, \sigma_{zz}^{(0)}(k) \right|^2 + \chi_{xx}(k) \left| \sigma_{zx}^{(0)}(k) \right|^2 \right] \\ &= \frac{\mu a^2}{4\pi(1-\sigma)} \int_0^{1/r_0} \frac{{\rm d}k}{k} \left\{ 1 - \left[1 + 2kh + 2k^2h^2 \right] {\rm e}^{-2kh} \right\} . \end{aligned}$$

The infrared divergence at $k = 0$ cancels out (the upper distance cut off is h instead of $R_{\max}$), but the large k divergence remains: it describes the dislocation core! The last step is inclusion of the capillary correction, as described in the main text. The relevant quantity is

$$\alpha_k = \gamma k^2 \chi_{zz}(k) = k d_0 \tag{49}$$

Since the relevant k are $\approx 1/d_0$, the need for a selfconsistent calculation is apparent. The net capillary energy, as "screened" by elastic response, is

$$\begin{aligned} U^{\rm cap} &= 2\pi \int_{-\infty}^{+\infty} {\rm d}k \left\{ \frac{\gamma k^2}{2} \left| u_z^{\rm el}(k) + u_z^{\rm cap}(k) \right|^2 + \frac{\left| u_z^{\rm cap}(k) \right|^2}{2\chi_{zz}(k)} \right\} \\ &= 2\pi \int_{-\infty}^{+\infty} {\rm d}k \frac{\gamma k^2}{2(1+\alpha_k)} \left| u_z^{\rm el}(k) \right|^2 . \end{aligned}$$

The expression for the total energy used in the text follows at once.

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