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Elastic Interaction Between Modulated Steps on a Vicinal Surface

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Abstract. — We study in this paper the elastic interaction between modulated steps, in both homoepitaxy and heteroepitaxy. The influence of this interaction on the step fluctuation spectrum is discussed. There are two important consequences that emanate from our analysis, i) in the homoepitaxy case the elastic interaction favors a modulated step profile. However, the gain in the elastic interaction does not generally overcome the line tension effect and ii) more importantly, for the heteroepitaxial case the elastic contribution to the energy modulation is of the same order as that of the line tension part. Elasticity results in an effective line tension (but singular), which may be either positive or negative depending on the phase shift between two adjacent steps. This analysis shows that the elastic contribution for non-zero wavevectors should be incorporated in any statistical theory of fluctuating steps, if one wants to account properly for experimental observations. We point out similarities between this problem, and others arising in quite different systems. We suggest that, in general, tensorial (dipolar and multipolar) interactions between lines, surfaces, etc. of electrostatic, magnetic, hydrodynamic, etc. nature, should lead to the same generic behaviour.

1. Introduction

Interaction between steps on a vicinal surface of crystals is a long-standing subject. It is a crucial question in many important physical problems, such as the equilibrium shape of crystals, the formation of domains on crystal surfaces, the fluctuation spectrum of steps, the terrace width distribution at equilibrium, etc.

Steps on vicinal surfaces are interface defects leading to elastic stress fields in the bulk. The long range elastic field of steps interferes so that an effective step-step interaction follows. The problem of interaction between straight steps in the framework of continuum elasticity has been the subject of various investigations [1,2]. The most important result which follows from these studies is that the interaction energy of two straight steps, separated by a distance $\ell$, is $\sim 1/\ell^2$ in the homoepitaxy case (i.e., when the last layer of the crystal has the same equilibrium structure as the bulk), and $\sim \ln \ell$ in the heteroepitaxy case (i.e., when the last layers have a different structure than the bulk, due to surface reconstruction or deposition of other species).

Several authors used the result for straight steps, in a mean-field approach, to study both the
fluctuation of steps at equilibrium and terrace width distributions [3], or step dynamics in step flow [4,5]. Incorporation of elasticity in the total energy of step modulations (i.e., for situations with nonzero wavevectors) remains to be formulated, and its far-reaching consequences to be elucidated. Dealing with these questions is the main purpose of this paper. This requires extending the work of Marchenko-Parshin [1] to modulated step profiles. When dealing with these questions we have also recognized that our investigation may be relevant to a wide list of physical systems, as explained below.

Let us outline the main results to emerge from our analysis. Firstly, contrary to what could have been expected à priori, in the homoepitaxial case (where, as we shall see, the step is a location of force doublets) a modulation of the step profile results in a decrease of the elastic energy. An inspection of the order of magnitude of the elastic interaction shows, however, that line tension should inhibit such a (potential) instability. Secondly, for heteroepitaxy (where the step is a location of elastic forces – in contrast to force doublets) the elastic contribution to modes with nonzero wavevectors results in an effective line tension, which can be either positive or negative, depending on the phase shift between two adjacent steps. Moreover, an inspection of the order of magnitude shows that the elastic contribution has the same order of magnitude as the line tension. As a consequence, its contribution must not be disregarded when dealing, for example, with step fluctuations. We shall also see that the elastic repulsion between straight steps (for \( q = 0 \), where \( q \) is the the wavevector) may overcome, in some situations, the diffusive attraction which usually leads to step-bunching during sublimation. In other words, elasticity may inhibit such an instability.

Another important feature of this work is the recognition that the type of (potential) instabilities caused by elastic interactions may arise, in a somewhat similar way, in other systems. The hydrated phospholipidic systems, which exhibit a lamellar phase, are typical examples. Indeed, the dipolar interaction between two layers is reminiscent of force-force interaction between steps. It comes out that a modulation of the bilayers reduces the energy, and may thus account for the appearance of rippled phases in these systems. We shall also suggest some experimental protocols in other systems (e.g., magnetic systems) where the interaction between dipole lines is expected to lead to a spontaneous modulation of the lines.

Here is a brief survey of the organization of the paper. In Section 2, we formulate the problem of step-step interaction for an arbitrary step modulation. We consider the homoepitaxial and heteroepitaxial cases separately. In Section 3 we discuss the order of magnitude of the elastic interaction and its implication on the fluctuation spectrum. Section 4 is devoted to a discussion of the results. Section 5 sums up our results and presents some outlooks. Details of the basic formulation together with some technical computations are relegated to appendices.

2. Elastic Interaction of Modulated Steps

To study the step interaction, we have to establish a close distinction between hetero- and homo-epitaxy. In the first case, the atoms belonging to the last layer are not of the same species as those in the underlying substrate.

Since these questions are not usually substantiated enough, we have felt it worthwhile to devote a detailed discussion to this problem.

Starting from elementary elasticity we shall use concepts of force or force doublet distribution along steps to derive general expressions for the interaction energy between two steps having arbitrary profiles. We shall then specialize our calculation to the case of small perturbations of the step profile. This will be sufficient if one is only interested in the linear stability analysis. Let us discuss the heteroepitaxial and homoepitaxial cases separately.
2.1. THE HETEROEPITAXY CASE. — First we consider an adatom on the substrate. This adatom locally creates a force on the substrate. Let \( f \) denote this force. The elastic displacement created by the force on the plane is given in standard textbooks of elasticity [6], and can be written as

\[
\mathbf{u}^{\text{force}}(\mathbf{r}) = \mathbf{A}(\mathbf{r})f,
\]

where \( \mathbf{r} \) is a two-dimensional vector. \( \mathbf{A} \) is a second-rank tensor:

\[
\mathbf{A}(\mathbf{r}) = \frac{1 + \sigma}{\pi E} \left( \frac{1 - \sigma}{r} \mathbf{I} + \frac{\sigma}{r^3} \mathbf{r} \mathbf{r}^{\perp} \right)
\]

where \( \mathbf{I} \) is the unit tensor, \( \mathbf{r} \mathbf{r}^{\perp} \) the second-rank tensor \( (r^{i}r_{j}) \), \( E \) the Young modulus and \( \sigma \) the Poisson coefficient.

Equilibrium implies that the total force felt by the substrate atoms is zero. However, in general, the dipole is not zero. Thus, the presence of an adatom is modelled by a force dipole. More precisely in order to compute the displacement induced by the adatom, we have to take a force distribution \( f_{i} \) at distance \( a_{i} \) (small compared to \( r \)) around the adatom located at a point \( M \) (Fig. 1), with the total force equal to zero. \( f_{i} \) is the force exerted by the adatom at point \( M \) on the substrate atom \( i \) in its neighborhood. The displacement at a point \( P \) (Fig. 1) is easily obtained from equation (1) to be

\[
\mathbf{u}^{\text{dipole}}(\mathbf{r}) = (\nabla \mathbf{A.a}_{i})f_{i} = \nabla \mathbf{A}(\mathbf{a}_{i}f_{i}^{\perp}) = \nabla \mathbf{A.D}
\]

where \( \nabla \mathbf{A} = (\partial \mathbf{A}_{ij}/\partial x_{\nu}) \) is a third-rank tensor and \( (\mathbf{D}_{\mu\nu}) = (a_{i\mu}f_{i}^{r}) \) is a second-rank tensor of dipolar moments. It is understood that repeated indices are to be summed over. Since \( \mathbf{D} \) is symmetric [1] and does not depend on \( r \), we can transform equation (3):

\[
\nabla \mathbf{A.D} = \left( \frac{\partial \mathbf{A}_{ij}(\partial x_{\nu})}{\partial x_{\nu}} D_{ij} \right) = \left( \frac{\partial \mathbf{A}_{ij}}{\partial x_{\nu}} D_{ij}^{\nu} \right) = \text{div}(\mathbf{A.D})
\]

To calculate the total displacement at \( P \), we have to integrate over all dipoles in the half monolayer limited by the step. Applying the divergence theorem, we find:

\[
\mathbf{u}^{\text{dipole}}(\mathbf{r}) = \int \int \text{d}\tau \text{div}(\mathbf{A.D}) = \int_{S} \text{d}s \mathbf{A.D.n},
\]

where \( n \) is the in-plane unit vector perpendicular to the step. So, the displacement caused by the half monolayer is equivalent to that of a force distribution \( \mathbf{f} = Dn \) localized along the step. In the isotropic case we consider here, \( D_{ij} = D\delta_{ij} \) and the vector \( \mathbf{f} = Dn \) is perpendicular to the step. In equation (5) \( \text{d}\tau \) stands for the surface element on the half monolayer, while \( \text{d}s \) designates the arclength element along the step.

Fig. 1. — Flux of force dipoles across the step give rise to force localized along it.
Thus it appears that the total effect amounts to considering a force distribution along the step. This is not surprising. Indeed, because the Lamé equations are conservative — expressing the fact that a volume interact elastically with its surrounding only through its surface, and thus the total force must be conservative — the integrated displacement over the half monolayer gives a contribution only through the contour, i.e., the step, in terms of a force dipole distribution flux across the step. This amounts to forces localized on the step. In conclusion of this discussion, we reiterate that the elastic problem reduces to considering steps as locations of force distribution.

A localized force \( \mathbf{f}_1 \) on a given step creates an elastic field \( \mathbf{u}_1 \), whose expression is given by equation (1). The interaction energy between two points belonging each to two neighboring steps separated by \( r \) reads (where the forces located at the two points whose interaction is considered are \( \mathbf{f}_1 \) and \( \mathbf{f}_2 \))

\[
\mathcal{E}_{\text{force}} = \mathbf{f}_1 \mathbf{u}_1(r) = \mathbf{f}_2 \mathbf{u}_2(-r)
\]

where \( \mathbf{u}_i \) is the displacement induced at the application point of \( \mathbf{f}_i \) by \( \mathbf{f}_j \). Using expression (1), we obtain:

\[
\mathcal{E}_{\text{force}} = \frac{1 + \sigma}{\pi E} \left( \frac{1 - \sigma}{r} \mathbf{f}_1 \mathbf{f}_2 + \frac{\sigma}{r^3} (r \mathbf{f}_1)(r \mathbf{f}_2) \right).
\]

Note that, fixing \( r \), the minimum energy is reached when all the three vectors \( \mathbf{f}_1, \mathbf{f}_2, r \) are parallel with \( \mathbf{f}_1 = -\mathbf{f}_2 \). For straight steps on a vicinal surface, all forces point to the same direction and this configuration has the maximum energy. Modulating steps modifies this result and, as we shall see, lowers the energy under precise circumstances.

Let us now compute the total step-step interaction energy. This is obviously given by integrating \( \mathcal{E}_{\text{force}} \) over the two step profiles

\[
\mathcal{E}_{\text{f-step}} = \frac{(1 + \sigma)(\rho f)^2}{\pi E} \int \int ds_1 ds_2 \left\{ \frac{1 - \sigma}{r} (\mathbf{n}_1 \mathbf{n}_2) + \frac{\sigma}{r^3} (r \mathbf{n}_1)(r \mathbf{n}_2) \right\},
\]

where \( \rho \) is the density of localized forces at steps (\( \rho \sim 1/a \) where \( a \) is the atomic spacing) and \( f \) the strength of localized forces, \( r \) the distance between two points at the two adjacent steps during integration and \( \mathbf{n}_i \) the local normal to step \( i \). Expression (8) is the general form of the interaction irrespective of the step profile. Since our aim is to investigate the stability of straight steps against infinitesimal perturbations, we shall confine ourselves to computing the step-step energy up to second order in the deformation. Since Fourier modes do not couple to this order, it suffices to consider one Fourier component. Moreover, we shall allow for an arbitrary phase shift between steps. More precisely, two neighboring step modulations are taken to have the following form (see Fig. 2)

\[
y_1 = \epsilon \cos(qx_1) \quad y_2 = \ell + \epsilon \cos(qx_2 + \phi)
\]

The quantities \( \ell, \mathbf{n}, \) etc., which appear in equation (8), are easily expressed in terms of \( y_1 \) and \( y_2 \). The calculation is straightforward, the algebraic details are presented in Appendix A. We find, per unit length of the step

\[
\mathcal{E}_{\text{f-step}} = \frac{(1 - \sigma^2)(\rho f)^2}{\pi E} \left\{ C_1 - \ln(\ell/L) + (\epsilon/\ell)^2 [1 - \ell q K_1(\ell q) \cos \phi] \right\}
\]

where \( C_1 = 2\sigma/(1 - \sigma) \) and \( K_1 \) is the modified Bessel function of first order. \( L \) is the step length.

Figure 3 represents the contribution proportional to \( \epsilon^2 \) (that is, we take the energy origin that corresponds to straight steps) in \( \mathcal{E}_{\text{f-step}} \) versus \( \ell q \) for various values of \( \phi \). A few remarks
Fig. 2. — Modelling of the elastic interaction of two modulated steps.

Fig. 3. — Elastic interaction energy of two modulated steps versus the wave number $q$, for different values of the phase shift, in the heteroepitaxy case.

are in order. From Figure 3 one sees that a modulation automatically leads to an increase of the elastic energy, since all curves have positive values. For in-phase fluctuations ($\phi = 0$) the energy is an increasing function of $q$, while it is a decreasing function for out-of phase fluctuations ($\phi = \pi$). More generally, for $\phi > \pi/2$ the energy decreases, and it increases otherwise.

It is instructive to expand the energy for small values of $q$. To second order in $q$, the energy reads

$$\Delta E_{\text{step}} = \frac{(1 - \sigma^2)(\rho f)^2}{\pi E} \left[ \frac{1 - \cos \phi}{\ell^2} + \frac{1}{2} \left( \frac{1}{2} + \ln 2 - \ln(\ell q) \right) q^2 \cos \phi \right] \epsilon^2$$

(11)

where $\Delta E$ designates the energy by taking the straight step as a reference. The important issue of the present calculation is that the energy behaves as $q^2 \ln q$ (to be compared with the line tension term, which behaves as $q^2$). Since $q\ell \ll 1$ the factor multiplying $q^2$ has a sign fixed by that of $\cos \phi$. For $\phi < \pi/2$, elasticity acts as a positive line tension, whereas for
\[ \phi > \pi/2, \] the elastic effect may be thought of as an effective negative line tension! If the energy magnitude were large enough (\textit{vide infra}), one would expect to be in a situation of spontaneous elasticity-induced morphological instability when non-equilibrium phenomena are involved. The basic reason is that the (non-variational) diffusive mechanism during sublimation enforces the \( \phi = \pi \)-mode. As we shall see later, according to the exploration of various regions of parameter space made so far, it seems that diffusion of adatoms (which is stabilizing during sublimation) inhibits such a potential instability.

A remark should be made at this point. Equation (5) shows that a step can be modelled by a distribution of localized forces. This is a general result: D represents the force–dipole induced by adatoms on the last layer and the localized forces \( \mathbf{f} \) along the steps are simply given by D.n. Assuming that D is isotropic, we conclude that these forces are perpendicular to the step. It is possible to extend this calculation to the anisotropic case. If the exact form of D is known, we can easily deduce the direction and strength of forces at the steps.

An important example is provided by Si(001) where dimerisation occurs on the last layer. There are two kinds of domain, say A and B, separated by steps, and the direction of dimerisation rotate by 90° between consecutive domains. Taking the \( x \)-direction parallel to one of the dimerisation directions, we can model D by [7]

\[
D = \begin{pmatrix}
  d_\parallel & 0 \\
  0 & d_\perp
\end{pmatrix},
\]

(12)

This amounts to localized forces \( \mathbf{f} = \mp(d_\parallel - d_\perp) \) at steps whose direction is opposite from one step to the next one. This situation contrasts with that considered above where D was taken to be isotropic. Of course, in general all crystals are anisotropic, and strictly speaking, D is anisotropic accordingly. The case of Si(001) is special in as much as the anisotropy results in an alternation of the force direction between two adjacent steps due to the \( 1 \times 2 \) and \( 2 \times 1 \) reconstruction. The total step-step energy is given by the same expression as equation (10) multiplied by \(-1\). This entails that in-phase step fluctuations will be enhanced by elastic effects. This is the case studied by Tersoff and Pehlke [8] who found indeed that in-phase step fluctuations were unstable, in agreement with our analysis. It must be emphasized, however, that these authors confined their study to the case of \( \phi = 0 \). Our calculation treats arbitrary fluctuations with regard to the phase. As is clearly seen from Figure 4 the most unstable configuration is obtained for \( \phi = \pi \) and \( q = 0 \), and not \( \phi = 0 \) and \( q = q^* \) (where \( q^* \) is a finite value obtained from a compromise between elasticity and line tension; see Fig. 4(b)). Otherwise stated, elasticity leads to (deterministically straight) step-pairing. Therefore, their conclusion regarding elasticity-induced morphological instability in a realistic situation (i.e., where all phases are permissible) is called into question. We must however be somewhat cautious: their calculation (including the bulk response) is fully anisotropic, and our present remark would be conclusive only after the authors have included an arbitrary phase in their treatment.

Before proceeding farther, let us now have a digression. Since the interaction energy between two forces behaves as in \( 1/r \), its integration over the straight steps diverges. This is the analog of the interaction between two charged lines in electrostatics. This term is not extensive, i.e., the interaction energy of \( N \) steps grows as \( N \ln N \). This longstanding puzzle is still a matter of debate.

2.2. The Homoeptaxy Case. — In this subsection we deal with homoepitaxy. The most important point is that the adatoms of the last layer are of the same nature as those of the substrate. Besides the fact, which holds for all situations, that the total force exerted by the last layer atoms on their neighbors is zero, here the total dipole vanishes too. It should be emphasized, however, that an isolated adatom, be it of the same nature as the underlying
substrate or not, creates a non-zero force dipole (see last subsection). The difference arises only when one considers a half monolayer. Indeed, in the present case the monolayer is not constrained (because there is no misfit). It behaves exactly as the bulk. We can present our reasoning by imagining the crystal structure to be a result of energy minimization with respect to the lattice spacing. Since the half monolayer atoms are in their natural bulk position, this amounts to requiring the force dipole to vanish. One can, if need be, convince oneself by resorting to a broken bond model. Thus, an adatom belonging to the half monolayer has a zero force dipole. The first non-vanishing contribution comes then from force quadrupoles only. A remark is in order. When we refer to the natural position of the atom belonging to the last layer, we have in mind the position in the plane of the surface crystal. Of course, the vertical position is affected by surface relaxation, which decreases exponentially with the penetration distance, as compared to the algebraic decay of displacement field due to in-plane forces. The contribution stemming from surface relaxation can thus be ignored for all practical purposes [9].

Having shown that an adatom on the surface can be modelled by a force quadrupole, we are in a position to determine the step-step elastic energy. The procedure follows exactly that presented above for heteroepitaxy. The total elastic energy associated with the monolayer can be written as being due to force doublets localized along the step, since the flux of quadrupoles implies dipoles along the integration contour. This distribution of force doublets perpendicular to the step is what was initially proposed by Marchenko and Parshin [1] and rederived in a more rigorous manner by Andreev and Kosevitch [2].

An important remark should be made. All we discussed here was about surface defects with zero total moment. However, as pointed out by Marchenko and Parshin [1], steps, as surface defects, must have non-vanishing total moments to compensate capillary forces. Consider a large radius region around a given step. Capillary forces acting on the edge of this region create a moment γa per unit length, where γ is the surface tension of the substrate and a the step height. Internal stress, modelled as force dipole with non-zero total moment and perpendicular to the surface, should compensate the moment induced by capillary forces. However, in the following, we neglect this effect. Actually, we are interested in the interaction energy of modulated steps, as a correction to the zeroth order approximation of straight steps. The derivation of this non-zero total moment dipole was based on capillary forces acting on a large

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Fig. 4. — Step energy in the case of Si(001) (Solid line: φ = π; dashed line: φ = 0) (a) elastic interaction; (b) elastic interaction plus line tension contribution.
The interaction energy between two force doublets (each on a different step) separated by a distance \( r \) is given by

\[
\mathcal{E}_{\text{doublet}} = \frac{(1 + \sigma)(\rho f')^2}{\pi \varepsilon} \left\{ \frac{\sigma}{r^3} - \frac{3\sigma}{r^5} \left[ (r n_1)^2 + (r n_2)^2 \right] + \frac{15\sigma}{r^7} (rn_1)^2 (rn_2)^2 - \frac{3(4\sigma - 1)}{r^5} (rn_1)(rn_2)(n_1n_2) + \frac{2\sigma - 1}{r^3} (n_1n_2)^2 \right\}
\]

(13)

For complementary information about details leading to the above expression see Appendix B (compare with Eq. (7)). Note that here, \( f' \) is the force doublet, i.e., has the dimension of a force multiplied by a length. For a fixed \( r \), the interaction energy is minimum when \( f_1' \) and \( f_2' \) are parallel and make an angle \( \theta \) with \( r \). This angle is given by:

\[
\cos^2 \theta = \frac{6\sigma - 1}{10\sigma}
\]

(14)

which is \( \theta \approx \pi/3 \) for typical values of \( \sigma \), \( \sigma \approx 1/3 \). This effect may favor modulating steps (see below).

To obtain the total step-step interaction energy, expression (13) is integrated over the two step profiles, exactly as in the heteroepitaxy case. When we specialize our expression to small deviations (with one Fourier mode as before) about the straight configuration, we obtain

\[
\mathcal{E}_{\text{d-step}} = \frac{2(1 - \sigma)^2 (\rho f')^2}{\pi \varepsilon \ell^2} + \frac{(1 + \sigma)(\rho f')^2 \varepsilon^2}{\pi \varepsilon \ell^4} \left\{ (1 - \sigma)(6 - (\ell q)^2) + \left[ [(1 - 7\sigma)(\ell q)^5 - \sigma(\ell q)^5] K_1(\ell q) + (1 - \sigma)(-3(\ell q)^2 + (\ell q)^4) K_2(\ell q) \right] \cos \phi \right\}
\]

(15)

As before, \( \rho \) is the number of doublets per unit length of the step. The first term in the above expression is the interaction energy of two straight steps, as given by Marchenko and Parshin [1]. This is the classical \( 1/\ell^2 \) repulsion between steps. We are interested here in the \( \varepsilon^2 \) term in this expression which corresponds to the contribution due to the modulation. Figure 5 displays the behaviour of \( \Delta \mathcal{E}_{\text{d-step}} \) (where the contribution from straight steps has been subtracted) as a function of \( q \ell \). A striking feature of the outcome of this calculation is that elasticity destabilizes straight steps, above a certain value of \( q \ell \approx 1 - 3 \): \( \Delta \mathcal{E}_{\text{d-step}} \) becomes negative). This is a new qualitative result, discovered here for the first time, to the best knowledge of the authors. This is a surprising feature inasmuch as two straight steps having the same sign repel each other. That is to say, contrary to what could have \textit{a priori} been expected on the basis of the well-known repulsion of two straight steps, their modulation results in a \textit{decrease} of energy.

This result can easily be recognized from equation (15) by noting that modified Bessel functions decrease as \( \exp(-\ell q) \) for \( q \gg 1/\ell \), so that \( \Delta \mathcal{E}_{\text{d-step}} \sim -q^2 \) for large values of \( q \), i.e., \( \Delta \mathcal{E}_{\text{d-step}} \) is a \textit{decreasing} function of \( q \) ! Elasticity \textit{at equilibrium} should lead to a spontaneous step modulation if it were acting alone. However, any step modulation is accompanied with a line tension energy. Thus, a modulation can take place only if the elastic energy amplitude overcomes the line tension effect. In fact, as we shall see in the next subsection, the line tension is sufficiently large (typically two or three orders of magnitude larger than the elastic energy) so that it penalizes energetically any modulation.

We would like here to anticipate by making the following important remark. The expression of the interaction energy between forces (as in the last subsection, see Eq. (7)) or force doublets...
Fig. 5. — Elastic interaction energy of two steps in the homoepitaxy case versus the wave number, for different values of phase shift, a) \( \phi = 0 \); b) dashed line: \( \phi = \pi/2 \), solid line: \( \phi = \pi \).

(Eq. (13)) are very reminiscent of dipole-dipole and quadrupole-quadrupole interactions in electrostatics. Therefore, the implication of our results may be generic in the sense that they can apply to electric, magnetic, hydrodynamic, etc. systems as it will be documented in the conclusion of the present paper. In particular, an interesting system is the pure phospholipidic membranes which may organize themselves – for a certain range of temperature and water content – in lamellar phases of the amphiphilic bilayers where the molecules have a polar head [11]. The dipolar interaction energy between two consecutive layers leads to a similar instability [12], which can manifest itself since such a system is devoid of a surface tension. We shall come back to this point later.

Let us come back to expression (15). For small \( q \), the \( \epsilon^2 \) term reads

\[
\Delta E^{d\text{-step}} = \frac{(1 + \sigma)(\rho^f)^2\epsilon^2}{\pi E \ell^4}\{6(1 - \sigma)(1 - \cos \phi) + [\sigma - 1 + \frac{3}{2}(3 - 7\sigma)\cos \phi](\ell q)^2 + O(\ell^3 q^3)\}. \tag{16}
\]

Taking a typical value of \( \sigma \approx 1/3 \), we easily find that for \( \cos \phi < 2/3 \), i.e. approximately \( \phi > \pi/4 \), \( \Delta E^{d\text{-step}} \) is a decreasing function of \( q \) for small \( q \). Here again one sees that elasticity plays the role of an effective line tension.

2.3. ORDER OF MAGNITUDE OF ELASTIC INTERACTIONS. — A potential instability caused by elasticity may manifest itself only if it is capable of overcoming the line tension effect. It is therefore of great importance to provide orders of magnitude of the elastic contribution. Let us concentrate on the small-q regime, where the elastic contribution (due to the step modulation) is \( \sim q^2 \). The \( q^2 \) prefactor is our definition of the amplitude (which we eventually compare to the line tension). Let us consider heteroepitaxy and homoepitaxy separately:

- In heteroepitaxy, it is clear from equation (11) that this amplitude, denoted by \( A_f \) is given by

\[
A_f \sim \frac{(\rho f)^2}{E} \tag{17}
\]

and \( f \) designates a force. We can give a crude approximation of this amplitude based on a dimensional analysis. \( f \) is a force, and depends on the mismatch between atomic
spacing in the surface and in the bulk, \( \delta a/a \). The only way to construct a force is (with
the additional assumption of linear elasticity which implies that \( f \sim \delta a/a \))
\[
f \sim (\delta a/a)Ea^2. \tag{18}
\]
Taking \( \rho \approx 1/a, \delta a/a \approx 3.10^{-2} \) and for \( E \) a typical value for silicon, \( E \approx 10^{11} \) Pa, \( a \approx 3 \) Å, we obtain \( A_f \approx 10^{-12} \) J/m.

The question of order of magnitude is not documented enough in the literature. Alerhand
et al. [7] and Webb et al. [13] used the spontaneous formation of stress domains on Si(001)
to evaluate the force strength at steps. Their analysis yields \( \rho f = 0.07 \) eV/Å². Using
this value, we obtain \( A_f \approx 10^{-12} \) J/m, which is coherent with our dimensional analysis.

This amplitude is to be compared to the line tension. A dimensional analysis (a typical
cohesion energy, a fraction of an eV per an atomic length) leads to \( \gamma \sim 10^{-10} - 10^{-11} \) J/m.
This is consistent with the value measured by Alfonso et al. [14] from the equilibrium
fluctuation spectrum on Si[111]. In the case of Si[001], however, where reconstruction
occurs, this value is much smaller. The value measured by Bartelt et al. [15], which is
based on the study of the step fluctuation spectrum, is in the range of 1–10 meV/Å
for the two types of steps, \( S_A \) and \( S_B \), i.e., \( 10^{-11} - 10^{-12} \) J/m, which is very close to the
amplitude of elastic interaction.

- In homoepitaxy, the amplitude of elastic interaction is obtained from equation (16) (we
  recall that we are interested in the \( q^2 \) term)
\[
A_d \sim \frac{(\rho f')^2}{E\ell^2}, \tag{19}
\]
where \( f' \) designates a force doublet. Note here that the interstep distance \( \ell \) enters
the amplitude, which is a consequence of the algebraic interaction (as opposed to the
logarithmic one).
From a dimensional analysis, we obtain
\[
f' \sim Ea^3. \tag{20}
\]
Taking a typical step spacing \( \ell \approx 100 \) atomic spacing we obtain \( A_d \approx 10^{-13} \) J/m.

Alfonso et al. [14], studied the terrace width distribution on clean Si(111) surface. They
give \( \ell^2 A_d \approx 10^{-30} \) J m. For a step spacing of 100a, we find \( A_d \approx 10^{-15} \) J/m, which is
not too far from the value derived from dimensional considerations.

3. Discussion

The main result obtained here is that, at least in the homo-epitaxy case, values of the line
tension and elastic interaction of steps are close. This should have an important influence on
the analysis of the fluctuation spectrum of steps at equilibrium.

For large wavelengths [10], the fluctuation spectrum of a train of steps reads:
\[
\omega \sim \frac{A_f(1 - \cos \phi)}{\ell^2} + (A_f e^{-\ell q} \cos \phi + \gamma)q^2. \tag{21}
\]
where we have disregarded the ln(\( \ell q \)) which originates from the elastic part.

The two important results can be stated as follows:
1- When ℓ is large, the steps fluctuate independently, i.e., their relative phase is not fixed. At low values of ℓ, however, and for out-of-phase fluctuations, the energy cost is larger than for in-phase fluctuations. As a result, one is tempted to expect that for small ℓ the in-phase fluctuation modes prevail. In a such case elasticity results in an (effective) positive line tension.

2- The elastic term is important only for \( q < \ell^{-1} \). In this regime, elasticity and line tension are additive. For \( \phi > \pi/2 \), elasticity acts as an effective negative line tension. For such a type of modes elasticity may cause a spontaneous modulation, if it is large enough. Whether this type of mode may be relevant or not is at present an open question which requires a detailed statistical study of a fluctuating train. For \( q \gg \ell^{-1} \), the elastic term is unimportant, as represented by the exponential decrease of the Bessel function for large arguments. The interstep distance \( \ell \) plays the role of a cut-off length below which elasticity is irrelevant with regard to step modulations.

Usually, in the experimental analyses of the step fluctuation spectrum, the elastic renormalization of the line tension, which can attain large values, is not taken into account. While the theoretical study of a train fluctuation is a problem which is relatively hard to tackle even at equilibrium, it is an important task for future investigations to properly incorporate elasticity in the model equations.

4. Conclusion and Outlook

To conclude, we extended the problem of elastic interactions to the case of modulated steps with arbitrary profiles. For homoepitaxy, we found that elasticity favors modulated steps! This is a striking feature inasmuch as two steps having the same sign repel each other. This phenomenon occurs irrespective of the phase shift. While line tension should penalize step modulations, this result is interesting by itself on the conceptual level.

Realizing that the general form of the force-force and dipole-dipole interaction energies bear a strong resemblance with electrostatic dipole-dipole and quadrupole-quadrupole interactions, we were naturally led to ask whether the ideas put forward here could apply to a wide list of systems. Of particular interest seem to be the hydrated amphiphilic systems which form – for a certain range of water contents and temperatures – lamellar phases, where the bilayer entities form a parallel array. Upon variation of the control parameter (e.g., the temperature) the lamellae become rippled [11]. To date, the origin of the rippling transition has remained obscure, albeit some key ingredients are beginning to emerge. Since the amphiphilic molecules have a polar head, it was natural to see whether the dipole-dipole interaction between adjacent bilayers may lead to an energy reduction due to a modulation. This is indeed what comes out from our calculation [12]. Because the membrane has (virtually) a zero surface tension (but a rigidity, giving rise to a \( q^4 \) contribution instead of \( q^2 \)), we expect the dipolar energy to become competitive with the rigidity energy. While the order of magnitude of the ripple wavelength which we calculated is smaller by one order of magnitude than that observed, this idea may open an interesting line of future inquiry.

Other situations where our ideas may stimulate new progress concern a variety of systems, going from electrostatics to hydrodynamics. Beside the above cited case, one can, for example, conceive of ways of testing our ideas on macroscopic magnetic systems by setting up experiments with lines supporting magnetic compasses. On the other hand, in hydrodynamics, it is known that two von Karmann vortex lines [16] are susceptible to modulations. Since the hydrodynamic equations are, for a two-dimensional flow, reminiscent of electrostatic equations, it is possible to formulate the line-line interaction in the present terms. Other various examples may be, probably, found in the case of vortex lines in superconductors, etc. Thus, it appears that anisotropic interactions (of vectorial, and tensorial – think of multipoles – origins) may
embrace a disparate variety of physical systems.

The other line of investigation reported here was the discovery of the relevance of the elastic contribution to the total energy modulation in the heteroepitaxy case. Future works on step fluctuations should incorporate such an effect with the aim to interpret experimental results properly. For phase shifts larger than \( \pi /2 \), the effect of elasticity is opposite to that of the line tension; it plays the role of a negative line tension. It is not yet clear whether in equilibrium the modes with \( \phi > \pi /2 \) are relevant or not. This question is currently under investigation.

What might the consequences in the out-of-equilibrium regime be? It is known that during step flow growth [17] the straight step may become morphologically unstable, the mode with \( \phi = 0 \) [18] being the most dangerous one. During sublimation, the straight step profile is stable, while the vicinal surface suffers a step-bunching instability (mode \( q = 0 \) and \( \phi = \pi \)). Because during sublimation the diffusive phenomenon enforces the mode \( \phi = \pi \), we may expect elasticity (recall that for \( \phi = \pi \) it plays the role of a negative line tension) to lead to a morphological instability. The incorporation of elastic effects in the Burton-Cabrera-Frank (BCF) [19] model results in a modified condition at the steps [12]. So far, we have found that during sublimation, diffusion (which is stabilizing) precludes an elastic induced morphological instability. This is attributed to the fact that, generally, the diffusion "force" is much larger than the elastic one.

In conclusion, the work presented here should serve as a basic framework for future investigations dealing with the problem of fluctuating steps. On the other hand, it has led to the identification of the relevance of the (tensorial) interaction in other systems that look extremely diverse. What we have learnt so far is that these types of interactions (e.g., dipolar interaction), familiar as they may appear, lead to unexpected results.

Appendix A

Interaction Energy of Two Modulated Steps: The Heteroepitaxy Case

We derive here the interaction energy of two lines of forces (Fig. 2). Step modulations are defined by:

\[ y_1 = \epsilon \cos(qx_1), y_2 = \ell + \epsilon \cos(qx_2 + \phi). \]  

(A.1)

Their interaction energy reads (Eq.(7))

\[ \mathcal{E}_{\text{step}} = \frac{(1 + \sigma)(\rho f)^2}{\pi E} \int \int ds_1 ds_2 \left\{ \frac{1 - \sigma}{r} (n_1 n_2) + \frac{\sigma}{r^3} (n_1)(n_2) \right\}, \]  

(A.2)

where \( f = fn_s \), and \( \rho \) is the force density at steps. For two points \( P_1(x_1, y_1), P_2(x_2, y_2) \) on steps, we develop terms in integral at the second order in \( \epsilon \). To shorten the notation, we use:

\[ \xi = (x_1 - x_2)/\ell, \quad \bar{\xi} = \epsilon /\ell, \quad \delta q = \ell q, \]

\[ \Delta = \cos(qx_1) - \cos(qx_2 + \phi) = \cos(qx_2 + \delta q) - \cos(qx_2 + \phi), \]

\[ \Pi = \sin(qx_1) \sin(qx_2 + \phi) = \sin(qx_2 + \delta q) \sin(qx_2 + \phi), \]

\[ \Sigma = \sin(qx_2 + \delta q) + \sin(qx_2 + \phi), \]

\[ g_i(\xi) = i(i + 2)(1 + \xi^2)^{-(i+1)/2} - i(1 + \xi^2)^{-(i+2)/2} \]  

(A.3)

Integration of the first term in equation (A.2) reads:

\[ \int \int ds_1 ds_2 \frac{(n_1 n_2)}{r} \]

\[ = \int \int d\xi dx_2 \left\{ (1 + \xi^2)^{-1/2} + \bar{\xi}^2 \left[ \Pi \xi^2 (1 + \xi^2)^{-1/2} + (1/2) \Delta^2 g_i(\xi) \right] \right\} \]
\[ = -L \ln (\ell/L') + L \left\{ 1 + \left[ \tilde{q}^2 K_0(\tilde{q}) + \tilde{q} K_1(\tilde{q}) - \tilde{q}^2 K_2(\tilde{q}) \right] \cos \phi \right\} \tilde{e}^2 \]
\[ = -L \ln (\ell/L) + L \left[ 1 - \tilde{q} K_1(\tilde{q}) \cos \phi \right] \tilde{e}^2. \]  

(A.4)

where \( L \) is the step length.

Integration of the second term in equation (A.2) gives

\[
\int \int d\xi_1 d\xi_2 \left\{ (1 + \xi^2)^{-3/2} - 3\tilde{e}^2 (2\Delta^2 + \tilde{q}\Delta \Sigma \xi) (1 + \xi^2)^{-5/2} + (1/2)\tilde{e}^2 \Delta^2 g_3(\xi) + \tilde{e}^2 \left[ \Delta^2 + \tilde{q}\Delta \Sigma \xi + \tilde{q}^2 \Pi \xi^2 \right] \right\} = L + L\tilde{e}^2 \left\{ -\tilde{q}^2 K_0(\tilde{q}) - 2\tilde{q} K_1(\tilde{q}) + \tilde{q}^3 K_2(\tilde{q}) + 5\tilde{q}^2 K_2(\tilde{q}) - \tilde{q}^3 K_3(\tilde{q}) \right\} \]
\[ = L + 0! \]  

(A.5)

Surprisingly, \( \tilde{e}^2 \) terms in equation (A.5) vanish exactly. For this term, the energy decrease due to modulation compensates exactly the energy increase due to augmentation of forces per projected length.

Adding these two contributions (A.4-A.5), we get the expression given in equation (10).

Appendix B

Interaction Energy of Two Modulated Steps: The Homoepitaxy Case

Here we compute expression (15), using the energy of two force doublets given by equation (13). We use notations of Appendix A. The interaction energy of two modulated steps reads

\[ E_{\text{step}} = \int \int d\xi_1 d\xi_2 E_{\text{doublet}}(\xi_1', \xi_2', \mathbf{r}). \]  

(B.1)

Integration of different terms of equation (13) gives

\[
\left( \ell^2/L \right) \int \int d\xi_1 d\xi_2 \frac{1}{r^3} [ (\mathrm{rn}_1)^2 + (\mathrm{rn}_2)^2 ]
\]
\[ = 2 + \left\{ 6 + q^2 - \left[ \tilde{q}^2 K_2(\tilde{q}) - \tilde{q}^3 K_3(\tilde{q}) \right] \cos \phi \right\} \tilde{e}^2 \]
\[ = 2 + \left\{ 6 + q^2 - \left[ \tilde{q}^3 K_1(\tilde{q}) + 3\tilde{q}^2 K_2(\tilde{q}) \right] \cos \phi \right\} \tilde{e}^2. \]  

(B.2)

\[
\left( \ell^2/L \right) \int \int d\xi_1 d\xi_2 \frac{1}{r^5} \left[ (\mathrm{rn}_1)^2 + (\mathrm{rn}_2)^2 \right]
\]
\[ = 8/3 + \frac{1}{3} \left\{ 24 + 2q^2 + \left[ -4\tilde{q}^3 K_1(\tilde{q}) - 4\tilde{q}^2 K_2(\tilde{q}) + 4\tilde{q}^4 K_2(\tilde{q}) + 10\tilde{q}^3 K_3(\tilde{q}) - 2\tilde{q}^4 K_4(\tilde{q}) \right] \cos \phi \right\} \tilde{e}^2 \]
\[ = 8/3 + \frac{1}{3} \left\{ 24 + 2q^2 + \left[ -6\tilde{q}^3 K_1(\tilde{q}) - 12\tilde{q}^2 K_2(\tilde{q}) + 2\tilde{q}^4 K_2(\tilde{q}) \right] \cos \phi \right\} \tilde{e}^2. \]  

(B.3)

\[
\left( \ell^2/L \right) \int \int d\xi_1 d\xi_2 \frac{1}{r^7} (\mathrm{rn}_1)^2 (\mathrm{rn}_2)^2
\]
\[ = 16/15 + \frac{1}{15} \left\{ 48 - 4q^2 + \left[ 8\tilde{q}^4 K_3(\tilde{q}) - 12\tilde{q}^3 K_3(\tilde{q}) + 9\tilde{q}^4 K_4(\tilde{q}) - \tilde{q}^5 K_5(\tilde{q}) \right] \cos \phi \right\} \tilde{e}^2 \]
\[
\begin{align*}
= & \frac{16/15}{15} \left\{ 48 - 4q^2 + \left[ -6q^3 K_1(\bar{q}) - q^5 K_1(\bar{q}) - 24q^2 K_2(\bar{q}) + 5q^4 K_2(\bar{q}) \right] \cos \phi \right\} \bar{\epsilon}^2. \\
(\ell^2/L) \int \int ds_1 ds_2 \frac{1}{r^5} (r n_1)(r n_2)(n_1 n_2)
= & \frac{4/3}{3} \left\{ 12 - 2q^2 + \left[ 3q^3 K_1(\bar{q}) - 2q^2 K_2(\bar{q}) + 2q^4 K_2(\bar{q}) + 5q^3 K_3(\bar{q}) - q^4 K_4(\bar{q}) \right] \cos \phi \right\} \bar{\epsilon}^2 \\
= & \frac{4/3}{3} \left\{ 12 - 2q^2 + \left[ 2q^3 K_1(\bar{q}) - 6q^2 K_2(\bar{q}) + \bar{q}^4 K_2(\bar{q}) \right] \cos \phi \right\} \bar{\epsilon}^2. \\
(\ell^2/L) \int \int ds_1 ds_2 \frac{1}{r^3} (n_1 n_2)^2
= & 2 + \{ 6 - \bar{q}^2 + \left[ 2q^3 K_1(\bar{q}) + \bar{q}^2 K_2(\bar{q}) - \bar{q}^3 K_3(\bar{q}) \right] \cos \phi \} \bar{\epsilon}^2 \\
= & 2 + \{ 6 - \bar{q}^2 + \left[ q^3 K_1(\bar{q}) - 3q^2 K_2(\bar{q}) \right] \cos \phi \} \bar{\epsilon}^2.
\end{align*}
\]

Summing these different contributions with their coefficients (which depend on \(\sigma\) as given by equation (13)), we obtain expression (15).

References

[10] Note that here we write the excitation spectrum where we assume that the only dissipation mechanism is at the step: atoms are absorbed and emitted at a finite rate. When other dissipation mechanisms are considered, we expect a modification in the spectrum.