

Elastic Interaction between Surface Defects in Thin Layers

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Elastic interactions between defects are investigated in thin layers. Unlike the classical repulsive law $1/r^3$ (r = defect separation), we discover that defects in thin layers may either attract or repel each other depending on the direction (though elastic deformation is isotropic) with respect to the local geometric force distribution caused by the defect. Moreover, the force distribution fixes the exponent in the power law $1/r^n$ (e.g., in a square lattice $n = 4$). We discuss the implication of this new behavior in various situations. [S0031-9007(98)08337-9]

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Elastic interactions embrace a myriad of situations ranging from physics to biology. They may come to the fore in a number of situations. To name but a few examples, they play a crucial role in the following: (i) metallurgy [1]; (ii) molecular beam epitaxy (MBE), especially in the fabrication of low dimensional nanostructures [2] (e.g., quantum dots and wires); (iii) in gels [3] (e.g., they are responsible for various phase transitions); (iv) in biological membranes (insertion of proteins leads to elastic distortions) [4]. A natural way for tackling elastic effects of an object immersed in a given system (e.g., an island grown on a substrate) is to start from elementary effects, such as the elastic strain caused by a localized force, and then sum up the contributions due to extended effects (lines—steps on vicinal surfaces; islands; etc.).

The study of elastic deformations due to point defects in the bulk is rather an old topic. It has given rise to various contributions, essentially developed in the 1950s and 1960s [5]. The study of surface defects has turned out to be more subtle [6–9]. It has been shown that identical point defects on the surface repel each other as $1/r^3$, r being their separation length. This result holds for a semi-infinite medium.

There are several important circumstances, however, where the medium is of finite thickness. Typical situations where 2D elasticity is particularly important concern Langmuir monolayers, phospholipidic bilayers, van der Waals epitaxy [10], adsorbed layers on a substrate [11], etc. A problem of much current interest for the latter example is multilayer growth and nanostructures (e.g., quantum dots [2]). It is therefore highly desirable to analyze elastic effects with a finite thickness, and ultimately in two dimensions, and to discuss their far reaching consequences. This is the main goal of this Letter.

Let us consider the situation where an atom (or molecule) is deposited on top of an elastic medium (semi-infinite solid, thin layer, etc.). On a time scale larger than that associated with sound propagation, the total force

caused by the atom on the medium is zero ($\sum_j \mathbf{f}_j = 0$; $j = x, y, z$). However, the total dipolar moment (not to be confused with the force moment usually used in mechanics) is not zero, $D_{ij} = \sum_l x_i^l f_j^l \neq 0$ (l runs over the forces around the defect). So the first effect of the defect is due to the moment.

Assuming that each defect creates a central force distribution, it has been shown [6] that the interaction energy between two defects on the surface of a semi-infinite medium is given by

$$E_{\text{int}} = \frac{\pi(1 - \sigma^2)a^2 f^2}{Er^3}, \quad (1)$$

where r is the interdefects distance, E the Young's modulus, and σ the Poisson ratio. This result shows that two identical defects repel each other. This is a classical result that plays a role of reference in this work.

We now turn to thin films. The ultimate limit is a very thin plate with an atomic thickness. In the absence of buckling, the displacement field $\mathbf{u}$ is purely two dimensional, and is a function of (x, y) . In that case the Lamé equation in the presence of a localized force is given by [12]

$$\nabla^2 \mathbf{u} + \frac{1 + \sigma}{1 - \sigma} \nabla(\nabla \cdot \mathbf{u}) = -\frac{2(1 + \sigma)}{Eh} \mathbf{f} \delta(\mathbf{r}). \quad (2)$$

Here Eh is the effective two dimensional Young's modulus (having the dimension of a force per unit length). For practical application in pure 2D we should substitute formally Eh by E_{2D} . For a film with finite thickness, E has the meaning of the true bulk Young's modulus, and h is the thickness. In that case h originates from integration of the displacement in the z direction [12]. It is easy to check that

$$\mathbf{u} = \frac{(1 + \sigma)^2}{2\pi Eh} \left[\frac{\sigma - 3}{1 + \sigma} \mathbf{f} \ln(r) + \frac{\mathbf{r}(\mathbf{r} \cdot \mathbf{f})}{r^2} \right] \quad (3)$$

is a solution. A simple dimensional analysis would lead to the result that the interaction between two defects in 2D should behave as $1/r^2$. We show here that this is in general not the case. Moreover, we show that the interaction may be either attractive or repulsive.

Let us first concentrate on the situation where each defect creates a central force distribution. We begin with the case of two opposite forces (dipole), with $2a$ being their separation. The dipolar field is given by $\mathbf{u}_d = \mathbf{u}(\mathbf{r} + \mathbf{a}) + \mathbf{u}(\mathbf{r} - \mathbf{a})$ [where $a \ll r$, and $\mathbf{u}$ is given by (3)]. A straightforward integration over θ (in polar coordinates) yields $\mathbf{u}_T = (1 - \sigma^2)af\mathbf{r}/2Ehr^2$. This expression bears a resemblance to that of the electric potential created in 2D by a central dipolar distribution. The interaction energy with a second defect at a distance r is given by $E_{\text{int}} \sim \int \mathbf{u}_T \cdot \mathbf{f}(r)d\mathbf{r}$. In view of the Gauss theorem this contribution vanishes identically. (This is easily checked since $\nabla \mathbf{u}_T = 0$.) That is to say, two defects with central force distributions in an infinite and isotropic medium do not interact elastically. The same result holds in 3D [13], and in fact in any dimension.

For a finite system the problem must be solved with appropriate boundary conditions. This entails that defects interact through their images. This leads in 2D [14] to an interaction which is typically of the order $f^2 a^2 / EL^2$, where L is the linear dimension of the system (we assume that r is small as compared to L). This is a vanishingly small contribution for large systems. This is an apparently striking feature. It is resolved by noting that the force distribution in a crystal is obviously not central. This means that a defect detects the discrete nature of matter, which is clearly anisotropic. As a consequence the *defects do indeed interact directly*. Even though the underlying elastic distortion may in some cases be isotropic (in particular a hexagonal 2D crystal obeys pure isotropic elasticity theory), the discrete nature of force distribution is felt at a long distance. It is not necessary as in 3D [15] to reconsider elasticity theory in a discrete version.

If a defect creates locally a n -fold force distribution (Fig. 1), using (3), we obtain

$$E_{\text{int}} = \frac{(1 + \sigma)f^2 g(\theta)a^n}{\pi E h r^n}, \quad (4)$$

where $g(\theta)$ is a function to be specified below, and θ represents the angle between the x axis and the vector $\mathbf{r}$. Let us consider explicitly some examples:

(i) In the presence of two opposite forces (Fig. 1a) the interaction behaves as $1/r^2$, and it can be both attractive or repulsive. Let us outline how this result is obtained. Each force creates a field given by (3). Let $\mathbf{u}_i^{(1)}$ ($\mathbf{u}_i^{(2)}$) denote the displacement field due to defect “1” (“2”) and $\mathbf{f}_i^{(1)}$ ($\mathbf{f}_i^{(2)}$) the i th force in the substrate. The interaction energy between two defects is defined as $E_{\text{int}} = -1/2 \sum_i (\mathbf{f}_i^{(2)} \cdot \mathbf{u}_i^{(1)} + \mathbf{f}_i^{(1)} \cdot \mathbf{u}_i^{(2)}) = -\sum_i \mathbf{f}_i^{(2)} \cdot \mathbf{u}_i^{(1)}$ (the defects are identical). This reads explicitly (see Fig. 1a)

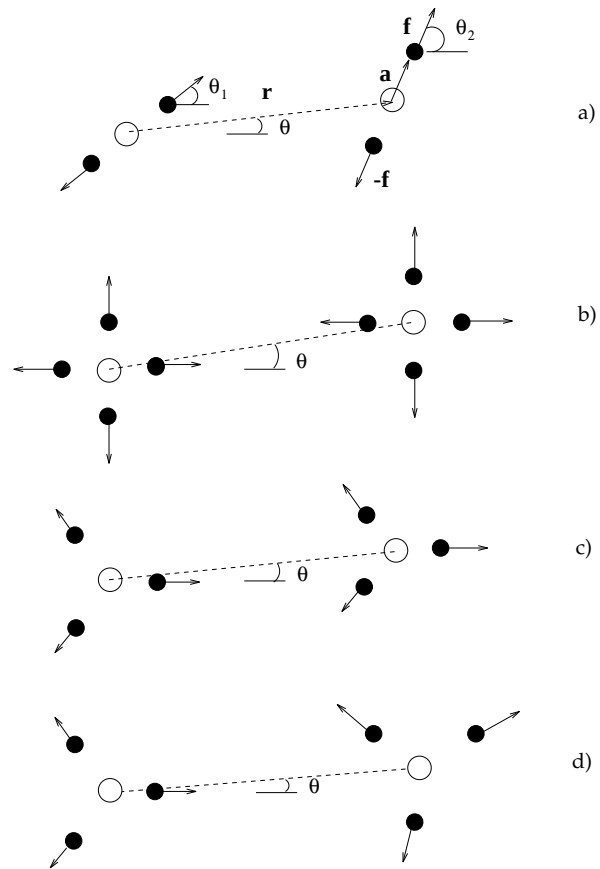


FIG. 1. Different geometries of the force distribution imposed by the crystal symmetry.

$$E_{\text{int}} = -\mathbf{f}^{(2)} \cdot \mathbf{u}(\mathbf{r} + \mathbf{a}) + \mathbf{f}^{(2)} \cdot \mathbf{u}(\mathbf{r} - \mathbf{a}). \quad (5)$$

Using for $\mathbf{u}$ expression (3), and expanding to leading order in a/r , we obtain expression (4) with $n = 2$, and

$$g(\theta) = -2(1 + \sigma) \left\{ \frac{(\sigma - 1)}{(1 + \sigma)} [\cos[2(\theta - \theta_1)] + \cos[2(\theta - \theta_2)]] - \cos[4\theta - 2(\theta_1 + \theta_2)] \right\}. \quad (6)$$

The interaction is attractive or repulsive depending on θ .

(ii) For a fourfold symmetry (Fig. 1b), we find in a similar manner that the first order contribution ($\sim 1/r^2$) vanishes identically due to symmetry. Higher order expansion in powers of a/r leads to $n = 4$. In this case we obtain

$$g(\theta) = (25 - 5\sigma) \cos(4\theta). \quad (7)$$

Here again both attraction and repulsion are possible. Indeed, the interaction is repulsive in the interval $-\pi/8 < \theta < \pi/8$, attractive for $\pi/8 < \theta < 3\pi/8$, and so on. Note that contrary to what could have been expected from a dimensional analysis ($1/r^2$), due to the

medium isotropy, this first contribution vanishes exactly. If allowance is made for crystal anisotropy—that is if the elastic distortions are taken to be anisotropic—analytical results [like Eq. (3)] are not available. We have developed a numerical method to handle this situation. We will simply outline briefly the method below.

(iii) For a threefold symmetry (Fig. 1c), several situations can be encountered. If the defect creates forces at the nodes of the hexagons, we find $E_{\text{int}} \sim 1/r^4$. Here,

$$g(\theta) = -27 \frac{(1 + \sigma)}{2} \cos(6\theta). \quad (8)$$

The interaction is attractive in the sector $-\pi/12 < \theta < \pi/12$, repulsive when $\pi/12 < \theta < \pi/4$, and so on. Another situation may arise. This is the case where the vertex around the defect may rotate from one position to another (Fig. 1d). We find $E_{\text{int}} \sim 1/r^3$. The force configuration in question may be typical in hexatic phases [16]. Here,

$$g(\theta) = 9 \frac{(1 - \sigma)}{4} \cos(3\theta). \quad (9)$$

For simplicity we give here the expression for the case where the relative rotation is π . The interaction is repulsive in the sector $-\pi/6 < \theta < \pi/6$, attractive for $\pi/6 < \theta < \pi/2$, and so on. Extensive discussions will be presented in the future.

In order to treat anisotropic elastic deformations, a numerical analysis seemed necessary. This step will allow us to check our analytical results in the case where isotropy is assumed. The numerical strategy is as follows. We consider atoms in a lattice of a given crystal symmetry. Each atom is coupled to its nearest neighbors with a constant springlike interaction. Let $\mathcal{M}$ denote the dynamical matrix associated with the oscillators, and $\mathbf{v}$ be the displacement vector of each atom ($\mathbf{v}$ has a dimension $2 \times N$, where N refers to the number of atoms, and the “2” to the two degrees of freedom in 2D). In the presence of two defects “1” and “2”, ϕ_1 and ϕ_2 represent the potential energy of interaction between the defects and the other atoms. The total energy in the harmonic approximation takes the form

$$E = \frac{1}{2} \mathbf{v}^t \mathcal{M} \mathbf{v} + \phi_1 + \phi_2. \quad (10)$$

The equilibrium value of the displacement is obtained by minimizing E with respect to $\mathbf{v}$, yielding

$$\mathbf{v} = \mathcal{M}^{-1}(\mathbf{f}^{(1)} + \mathbf{f}^{(2)}), \quad (11)$$

where we have set $\mathbf{f}^{(i)} = -\partial\phi_i/\partial\mathbf{v}$, the force. Reporting into (10) and subtracting the contribution due to the defects if they were alone, we obtain for the interaction energy $E_{\text{int}} = -1/2(\mathbf{f}^{(2)} \cdot \mathbf{v}^{(1)} + \mathbf{f}^{(1)} \cdot \mathbf{v}^{(2)})$. The question amounts thus to inverting the matrix $\mathcal{M}$ [Eq. (11)]. Two important remarks have to be made. First, $\mathcal{M}$ has to be

inverted after projecting the vectorial space of displacement into subspace orthogonal to any degenerate modes. Second, periodic boundary conditions have to be considered in order to take advantage of the translation invariance of $\mathcal{M}$ and calculate its inverse on the plane wave basis. Inversion of $\mathcal{M}$ in the case of a square lattice has led to the following results. (i) If the dynamical matrix is taken to be that of an isotropic medium we find the above-mentioned $1/r^4$ for a fourfold force distribution, and $1/r^2$ for a twofold distribution. (ii) If $\mathcal{M}$ is taken to be fully anisotropic, then we find for the fourfold distribution that the leading contribution is $1/r^2$ (instead of $1/r^4$). Here again both attraction and repulsion are possible.

The question thus arises of to what extent interaction laws in a thin, but finite, film would produce the semi-infinite limit [11]. Qualitatively, we expect that if defects interdistance is large in comparison to the film thickness, then the interaction should be effectively of a 2D nature. Conversely, for short separations in comparison to the thickness, semi-infinite behavior should prevail. Evaluation of the elastic field in a semiexplicit form (in a form of integrals that are to be tabulated) created by a localized force at the surface of a thin film are known [17], but are too much involved in order to be listed here. Using that field it is possible to evaluate numerically the defect-defect interaction by using the very definition of the interaction energy presented above. We find that at $r/h \ll 1$ the interaction is repulsive and is given by $1/r^3$, as expected. At larger separation ($r/h \gg 1$), we recover the above-discussed 2D interaction. If the orientation in 2D is such that the interaction is attractive, the full interaction exhibits a minimum at $r \sim h$, as shown in Fig. 2.

It must be emphasized that the film need not be isolated. It can adhere to a substrate of arbitrary thickness. The 2D results apply, then, provided that the two defects are at a distance of the order or larger than the film thickness [11].

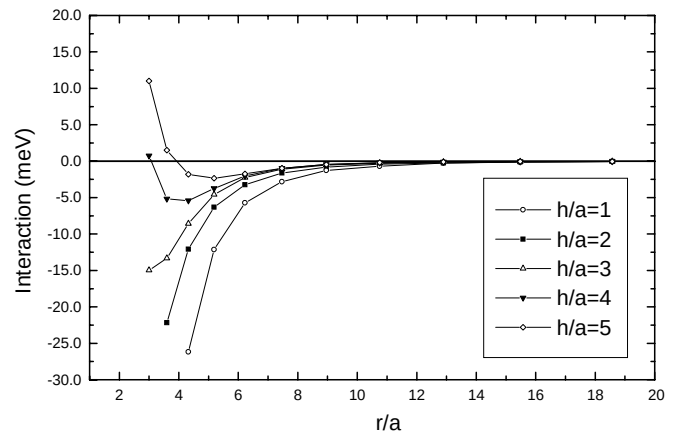


FIG. 2. Elastic interaction in the case of a fourfold symmetry (Fig. 1b) for different layer thicknesses. $F \approx 3 \times 10^{-9}$ N, $\sigma \approx 0.3$ and $E \approx 3 \times 10^{11}$ Pa (typical silicon values).

Let us present further results along with some general implications. Steps, or a chain of atoms on a thin film, are locations of force dipole [18]. As seen above, a dipole creates a field which behaves as $1/r$. In order to evaluate the field due to a step, we integrate this interaction over a line from $-\infty$ to ∞ . We find that the interaction energy between two linear defects vanishes identically, since it involves the combination $u_s(\ell + a)f - u_s(\ell - a)f$ (u_s is the displacement due to the step—independent of the distance—and ℓ is the interstep distance). In contrast, if a semimonolayer of some nature is deposited on top of a substrate of a different nature, then the edge is a location of force monopoles [18] (this holds also in general if the surface stress tensor is anisotropic; that is if its values on both sides of the linear defect are different). We find in that case, upon integration of (3) along the line, the displacement due to a step. Multiplying by a force and integrating over a unit length leads to the edge-edge interaction energy

$$E_{\text{int}} = -\frac{N(1 - \sigma^2)}{Eh} \mathbf{f}^{(1)} \cdot \mathbf{f}^{(2)} \ell, \quad (12)$$

where N is the density of monopoles. This interaction is attractive if the two monopoles are antiparallel, and repulsive otherwise. This result differs from the classical law $\ln \ell$ encountered for a semi-infinite medium. The present interaction is much stronger.

Another important implication concerns the quantum dots fabrication. It is still a puzzling question of how and by which mechanism island organization (e.g., InGaAs [2]) takes place. A moving atom (say during growth and nucleation) would follow a path of attraction until it sticks to an island. Thus one would expect clustering to be enhanced. If a film of finite thickness h is on top of a substrate (as is often the case in the production of nanostructures) repulsion takes place at shorter distances and clustering may become prohibited. Atoms are attracted at a large distance (along the sectors of attraction). When they approach the island, the atoms feel a repulsion (when the distance to the island becomes smaller than h). This will give rise to a typical length scale of an elastic nature which should lead most likely to a selection of a typical length separation between growing islands. We are currently dealing with this question by means of Monte Carlo simulations.

Finally, this work can be of interest to a wide variety of systems. For example, Langmuir monolayers are truly two dimensional objects. Thus defects on the monolayers should lead to interactions of the nature presented here. The same should hold when macromolecules are inserted into phospholipidic bilayers. However, in these systems buckling may become decisive. Thus it is important when dealing with 2D objects to identify first the soft degrees of freedom for a given force configurations. Determining the microscopic configuration of the force distribution requires *ab initio* calculation. Moreover, if out-of-plane

deviations are large enough, a nontrivial coupling between in and out-of-plane strain may arise. It is known that buckling belongs, strictly speaking, to a domain where leading order elastic theory does not apply. The full deformation equations are nonlinear.

In summary, this work has given a first set of nontrivial results on defect elasticity theory in 2D and thin films (be it isolated or on top of a substrate). We have given a short list on general implications. This study opens several issues in different fields.

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