

Revisiting Elastic Interactions between Steps on Vicinal Surfaces: The Buried Dipole Model

G. Prévot and B. Croset

Groupe de Physique des Solides, Universités Paris 6 et 7, UMR-CNRS 75-88, 140, rue de Lourmel, 75015 Paris, France
(Received 22 January 2004; published 25 June 2004)

We expose a new analytical method for computing elastic displacements and interactions due to steps on vicinal surfaces. The model of a “buried dipole” allows us to take into account the specific geometry of the step while performing anisotropic linear elasticity calculations. The displacements found show a remarkable agreement with molecular dynamics simulations for Cu and Pt (001) and (111) vicinals. The interaction energy between steps strongly depends on the dipole direction.

DOI: 10.1103/PhysRevLett.92.256104

PACS numbers: 68.35.Gy, 46.25.Cc

Vicinal surfaces have been the subject of numerous fundamental studies concerning, for example, crystal shape, faceting, or roughening transition [1–6]. They are today widely studied due to their interest in catalysis [7] or for growing well-ordered thin films or nanostructures [8,9], steps acting as a regular array of nucleation centers. If the miscut gives the mean interstep distance, the step spacing regularity depends, in particular, on the step interactions. Conversely, these interactions can be derived from terrace width distribution measurements [2,3].

Since the pioneering prediction of Marchenko and Parshin (MP) [10], steps have been shown to interact elastically. In their model, steps on a vicinal surface are equivalent to lines of point dipoles on the nominal surface. Isotropic calculations lead to a E_2/D^2 repulsive interaction energy between identical steps separated by a distance D . Most experiments agree with such a law [2,3,11]. However, some experimental studies [12,13] have shown the possibility of attractive interactions, in full contradiction with MP predictions, but other interactions could not be excluded, for example, electronic or electrostatic interactions [14,15]. Very recently, it has been shown that the elastic interaction could be obtained separately from the other contributions through x-ray measurements of the atomic relaxations [16]. As a result of this study the MP model was inadequate for a correct evaluation of the elastic displacements and the E_2 coefficient predicted was only approximate [16].

The MP model of the step interactions in the frame of elastic theory of continuous media is based on three main approximations: (i) the stepped surface is replaced by a straight one with periodic lines of force dipoles; (ii) the two components of the force dipole are taken on the surface, i.e., the lever arm of the dipole is parallel to the surface, the torque or stretch nature of the dipole being due to the force angle ϕ with respect to the lever arm; (iii) the elastic medium is taken as isotropic. Up to now, the attempts to calculate step interactions for anisotropic crystals have been limited to a few geometric configurations, for example, $\langle 100 \rangle$ line dipoles on the (001) surface of a cubic crystal [17]. Moreover, it has been shown [18] that it was also necessary to take into account

the real geometry of the step, which could not be reduced to a point dipole on a flat surface.

In this paper, we present anisotropic linear elasticity calculations for regular vicinal surfaces. Going in the reciprocal space we show that the elastic displacements and associated interactions can be obtained, taking into account both crystalline anisotropy and local step geometry. Only the first MP approximation is needed. The comparison with numerical simulations demonstrates its validity. The elastic displacements are highly anisotropic and the directions of both the force and lever arm play a major role for the interaction. These two conclusions are missed by the other MP approximations.

We restrict our study to steps whose direction y is perpendicular to a plane of symmetry. They are modeled by a periodic distribution of extended force dipoles. The dipoles consist of a couple of opposite forces applied on the step edge (S) and step corner (C). The calculations are done on a plane surface so that S is located at the surface ($z = 0$) and C is in the bulk ($z = z_0 > 0$) (see Fig. 1). Let us consider first a harmonic surface force distribution in the bulk $\vec{F}(x, z)$ given by

$$\vec{F}(x, z) = \vec{f} \exp(iqx) \delta(z - z_0), \quad \text{with } \vec{f} = (f_x, 0, f_z). \quad (1)$$

We can search for displacements $\vec{u}(x, z)$ given by

$$\vec{u}(x, z) = \sum_l \vec{a}_l^s \exp(ik_l q z) \exp(iqx), \quad (2)$$

where $s = “-”$ when $z < z_0$ and $s = “+”$ when $z \geq z_0$.

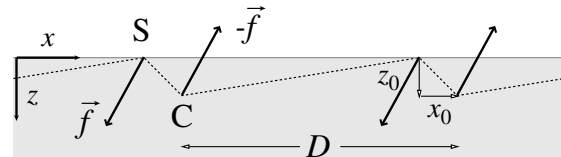


FIG. 1. Force dipole distribution equivalent to the steps. The full line indicates the surface used for elastic calculations whereas the dotted line indicates the geometry of the real surface.

Writing the bulk mechanical equilibrium leads to a fourth degree polynomial equation for k , of which the coefficients are polynomials of the elastic constants. Therefore, four values of k , conjugated two by two, satisfy the secular equation. Noting k_1 and k_2 , the solutions corresponding to $\text{Im}(k_1) \geq \text{Im}(k_2) > 0$ and $k_3 = k_1^*$, $k_4 = k_2^*$, we have $\vec{a}_3^+ = \vec{a}_4^+ = \vec{0}$. It is worth noting that for a general orientation of the surface, the displacements are attenuated in depth due to the imaginary part of k_1 and k_2 , but undergo also a linear drift due to their real part. Using the equality between stress discontinuities and external forces at $z = 0$ and $z = z_0$ and the continuity of the displacements at $z = z_0$, all the components $a_{l\alpha}^s$ of the \vec{a}_l^s can be found:

$$a_{l\alpha}^s = \sum_{\beta=x,z} \frac{f_\beta}{q} \left(\sum_{m=1}^4 b_{lm\alpha\beta}^s \exp(-ik_m q z_0) \right), \quad (3)$$

where the $b_{lm\alpha\beta}^s$ are rational fractions of the elastic constants and of k_l but do not depend on q . Details of the calculations will be given elsewhere[19]. This result also applies to a force distribution at the surface. We now consider the force dipole distribution shown in Fig. 1. To avoid infinite displacements and energy, the dipole distribution $\vec{F}(x, z)$ must be smoothed by a cutoff of length a_c . The choice of a Lorentzian broadening allows the harmonics resummation [20]. We take

$$\vec{F}(x, z) = \frac{\vec{f}}{D} \sum_{n=-\infty}^{\infty} \exp(inGx - |n|Ga_c) \delta(z) - \exp[inG(x - x_0) - |n|Ga_c] \delta(z - z_0), \quad (4)$$

where $G = 2\pi/D$. The displacements due to the $n = 0$ dipole layer are easily obtained and the total displacements write

$$u_\alpha(x, z) = \sum_{\beta} \frac{f_\beta}{\pi} \left[\text{Re} \left(\sum_{l=1}^4 \sum_{m=1}^4 b_{lm\alpha\beta}^- \log\{1 - \exp[G(ik_l z - ik_m z_0 + ix - ix_0 - a_c)]\} \right) - \sum_{l=1}^2 \sum_{m=1}^4 b_{lm\alpha\beta}^+ \log\{1 - \exp[G(ik_l z + ix - a_c)]\} + c_{\alpha\beta}(z_0 - z) \right], \quad \text{for } z < z_0,$$

$$u_\alpha(x, z) = \sum_{\beta} \frac{f_\beta}{\pi} \text{Re} \left(\sum_{l=1}^2 \sum_{m=1}^4 b_{lm\alpha\beta}^+ \log \frac{1 - \exp[G(ik_l z - ik_m z_0 + ix - ix_0 - a_c)]}{1 - \exp[G(ik_l z + ix - a_c)]} \right), \quad \text{for } z_0 \leq z, \quad (5)$$

where $c_{\alpha\beta}$ are rational fractions of the elastic constants. The elastic energy writes $E = -\sum_{\alpha,\beta} f_\alpha f_\beta H_{\alpha\beta}$ with

$$H_{\alpha\beta} = \frac{G}{4\pi} c_{\alpha\beta} z_0 + \frac{1}{2\pi} \text{Re} \sum_{l=1}^4 \sum_{m=1}^4 \left(b_{lm\alpha\beta}^+ \log \frac{1 - \exp(-2Ga_c)}{1 - \exp[G(ik_l z_0 + ix_0 - 2a_c)]} + b_{lm\alpha\beta}^- \log \frac{1 - \exp[G(ik_l z_0 - ik_m z_0 - 2a_c)]}{1 - \exp[G(-ik_m z_0 - ix_0 - 2a_c)]} \right). \quad (6)$$

From parity arguments, it is easy to show that the terms of odd order greater than 2 are zero in the series expansion of $H_{\alpha\beta}(G)$. Moreover it can be shown [19] that the first order term is zero leading to

$$E = E_0 + E_{\text{int}}(D) = E_0 + \frac{E_2}{D^2} + o\left(\frac{1}{D^4}\right),$$

$$\text{with } E_0 = \frac{f_\alpha f_\beta}{2\pi} \text{Re} \left(b_{lm\alpha\beta}^- \log \frac{2a_c + iz_0(k_m - k_l)}{2a_c + i(x_0 + z_0 k_m)} + b_{lm\alpha\beta}^+ \log \frac{2a_c}{2a_c - i(x_0 + z_0 k_l)} \right),$$

$$\text{and } E_2 = \frac{\pi f_\alpha f_\beta}{12} \text{Re} \{ x_0^2 (b_{lm\alpha\beta}^- + b_{lm\alpha\beta}^+) + 2x_0 z_0 (b_{lm\alpha\beta}^- k_m + b_{lm\alpha\beta}^+ k_l) - 4ia_c z_0 k_l (b_{lm\alpha\beta}^- - b_{lm\alpha\beta}^+) + z_0^2 [b_{lm\alpha\beta}^- (2k_m - k_l) k_l + b_{lm\alpha\beta}^+ k_l^2] \} \quad (7)$$

in which the summation on l, m, α , and β is implicit. The $a_c z_0$ term in E_2 must be omitted since it is related to the particular choice of a Lorentzian broadening for the force distribution and has no physical meaning.

Figure 2 shows the comparison of E_2 obtained from Eq. (7) for A-type steps on a Cu(111) vicinal with wide terraces, taking into account the exact geometry of the step ("buried dipole") or using a dipole with lever arm parallel to the surface ($z_0 = 0$) ("surface dipole") with the same results obtained using isotropic elastic constants for Cu. Both for anisotropic and isotropic cases, the major result is the strong dependence of the elastic inter-

action energy with the force direction ϕ for a buried dipole: The maximum of repulsive interaction energy is obtained for a dipole close to a pure torque ($\phi = 90^\circ$), whereas for a stretch dipole ($\phi = 0^\circ$), E_2 practically vanishes. This general behavior is also observed for a (001) orientation. Even more, full explorations of face orientations show that in special cases, for example, on Cu(119) or Au(332), the interactions can become slightly attractive for stretch dipoles. However higher order terms in the series expansion of E have to be considered when E_2 becomes negligible. For a surface dipole, the angular

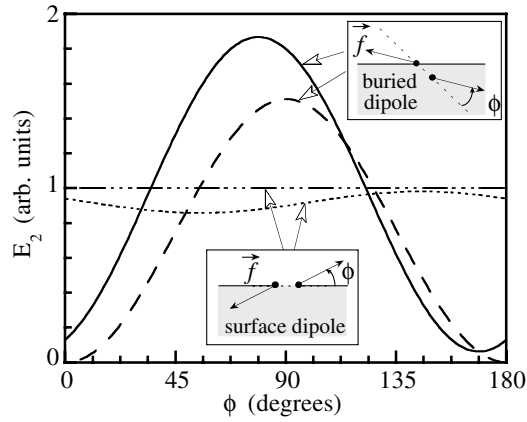


FIG. 2. Angular dependence of the interaction energy between steps on $\text{Cu}(n-1, n-1, n+1)$ with $n \rightarrow \infty$. Full and dashed lines: buried dipole ($z_0 = a_0/\sqrt{3}$, $x_0 = a_0/\sqrt{6}$). Dotted and dash-dotted lines: surface dipole ($z_0 = 0$, $x_0 = a_0/\sqrt{2}$). Full and dotted lines: anisotropic calculations. Dashed and dash-dotted lines: isotropic calculations. a_0 is the lattice constant and the values are normalized to the MP value of the interaction.

dependence of the interaction energy is quite weak and disappears for the isotropic case as predicted by Marchenko's formula. Therefore, the surface dipole model misses the strong dependence of the interaction energy with the force direction which is accounted for in the buried dipole model.

To confirm our results and to test the validity of our model, we have compared the analytical calculations with quenched molecular dynamics (MD) simulations. We restrict our presentation to Cu and Pt vicinals of (001) and (111) surfaces. For MD, we use a semiempirical many-body potential well adapted for transition and noble metals [21]. The parameters of the potential, fitted on the elastic constants and cohesive energy, are given in [22,23]. The simulations are performed on slabs of thickness e in the z direction, with periodic boundary conditions along x and y . The specific elastic displacements \vec{u}^{MD} due to the steps are obtained by subtracting to the relaxations the mean contribution of the terraces using the same procedure as in [16]. \vec{u}^{MD} are compared to the elastic displacements \vec{u}^{Elast} given by Eq. (5), taking \vec{f} as a free parameter. In the fitting procedure, we take into account all atoms except step edge and corner atoms. We are much more sensitive to the torque component p_T of the dipole than to the stretch component p_S for which the displacements are smaller in the bulk.

Figure 3 shows the comparison for Cu(15 15 16). The agreement is remarkable, in particular, on the linear drift and attenuation coefficients, given, respectively, by $\text{Re}(k_m)$ and $\text{Im}(k_m)$. Far from the surface, \vec{u} is given by the first harmonic of the k_2 mode. The displacements are then about 3 times higher along z than along x . Such anisotropy cannot be reproduced either with isotropic calculations or with surface dipoles. Both buried dipoles and anisotropic crystallinity are thus necessary for a

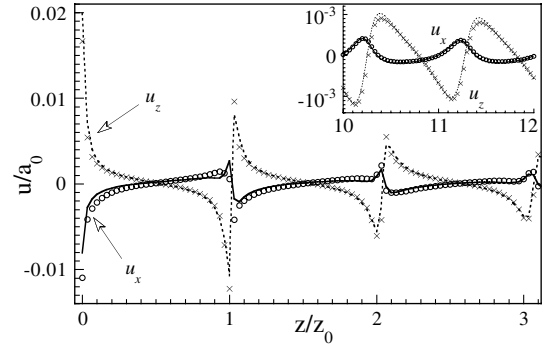


FIG. 3. Elastic displacements on Cu(15 15 16): comparison between MD and elastic results with $p_S = 0.22$ nN and $p_T = 0.19$ nN. Circles: u_x^{MD} ; crosses: u_z^{MD} ; full line: u_x^{Elast} ; dotted line: u_z^{Elast} . The inset shows the displacements deeper in the bulk. Note that the abscissas are obtained by following the atomic positions along the $[10\bar{1}]$ axis of the (111) plane.

correct description of \vec{u} . The inset of Fig. 3 corresponds to a depth at which the harmonics of intermediate order are still visible. The excellent fit of the MD results by the analytical expression of Eq. (5) in this region clearly indicates that a force dipole applied on the step edge and step corner fully accounts for the force distribution associated with the steps in the case of Cu(111) vicinals. This also justifies the choice of performing the elastic calculations on a planar surface. The minor differences appearing on Fig. 3 for $z \approx 0$ are probably due to the modification of the elastic constants near the surface in MD calculations, due to the many-body nature of the potential used. The agreement is also excellent for Cu(001) and Pt(001) vicinals, but less good for Pt(111) vicinals especially near the surface (see Fig. 4). This could be due to the fact that the force distribution is in that case more complex than a dipole.

In the MP model [10], p_T is equal to the product of the surface stress τ of the nominal surface by the step height z_0 . In the MD simulations, we have computed τ for the different surfaces studied. The comparison between τz_0 and p_T is given in Table I. p_T is very close to τz_0 . This indicates that the value of surface stress of a nominal

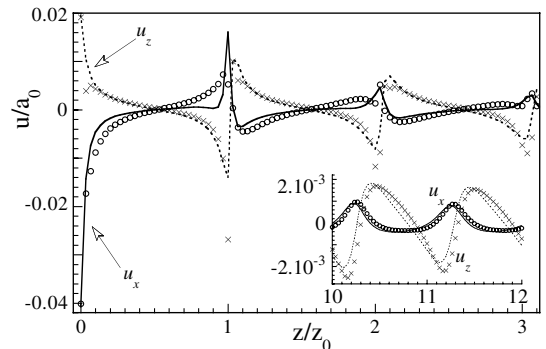


FIG. 4. Elastic displacements on Pt(15 15 16): comparison between MD and elastic results with $p_S = 0$ and $p_T = 0.61$ nN. Same legend as in Fig. 3.

TABLE I. Values, in the limit $D \rightarrow \infty$, of the torque dipole density p_T , of τ_{z_0} , and of E_2 , obtained from Eq. (7) or from Eq. (8). The precision is 10% on p_T and 20% on E_2 .

Surface	p_T (nN)	τ_{z_0} (nN)	E_2 (eV Å)	
			Elastic	MD
Cu(001)	0.25	0.237	0.056	0.076
Cu(111)	0.19	0.185	0.020	0.025
Pt(001)	0.39	0.407	0.17	0.17
Pt(111)	0.56	0.535	0.13	0.27

surface can be obtained through the determination of p_T on a vicinal surface.

The interaction energy $E_2^{\text{Elast.}}$ is obtained through Eq. (7), using the value of \vec{p} which gives the best fit to the displacements \vec{u}^{MD} . Since $E_2 \approx 0$ for a stretch dipole, it is mainly given by p_T (cf. Fig. 2). For all vicinals studied, the higher order terms in Eq. (7) were always negligible. However, it must be noticed that E_2 varies implicitly with D since crystalline orientation and p vary with D . Concurrently, the step interaction energy $E_{\text{int}}(D)$ can be obtained directly from the dependence of the surface energy γ given by MD results with the miscut θ :

$$\gamma(\theta) = \gamma(0) \cos(\theta) + \frac{1}{D} [E_0^{\text{MD}} + E_{\text{int}}^{\text{MD}}(D)]. \quad (8)$$

The comparison between the two methods is given in Fig. 5. The agreement is excellent for (001) vicinals. For Cu(111) vicinals, MD results exhibit a softening of the interaction for short interstep distances which is not recovered with Eq. (7). Nevertheless, for these short interstep distances, a linear elasticity approach is questionable. For Pt(111) vicinals, the agreement is less good, with $E_2^{\text{Elast.}}/D^2$ being approximately 40% less than $E_{\text{int}}^{\text{MD}}$. We have previously noticed that, in this case, a force distribution reduced to a dipole seems insufficient. $E_{\text{int}}^{\text{MD}}$ is well fitted by $E_2^{\text{MD}}/D^2 + E_3^{\text{MD}}/D^3$. The values of E_2^{MD} are compared in Table I to $E_2^{\text{Elast.}}(D \rightarrow \infty)$. The greater discrepancy between the two values is of course obtained for Pt(111). For Cu, E_2 is 5 to 10 times higher than the values found by statistical analysis of STM images [2,3]. This is partly due to the neglecting of the negative E_3 term in the STM studies. This could also be due to the differences between the interaction of straight steps at 0 K discussed here and the interaction of meandering steps above the roughening transition temperature, for which a 1D model is questionable.

To conclude, comparisons with MD calculations demonstrate the validity of the buried dipole model for vicinals of Cu(100), Pt(100), and Cu(111). Our calculation clearly shows that in order to reproduce the elastic interaction energy dependence with dipole direction, the environment difference between the step edge and the step corner must be considered. We show that analytical results

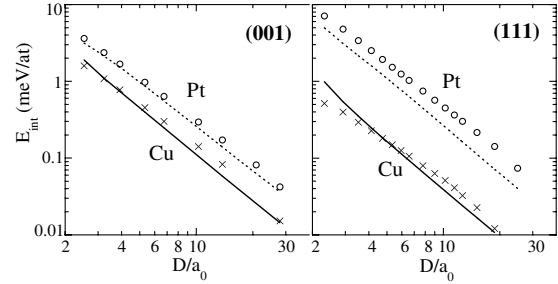


FIG. 5. Comparison of the step interaction energy derived from MD and E_2/D^2 obtained from Eq. (7). Symbols: MD results; lines: elastic results.

in the frame of anisotropic linear elasticity can be easily obtained by Fourier transform. The method could be applied to a wide variety of phenomena governed by the distribution of elastic forces near the surface such as self-assembly or self-ordering [20].

The authors thank L. Barbier and D. Calecki for fruitful discussions.

- [1] J. Lapujoulade, Surf. Sci. Rep. **20**, 195 (1994).
- [2] H.-C. Jeong and E. D. Williams, Surf. Sci. Rep. **34**, 171 (1999).
- [3] M. Giesen, Prog. Surf. Sci. **68**, 1 (2001).
- [4] J.-C. Boulliard and M. P. Sotro, Surf. Sci. **414**, 131 (1998).
- [5] V. Repain *et al.*, Phys. Rev. Lett. **84**, 5367 (2000).
- [6] M. C. Desjonquères, D. Spanjaard, C. Barreateau, and F. Raouafi, Phys. Rev. Lett. **88**, 056104 (2002).
- [7] P. Gambardella *et al.*, Phys. Rev. Lett. **87**, 056103 (2001).
- [8] V. Marsico, M. Blanc, K. Kuhnke, and K. Kern, Phys. Rev. Lett. **78**, 94 (1997).
- [9] Y. Garreau, A. Coati, A. Zobelli, and J. Creuze, Phys. Rev. Lett. **91**, 116101 (2003).
- [10] V. I. Marchenko and A. Ya. Parshin, Sov. Phys. JETP **52**, 129 (1980).
- [11] L. Barbier, L. Masson, J. Cousty, and B. Salanon, Surf. Sci. **345**, 197 (1996).
- [12] J. Frohn *et al.*, Phys. Rev. Lett. **67**, 3543 (1991).
- [13] W. W. Pai *et al.*, Surf. Sci. **307-309**, 747 (1994).
- [14] N. Garcia and P. A. Serena, Surf. Sci. **330**, L665 (1995).
- [15] A. C. Redfield and A. Zangwill, Phys. Rev. B **46**, 4289 (1992).
- [16] G. Prévot, P. Steadman, and S. Ferrer, Phys. Rev. B **67**, 245409 (2003).
- [17] L. E. Shilkrot and D. J. Srolovitz, Phys. Rev. B **53**, 11120 (1996).
- [18] R. V. Kukta, A. Peralta, and D. Kouris, Phys. Rev. Lett. **88**, 186102 (2002).
- [19] B. Croset and G. Prévot (to be published).
- [20] O. L. Alerhand, D. Vanderbilt, R. D. Meade, and J. D. Joannopoulos, Phys. Rev. Lett. **61**, 1973 (1988).
- [21] V. Rosato *et al.*, Philos. Mag. A **59**, 321 (1989).
- [22] G. Prévot *et al.*, Surf. Sci. **506**, 272 (2002).
- [23] F. Cleri and V. Rosato, Phys. Rev. B **48**, 22 (1993).