Elastic Interaction of Surface Steps: Effect of Atomic-Scale Roughness

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Elastic interactions of atomic steps can greatly impact surface morphology. Recent atomistic calculations and experimental observations find the standard dipole model of steps is valid only for very large step separations. In this Letter, a new model is presented that displays remarkable agreement with atomistic predictions for step separations larger than just a few step heights. It is shown that the interaction energy of steps exhibits a novel intermediate-ranged behavior and that, for particular systems, step interactions switch from repulsive to attractive as separation distance decreases.

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Film quality during epitaxial growth depends largely on the nature of surface evolution. As rough surfaces tend to develop defects (e.g., grain boundaries, dislocations, and voids), a flat surface is often desired throughout growth. On the other hand, certain applications require isolated material deposits in island or wire configurations. If properly controlled, roughening can be used to pattern such devices.

A surface evolves by step nucleation and propagation, as adatoms cluster into monolayer islands or attach to existing steps. The tendency is for steps to move towards low-energy configurations. To determine their interaction energy, Marchenko and Parshin [1] modeled steps as force dipoles on a half-plane. Each step dipole has a moment $h\tau$ and a dilatation $h\omega$ (h is step height, τ is surface stress, and ω characterizes step structure). The calculated interaction energy of two steps (labeled i and j) a distance d_{ij} apart is

$$\phi_{ij}^{\mathrm{MP}}(d_{ij}) = Ch^2(\omega^2 + n_i n_j \tau^2)d_{ij}^{-2}, \qquad (1)$$

where $C = (1 - \nu)/\pi\mu$ is a positive constant, ν is Poisson's ratio, μ is shear modulus, and $n_k = \pm 1$ denotes the orientation of a step k (whether it faces to the right or left). According to the model, interaction energy varies monotonically with separation, which means two steps will either repel or attract irrespective of their separation distance. This is a first order approximation valid for small values of h/d_{ij} [2].

The Marchenko-Parshin (MP) model has been used extensively over the past twenty years, and it is the basis for our understanding of step energetics. There are examples, however, where the model fails to explain experimental observation. Terrace width distributions measured on Cu [3] and Ag [4] indicate that step-step interactions may be either attractive or repulsive depending on their separations, and observed equilibrium shapes of Pb crystallites suggest a possible long-ranged attraction between like-oriented steps that opposes the repulsion determined by Eq. (1) [5]. Another example is the mesoscopic self-organization of steps on Pt [6], where periodic arrays consisting of several ascending steps followed by several descending steps form on a vicinal surface. Evidence suggests that these rippled morphologies represent minimum energy configurations, but the MP model suggests they are unstable [7–10]. The self-organization mechanism remains unclear.

Atomistic calculations are also found to exhibit features the MP model does not explain. Displacements about steps [2,11,12] and interaction energies between steps [12–16] agree only roughly with the model, and in certain cases substantial differences are observed. For example, the interaction energy of two opposite facing ⟨100⟩ steps on {001} tungsten was found to vary as $d_{ij}^{-1.6}$ for separations less than about 250 step heights [12], unlike the d_{ij}^{-2} behavior of Eq. (1). On the same surface, opposite facing ⟨110⟩ steps were found to attract, whereas the MP model suggests they repel. These effects cannot be explained by short-ranged corrections of quadrupoles and higher-order multipoles to Eq. (1).

Many of these observations can be understood with a departure from the half-plane simplification to account explicitly for the *stepped* geometry. Kukta and Bhattacharya [17] calculated the displacement field of an isolated step and found that the first-order correction is of a logarithmic order, weaker than a dipole field but stronger than a quadrupole field. A similar correction ought to appear in the interaction energy, establishing a distinct intermediate ranged behavior.

The model is fairly simple. The elastic field is that of a dipole applied on a *stepped surface,* as depicted in Fig. 1, rather than a flat surface. At points within a distance b from the center of the step, the surface shape (or step structure) is arbitrary, whereas the terraces ($|y| > b$) are flat. Length b is on the order of step height h and can be regarded as a structural parameter, for example, step "width." Quadrupole and higher-order source fields can be introduced to account for further structural details but are omitted because they decay much faster than a dipole field. The traction-free boundary condition is enforced on each terrace, and equilibrium within $r < b$ is enforced by balancing forces and moments about the step.

A series expansion of the elastic field, convergent for small values of h/r, can be obtained to any order accuracy. Up to second order, displacement on the terraces of an

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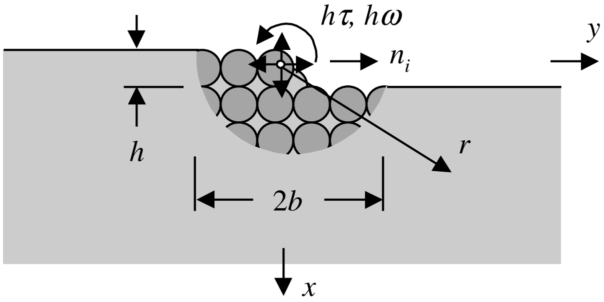

FIG. 1. Schematic of a surface step. The elastic field is determined by a dipole moment $h\tau$ and dipole dilatation $h\omega$.

isolated step are calculated as [17]

$$u_x = -Cn_i\tau\frac{h}{y} + Cn_i\omega\frac{h^2}{2y|y|} + \cdots, \tag{2}$$

and

$$u_y = C\omega\left\{\frac{h}{y} + \frac{2n_ih^2}{\pi y^2}\ln\frac{b}{|y|}\right\} - C\tau\frac{h^2}{2y|y|} + \cdots. \tag{3}$$

To leading order the result is equivalent to the MP model. Note that the displacements of a dipole and quadrupole on a half-plane decay as $1/r$ and $1/r^2$, respectively. The logarithmic term in Eq. (3) decays at a rate intermediate to these two source fields and is a first-order correction to the MP model. Comparing Eqs. (2) and (3) to atomistic results establishes values for the constants ω and τ, thus characterizing the leading order and first-order correction for a particular step [18]. The parameter b cannot be distinguished from a quadrapole source and is therefore associated with a correction of higher order.

To evaluate the interaction energy between steps, one needs the elastic field for the case of two steps. Unfortunately, the result cannot be obtained by superposing isolated step fields because adding a step alters the domain. Nevertheless, the calculation is tractable, and a closed-form solution has been obtained following the approach outlined by Kukta and Bhattacharya [17]. As the explicit equations for the stresses and displacements are too lengthy to include here, they will be presented in a different communication.

Eshelby [19] offers two methods for calculating the interaction energy between sources of stress: a direct and an indirect method. When the sources are decoupled, the direct method is possible. However, in the present system the elastic fields of the two steps are highly coupled and therefore the alternative indirect method is needed. Here the variation in the total energy with respect to step position is evaluated, and then the interaction energy is calculated by integrating the result. Consider two steps, one located at $y = y_i$ and the other at $y = y_j$. Step orientations are denoted by n_i and n_j, which have the value of $+1$ if the step faces the $+y$ direction, as in Fig. 1, or -1 if it faces the opposite direction. Holding the position of step j fixed, the energy release rate for the motion step i in the y direction is given by [19]

$$F_i(y_i) = -\frac{\partial E_{\text{tot}}}{\partial y_i} = \mathbf{e}_y \cdot \int_{C_i} \mathbf{Pq}\, dL, \tag{4}$$

where $\mathbf{e}_y$ is a unit vector in the y direction, $\mathbf{P} = W\mathbf{I} - (\nabla\mathbf{u})^T\boldsymbol{\sigma}$ is Eshelby's energy-momentum tensor, W is strain energy density, $\mathbf{I}$ is the identity tensor, ∇ is the gradient operator, $\mathbf{u}$ is displacement, $\boldsymbol{\sigma}$ is stress, and superscript T denotes the transpose of a tensor. The integration contour in Eq. (4) envelops step i, starting on its lower terrace and ending on its upper terrace, and its outward unit normal is $\mathbf{q}$. It is noted that the integral is path independent, which provides a useful check of the lengthy calculation. The total energy E_{tot} consists of the interaction energy $\Phi_{ij}(d_{ij})$, which depends on the relative position $d_{ij} = y_i - y_j$ of the steps, plus contributions that are independent of step position such as step and surface excess energies. Equation (4) sifts out all contributions but the interaction energy, and therefore one may calculate the interaction energy as

$$\Phi_{ij}(d_{ij}) = -\int F_i(y_i)\, dy_i. \tag{5}$$

To third order in h/d_{ij}, the result is

$$\phi_{ij}(d_{ij}) = \phi_{ij}^{\text{MP}}(d_{ij}) - \frac{C\omega^2 n_{ij}}{\pi}\frac{h^3}{d_{ij}^3}\left\{4\ln\frac{b}{|d_{ij}|} + 1\right\}, \tag{6}$$

where ϕ_{ij}^{MP} is the MP result given by Eq. (1) and $n_{ij} = n_i - n_j$.

The roughness correction, which is the last term in Eq. (6), vanishes for the case of like-oriented steps ($n_i = n_j$). Hence, the interaction energy is no different from the MP model. Atomistic calculations support this result, with energies exhibiting an almost exact $1/d_{ij}^2$ decay except for separations of a few step heights, where dipole-quadrupole interactions of the form $1/d_{ij}^3$ become strong [12]. The correction is nonzero for steps of opposite orientation, and, depending on the relative magnitudes of τ and ω, it may be significant for quite large step separations. In fact, when $\tau = \omega$, the contribution ϕ_{ij}^{MP} vanishes and the correction becomes the leading-order term.

Figures 2 and 3 compare the predictions of Eqs. (1) and (6) with interaction energies calculated with EAM (embedded atom method) for unlike-oriented steps on the $\{001\}$ surface of bcc tungsten. Step height is $h = 1.5826$ Å, which is half a unit cell, and $C = (1-\nu)/\pi\mu = 0.2286$ Å^3/eV. Surface stress is isotropic with a value $\tau = 0.188$ eV/Å^2, and, with this value, Eq. (2) demonstrates excellent agreement with EAM displacements. Comparing Eq. (3) with EAM displacements determines values of $\omega = 0.319$ eV/Å^2 for a $\langle 100\rangle$ oriented step and $\omega = 0.241$ eV/Å^2 for a $\langle 110\rangle$ step. The values differ because the two orientations have different atomic structures.

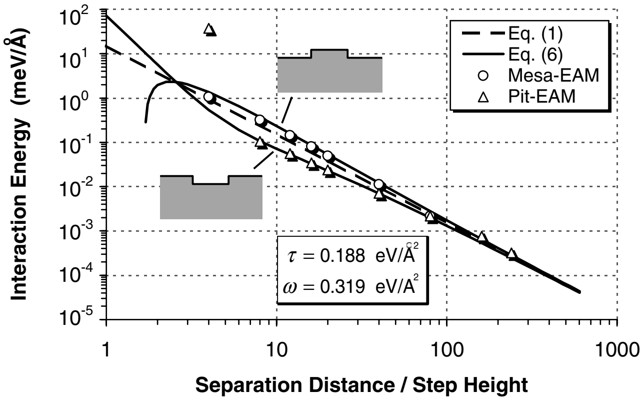

FIG. 2. Interaction energy versus separation distance for unlike-oriented $\langle 100\rangle$ steps on $\{001\}$ W. The value $b = 2h$ is used for Eq. (6).

The case of $\langle 100\rangle$ steps in both mesa (monolayer island) and pit (surface depression) configurations is plotted in Fig. 2. The MP model suggests that the interaction is equivalent for these two configurations; however, both Eq. (6) and EAM show different trends for separations less than about 200 step heights. The interaction energy for a mesa is about 20% greater than a pit for a step separation of $200h$ and about 225% greater for a $10h$ separation. Over a range of separations, between about $10h$ and $100h$, the interaction energy for the pit appears to decay similar to $d_{ij}^{-1.6}$, as observed elsewhere [12]. Equation (6) agrees well with EAM results for separations larger than $4h$. Slight differences for the largest separations plotted are due to finite domain effects in the EAM calculation. Recall that the value of b is on the order of h and that its effect is short ranged and cannot be distinguished from that of a quadrupole source.

For $\langle 110\rangle$ steps, the roughness correction is more significant because the difference between ω and τ is smaller. For example, while Eq. (1) suggests mesas and pits have the same interaction, Eq. (6) determines the mesa's interaction energy to be about 60% larger than the pit's for a $200h$ separation distance (compared with 20% in the $\langle 100\rangle$ case). For smaller separations, the difference can be several orders of magnitude. Figure 3 is a log-log plot of the absolute value of interaction energy for a pit, comparing the two continuum models with EAM results. Negative and positive values are labeled in the figure. Equation (1) suggests the step-step interaction is repulsive for all separations, while both Eq. (6) and EAM determine transitions from repulsion, to attraction, and back to repulsion as the steps become closer. Equation (6) determines the attractive region to lie between the maximum energy at $|d_{ij}| \approx 20h$ and the minimum energy at $|d_{ij}| \approx 4h$. The separation distance $|d_{ij}| \approx 4h$ corresponds to a metastable state. While the model may not be accurate for such small separations, EAM also suggests a metastable state near this separation distance. The mesa interaction (not plotted) has the same features as that in Fig. 2; however, its difference from the MP model is larger. In the case where ω is less than, but nearly equal to, τ, the mesa configuration would exhibit transitions between repulsion and attraction, while the pit interaction would always be attractive.

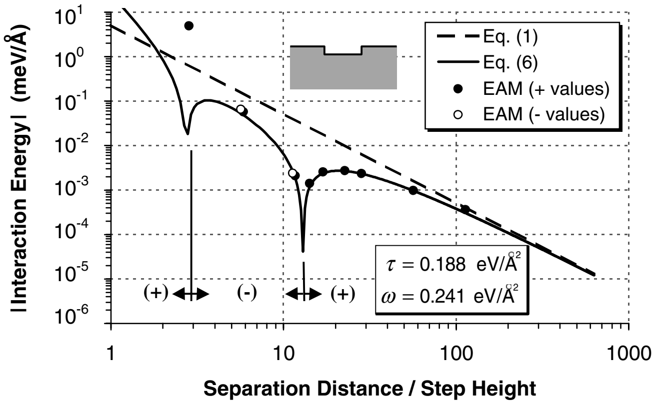

FIG. 3. Interaction energy versus separation distance for $\langle 110\rangle$ steps on $\{001\}$ W in a pit configuration. The value $b = 1.4h$ is used for Eq. (6).

Considering the pair interaction alone, one might conclude that roughness corrections are not important if all steps are oriented in the same direction, for example, on a vicinal surface. This is not the case, however, because one must account for threefold interactions between steps to capture the first-order effect of roughness. The energy of a multiply stepped surface can be written as

$$E_{\mathrm{tot}} = \sum_i \psi_i + \frac{1}{2}\sum_i \sum_{j\neq i} \phi_{ij}(d_{ij}) + \frac{1}{6}\sum_i \sum_{j\neq i} \sum_{k\neq i,j} F_{ijk}(d_{ij}, d_{jk}), \tag{7}$$

where ϕ_{ij} is given by Eq. (6) and F_{ijk} is threefold interaction between steps, which is of the same order as the last term in Eq. (6). The explicit form of F_{ijk} will be presented elsewhere, but a simple example is offered here to demonstrate its significance for like-oriented steps.

Consider the interaction between a step bunch, with N like-oriented steps, and a single step of the same orientation that are a distance d apart. Each step has the same height h, "width" parameter b, and constitutive constants τ and ω. Assuming the distance δ between bunched steps is small compared to d, the bunch can be approximated by a single step of height Nh, width parameter $(b/\delta)^{1/N}\delta$, and constitute constants τ and ω. This is deduced by building step bunches from the elastic field of two steps and explicitly taking their separation to be small compared to distance from the pair. The elastic field for the case of two different height steps was used to proceed beyond a two-step bunch. The interaction energy E_{int} between the bunch and the single step is calculated as

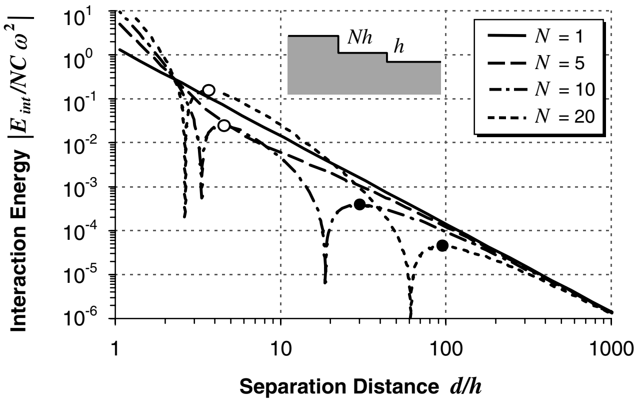

FIG. 4. Absolute value of interaction energy versus distance between an N-step bunch and a single step as determined by Eq. (8). Filled and open circles denote maxima and minima, respectively. Constants τ and ω are given in Fig. 2.

$$\frac{E_{\mathrm{int}}}{NC\omega^2} = \left\{\frac{\tau^2}{\omega^2} + 1\right\}\frac{h^2}{d^2} \pm \frac{(N-1)h^3}{3\pi d^3}\left\{12\ln\frac{b}{d} + 1\right\}, \tag{8}$$

where $\delta = b$ was assumed for simplicity. The last term is the roughness correction, where the plus sign is used when the step lies below the bunch, as in the inset of Fig. 4, and the minus is used if it lies above the bunch.

Figure 4 plots the case where the step lies below the bunch for $\langle 100 \rangle$ steps on $\{001\}$ W. The solid line corresponds to $N = 1$ and is also the prediction of the MP model for arbitrary N, which suggests the features repel for all separations. For $N = 5$, the step and the bunch also repel irrespective of separation, but for d less than about $100h$ a substantial change in the character of the interaction is apparent. For bunch sizes $N > 6$, the energy has a minimum and a maximum, which bound a range of separations where the interaction becomes attractive. For example, the respective minima and maxima occur at $d \approx 4h$ and $d \approx 29h$ for $N = 10$ and $d \approx 3.5h$ and $d \approx 92h$ for $N = 20$. The size of the attractive region is found to grow at a rate faster than N, which should have significance for coarsening of step bunches and facets. This has not yet been tested with atomistics. When the step lies above the bunch, Eq. (8) determines that the features attract (for $N > 1$) only when they are very close together (less than a few step heights apart). However, the model is not accurate for such separations.

It is found that roughness can have a significant effect on the elastic interactions of atomic steps, and in certain cases it determines a transition between repulsion and attraction with separation distance. In addition to altering the pair interaction between steps, the lowest order contribution of roughness to energy of crystal surface adds threefold step interactions that may have nonintuitive consequences. It is hoped that these will explain certain unresolved issues such as the mesoscopic self-organization of steps on Pt [6].

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