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Critical step undulation by elastic interaction

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Abstract

Theoretical explanation of the transition from a straight step to a wavy step that was observed on Si(0 0 1) is presented. The origin of this transition is the instability developed due to a negative logarithmic divergence of the wavy step elastic self-interaction. Above the critical temperature the straight step is stabilized by the interaction between different steps. The instability results in a second order phase transition with a finite critical wavelength. The predicted mean square fluctuation near the transition point deviates from the wavelength square law predicted by the edge-stiffness theory, and consists of the critical growth of the unstable soft mode, yielding a peaked power spectrum.

Keywords: Non-equilibrium thermodynamics and statistical mechanics; Surface structure, morphology, roughness, and topography; Vicinal single crystal surfaces

Undulation of steps on Si(0 0 1) [1–3] has often been cited as an example of two-dimensional pattern formation arising from surface stress anisotropy. For this surface, a bulk screw axis $C_4\tau$ in the space group generates two possible (0 0 1) surface structures (even without surface reconstruction). These structures, referred to as (1×2) and (2×1), are separated by monoatomic step. Two types of steps structures are possible, referred to as "A" and "B", depending upon the orientation of the structure on the upper terrace. In heavily boron-doped Si, unusual morphology was observed consisting of alternating wavy B-step and a straight A-step, the nearest B-step being out of phase [2,3] as demonstrated schematically in Fig. 1. Using low-energy electron microscopy (LEEM), Hannon et al. [3] investigated the transition from a straight to a wavy B-step that occurs at roughly 960°C. Specifically, they measured the fluctuations of the A- and B-type steps. The inspection of the images shown in Refs. [2,3] reveals most intriguing phenomena, namely, the transition occurs at a finite wavelength. From these micrographs, we estimated the critical wavelength to be roughly 0.5 μm.

The origin of the transition from straight to undulated step configuration has been a subject to an ongoing debate. Hannon et al. [3] attributed the undulation to the reduction of the single A-step free energy. According to this theory, the steps curve to include A-type-step segments. In an attempt to show this effect, they considered the temperature dependence of the step free energy by

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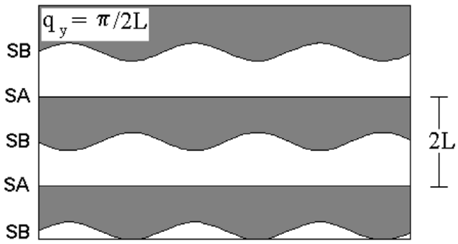

Fig. 1. Schematic presentation of the observed wavy step structure in doped Si(0 0 1). SA and SB represent the "A-step" and "B-step" correspondingly.

studying the step stiffness that was extracted from their fluctuation measurements. This was done by the application of the capillary theory by Bartelt and Tromp [4] that ignored elastic interaction between steps and predicted that the step mean square fluctuation decreases as the square of its wave number. This relation was approximately confirmed for the A-step, which is stable at the transition temperature against wavy fluctuations. In contrast to this theory, Pelz et al. [1,5] pointed out that the elastic interaction between steps (that explains the low-temperature ordering of the step pattern) reduces the total A-step energy. In reply, Hannon et al. [6] argued that the discrepancy is not evident at high temperature, before the pattern is fully developed.

Being the core of the debate and the essence of the present work, the steps elastic interaction on Si(0 0 1) calls for a particular presentation. This interaction belongs to the general problem of line defect interaction via negative logarithmic divergence. This divergence was predicted for the case of interacting boundaries between two dimensional phases on crystal and liquid surfaces as well as the interaction between domain boundaries in 2D ferromagnets [7–9]. Similarly, Ref. [10] considered the equilibrium structure of striped domain via logarithmic divergence, with isotropic edge energy. Experimental evidence for such interaction was found in the measurements of the ratio between the widths of straight terrace strips of (1×2) and (2×1) phases as a function of the external strain on Si(0 0 1) [11]. Similar phenomenon was found in the measurements of the variation of the width of oxygen monolayer strips with the adsorbed oxygen coverage on a copper surface [12]. Both experiments were interpreted in Refs. [13,14]. The thermodynamic of the striped domain case is different than the vicinal surface case since the domain width is not constrained by external force. The interaction between two wavy steps was studied in Ref. [15]. [1] The energy of a periodic array of wavy steps on Si(0 0 1) was examined numerically in Refs. [16,17]. In Ref. [17] Ebner et al., considered particularly the stability of step-train in-phase. (We shall show that this structure is not the first to become unstable.) None of these works attempted to study the transition from a straight step to a wavy step, as observed for the doped Si. The study of the influence of step–step interaction on the single step auto-correlation function involved various approximations and was recently reviewed in Ref. [18]. These models do not account for the detailed elastic interaction between all the steps or for the elastic self-interaction. A unique attempt to calculate the step shape spectrum in a step–train structure was concluded with the remark that the problem is relatively difficult [13].

In order to examine the role of the elastic interaction in the step fluctuation and in the undulation transition, the present letter analyzes both contributions of the step formation and the elastic interactions to the total system energy and to the step fluctuations. It is shown that the *self-interaction* of the wavy step (B-step) *destabilizes* a soft mode near the critical point, causing the undulation transition. The elastic interaction between the *unlike steps stabilizes* the straight step above the critical temperature. In contrast to the capillary theory, the present theory predicts that near the critical temperature the B-step fluctuations do not decrease as the inverse square of the wave number, and exhibit strong amplitude growth close to the critical point (see Fig. 4 below). We interpret the observed undulation instability as a second order phase transition with a finite wavelength at the transition temperature. The nature of this transition is similar to the one discussed for the flattening transition of a rough interface [19].

[1] Compare with 2D ferromagnet case in Refs. [7–9].

The energy of an ensemble of parallel surface steps is divided into two contributions:

(a) The energy of steps as the line defect: This energy is given by $\int \gamma_{A,B}(\phi)\, dl$, where $\gamma_{A,B}(\phi)$ is the energy density per unit length of A-step or B-step with orientation ϕ, and dl is the step length element; and,

(b) The steps elastic interaction, discussed below.

As the point symmetry of the surface is C_{2v}, the principle axes of the surface stress tensor [20], β^1 and β^2, are parallel to the reflection lines, e.g. along x and y. Therefore, the two surface structures are represented by the diagonal surface stress tensors, i.e.: $\beta^1_{xx} = \beta^2_{yy} = \beta_1$ and $\beta^1_{yy} = \beta^2_{xx} = \beta_2$. A given step running along x with a shape function $\eta(x)$ exerted a net force due to the surface stress, namely:

$$|f_x| = (\beta_1 - \beta_2)\frac{d\eta}{dx}, \quad |f_y| = (\beta_1 - \beta_2) \qquad \left(\left|\frac{d\eta}{dx}\right| \ll 1\right)$$

The force components are positive or negative depending upon whether the step is of type A or B. The interaction between step elements is

$$-\frac{1}{2}\int dx\, dx' \chi_{kl}(|x - x'|) f_k(x) f_l(x')$$

where χ is the elastic Green function of the surface. Consider an array of alternating infinitely long parallel A-step and B-step separated by terraces of width L. The shape of the nth steps (A or B) is represented by periodic functions $\eta_n(x)$. After substituting the force exerted on the step edge (as above), we express the elastic interaction to lowest order of $\eta_n(x)$, assuming a periodic step function (first order term vanishes):

$$(-1)^{n-n'}\Lambda \int dx_n \int dx_{n'} \left[\frac{1}{2}(\eta_n - \eta_{n'})^2 \left(r_{nn'}^{-3} - 3L^2 \times (n - n')^2 r_{nn'}^{-5}\right) - \eta'_n \eta'_{n'} r_{nn'}^{-1}\right]$$

where

$$r_{nn'}^2 = (x_n - x_{n'})^2 + L^2(n - n')^2$$

and

$$\Lambda = \frac{(1 - \sigma^2)(\beta_1 - \beta_2)^2}{4\pi E}.$$

Here E is the Young modulus and σ the Poisson ratio. The first two terms in the integrand are the expansion of the interaction due to displacements and forces normal to the step, and the third term arises from displacements parallel to the average step direction.

In the small amplitude limit we Fourier expand the step shape function, $\eta_n^{A,B}(x) = \sum_{\vec{q}} \eta_{\vec{q}}^{A,B} \times e^{iq_x x + 2nLqy}$. In this notations, the undulation mode represented in Fig. 1 is characterized by ($q_y = \pi/2L$, $\eta_q^A = 0$, $\eta_q^B = \eta_{q_x}$). In the harmonic approximation the total energy is given by:

$$E = \frac{1}{2}\sum_{\vec{q}}\{\lambda_q^{AA}|\eta_q^A|^2 + \lambda_q^{BB}|\eta_q^B|^2 + \lambda_q^{AB}\eta_q^A(\eta_q^B)^* + \lambda_q^{BA}\eta_q^B(\eta_q^A)^*\}. \tag{1}$$

Here the coefficients are calculated from the above:

$$\lambda^{AA,BB} = \frac{\Lambda}{2L^3}\left\{-(Lq_x)^2 \ln\left(\frac{1}{q_x L_{A,B}}\right) + 4\sum_{n=1}^{\infty}\left[\cos(2nLq_y)\frac{Lq_x}{2n}K_1(2Lq_x n) + \frac{1}{(2n-1)^2} - \frac{1}{(2n)^2}\right]\right\} \tag{2a}$$

$$\lambda^{AB} = \lambda^{BA} = -\frac{\Lambda}{2L^3}\sum_{n=1}^{\infty} 4\cos((2n-1)Lq_y) \times \frac{Lq_x}{2n-1}K_1(2Lq_x(2n-1)), \tag{2b}$$

and represented by elements of a matrix designated hereafter as the "[λ]-matrix". In Eqs. (2a) and (2b), $L_{A,B} = a_{A,B}\exp(\tilde{\gamma}_{A,B}/\Lambda)$, a_A and a_B are the elastic cutoff for the A-step and B-step, $\tilde{\gamma}_{A,B} = \gamma_{A,B} + (d^2\gamma_{A,B}/d\phi^2)$ is the step stiffness and K_1 is the modified Bessel function of order 1. The logarithmic term includes the contributions of the line defect energy and elastic self-interaction. This term becomes negative when $q_x L_{A,B} < 1$, revealing the origin of the undulation instability. However, the other interaction terms that arise from the elastic step–step interaction, can stabilize the straight step morphology.

In the general case ($qL \sim 1$), instabilities are identified by negative values of the eigenvalues of $[\lambda]$. These are given by:

$$\lambda_q^{+,-} = (\lambda_q^{AA} + \lambda_q^{BB})/2 \pm \left\{ \left(\lambda_q^{AA} - \lambda_q^{BB}\right)^2/4 + (\lambda_q^{AB})^2 \right\}^{1/2}$$

As $\lambda_q^+ > \lambda_q^-$, we search for instabilities in λ_q^-. When $q_y = \pi/2L$, the eigenvectors associated with the λ_q^- eigenvalues (for any q_x) are ($\eta_q^A = 0$, $\eta_q^B = \eta_{qx}$), which implies possible instability of the type shown in Fig. 1. In order to explore such instabilities, we write the energy as

$$E = \frac{1}{2}\sum_{q_x>0} \lambda^-(q_x)|\eta_{q_x}|^2,$$

where $\lambda^- = \lambda_q^{BB}$ and $q = (q_x, \pi/2L)$. In Fig. 2 we plot the numerical values of λ as function of q_xL and L/L_B. It is seen that instability occurs when $L \approx 4.1L_B$. The critical wave number is independent of L_A; and, from Fig. 2, $q_c \approx 0.63/L_B$. Moreover, the numerical investigation of λ_q^- in the q_x, q_y plane shows that there is a single minima (with the above critical values), indicating that the instability shown in Fig. 1 is developed before any other possible instability occurs.

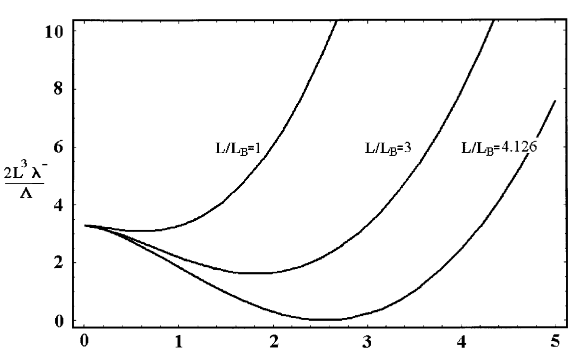

Fig. 2. The lower eigenvalue of λ vs. Lq_x. Curves of different values of L/L_B are presented, including the critical curve $L/L_B \approx 4.12$.

In view of Landau theory [21], the instability can result in a second order phase transition since the third order term of the energy expansion in amplitude vanishes by translational invariance. The critical temperature T_c, is determined by the dependence of L_B on the temperature. The amplitude $|\eta_{q_c}|$ grows near the critical temperature as $|T - T_c|^{1/2}$.

Having found the parameters leading to the instability, we are now able to evaluate the mean square fluctuation near the critical point. The amplitude of the step fluctuation is given by the elements of the inverse matrix $[\lambda^{-1}]$ (see e.g., Ref. [22]), in particular:

$$\langle|\eta^{A,B}(q_x)|^2\rangle = \frac{2T}{\ell}\int_0^{\pi/2L} \frac{dq_y}{2\pi}\frac{\lambda_{\vec{q}}^{BB,AA}}{\lambda_{\vec{q}}^{AA}\lambda_{\vec{q}}^{BB} - (\lambda_{\vec{q}}^{AB})^2}, \quad (3)$$

where T is the temperature, and ℓ is the step length.

When L_A and L_B are comparable to the terrace width L, the fluctuations in the limits of small wavelength are:

$$\langle|\eta^2|\rangle \propto \frac{1}{q_x^2 \ln q_xL} \quad (q_xL \gg 1);$$

In the limit of large wavelength, (3) yields:

$$\langle|\eta^2|\rangle \propto \frac{1}{q_x} \quad (q_xL \ll 1);$$

and finally, near the transition point, the mean square fluctuations of the B-step diverge at the critical wavelength:

$$\langle|\eta^2|\rangle \propto \frac{1}{\sqrt{A|T-T_c|^2 + (q_x - q_c)^2}} \quad (q_x - q_c)L \ll 1$$

where A is a constant.

We use Eq. (3) to calculate the fluctuation of the A-step and compare it with the results of Ref. [3]. For this purpose, we evaluate the integral in Eq. (3) numerically for a general wave number q_x, and at temperature 966°C. The integral depends on L/L_A and by fitting to the data of Ref. [3], we found: $L/L_A \approx 0.1$. Fig. 3 shows the so obtained

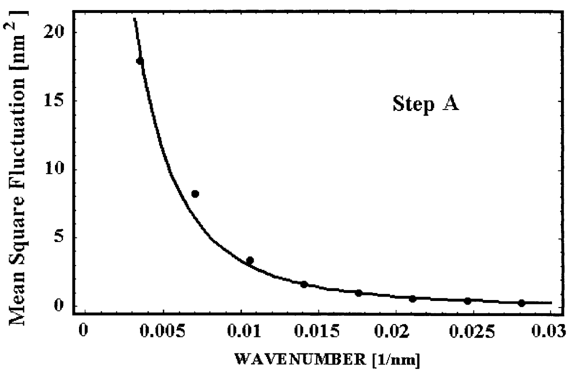

Fig. 3. Fitting the present theory (—) to the experimental (●) power spectrum of the A-step shape function near the undulation transition of the B-step. Experimental points are measured at 966°C, and are presented in Ref. [3]. The elastic contribution to the A-step spectrum is too small to be distinguishable from the edge-stiffness theory (Fig. 4 in Ref. [3]).

spectrum of A-step. The step stiffness is extracted by assuming $L = 180$ nm, $a_{\mathrm{B}} = 0.3$ nm, $|\beta_1 - \beta_2| \approx 16$ eV/nm^2, $E = 670$ eV/nm^3 and $\sigma = 0.3$, i.e. $\tilde{\gamma}_{\mathrm{A}} = \Lambda \log(L_{\mathrm{A}}/a_{\mathrm{A}}) = 0.24$ eV/nm. This value is comparable to the one suggested in Ref. [3]. Indeed, Fig. 3 is similar (though different presentation) to Fig. 4 in Ref. [3], and the elastic interaction does not contribute significantly to the A-step fluctuations near the undulation point of the B-step (see the limit $q_x L \gg 1$ below Eq. (3)).

The B-step fluctuations, depend on the coherence length L_{B}. Therefore, before applying Eq. (3) to the B-step fluctuations, lets discuss this length and its physical significance. For this purpose, we presume that the critical wavelength is 500 nm (this value may be estimated for a particular experiment by examining the LEEM images near the critical temperature). Employing the relation between the critical wave number and the coherence length ($q_{\mathrm{c}} \approx 0.63/L_{\mathrm{B}}$), we suggest: $L_{\mathrm{B}} \approx 50$ nm. Accordingly, the B-step stiffness is found: $\tilde{\gamma}_{\mathrm{B}} = \Lambda \log(L_{\mathrm{B}}/a_{\mathrm{B}}) = 0.15$ eV/nm. This value is larger by a factor 15 than the one suggested in Ref. [3]. The difference originates at the contribution of the elastic interaction that was ignored in the previous theory. In this context, Ref. [5] pointed out that the possible error in the estimation of the step stiffness in Ref. [3] is due to neglecting the negative elastic interaction between unlike steps. The present theory shows that the interaction between the unlike steps introduces a positive contribution (stabilizing the straight step above the critical point). Hence, the stiffness value extracted in Ref. [3] is different from the present value as a result of ignoring the negative self-step interaction. (At the critical point this interaction balances the contributions due to the capillary and the unlike-step interactions.) By the same argument, the consideration of step features with a radius of curvature is *smaller* than the coherent length (at the corners of the "triangular step facet" or at the apex of lens-shape islands [3,23]) should take into account the elastic interaction. To conclude the discussion of the coherence length, the fact that the transition occurs only in doped specimens implies that doping changes the coherence length. This can be due to the variation of the step stiffness and the surface stress upon segregation to the edge and to the surface terraces. In the absence of experimental data on the fluctuation of the B-step, we calculate in Fig. 4 $\langle|\eta^2_{q_x}|\rangle$ for the above coherence length ($L_{\mathrm{B}} \approx 50$ nm). In contrast to the step A fluctuations, the predicted spectrum of the B-step deviates significantly from the q_x^{-2} power law of the existing step-stiffness theory. The peak seen in this figure manifests the development of a soft mode near the undulation transition, and located close to the critical wave number. Therefore, experimental

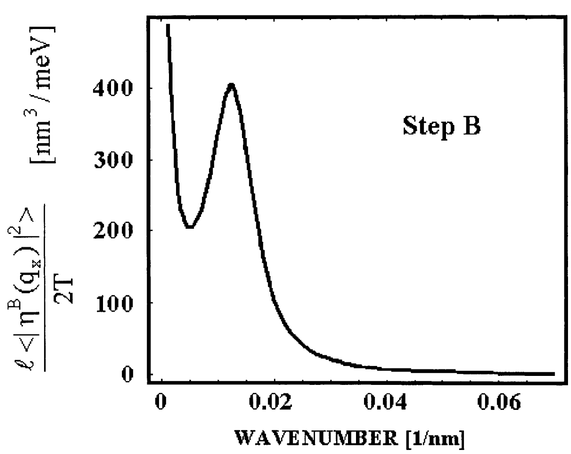

Fig. 4. Prediction of the B-step fluctuations near its undulation transition (966°C). The peaked spectrum manifests the growth of a soft mode. This spectrum deviates significantly from the q_x^{-2} power law of edge-stiffness theory.

measurements of the B-step fluctuations near the critical point may reveal (or not) the significance of the self-elastic interaction by searching for the typical spectrum as demonstrated in Fig. 4.

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