

## Pre-roughening induced deconstruction in Si and Ge(001) type crystal surfaces

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1997 J. Phys. A: Math. Gen. 30 397

(<http://iopscience.iop.org/0305-4470/30/2/007>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.110

The article was downloaded on 02/06/2010 at 06:02

Please note that [terms and conditions apply](#).

## Pre-roughening induced deconstruction in Si and Ge(001) type crystal surfaces

Marcel den Nijs

Department of Physics, University of Washington, Seattle, WA 98195, USA

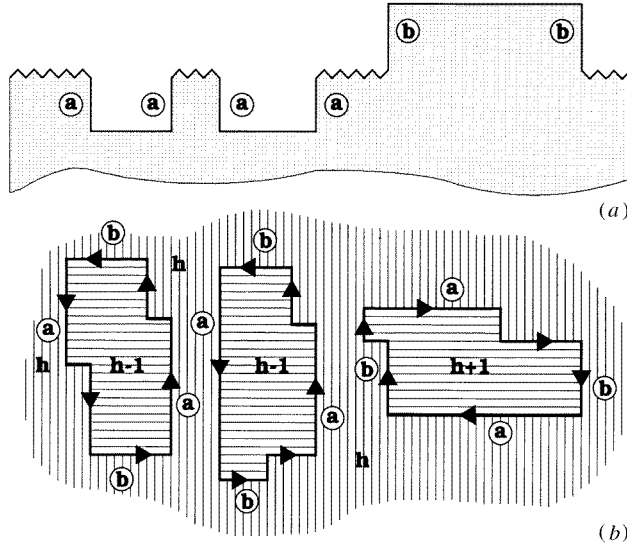
Received 29 July 1996

**Abstract.** The (001) facets of Si and Ge have a uniaxial structure which changes direction at each mono-atomic step. This type of topology is unusual and new for the theory of surface roughening and reconstruction phase transitions. It can be incorporated into the solid-on-solid model description. The phase diagram includes pre-roughening transitions and disordered flat phases without the need for step–step interactions. The competition between this and the  $1 \times 2$  reconstruction in Si(001) can be described by a generalized 4-state clock-step model. This leads to the prediction that Si(001) and Ge(001) undergo a pre-roughening induced simultaneous deconstruction transition.

The (001) facets of Si and Ge have been studied extensively [1–5]. These surfaces are reconstructed in a  $1 \times 2$  structure in which the dangling bonds on the surface form dimers. Most theoretical research has focused on low temperatures aspects (close to room temperature), such as elastic stress and the crossover from mono- to bi-atomic steps in vicinal surfaces. The latter results from the presence of two types of mono-atomic steps, so-called  $S_A$ - and  $S_B$ -type steps. They originate from the quite unusual topology of the Si(001) surface. The purpose of this paper is to investigate the consequences of this topology for the interplay between surface roughening and reconstruction in non-vicinal surfaces at high temperatures. A solid-on-solid (SOS) model that incorporates these topological properties will be introduced. Its phase diagram contains interesting phase transitions: pre-roughening (PR) without the need of step-step interactions, and PR induced simultaneous deconstruction.

SOS models with short-range interactions provide an adequate description of surface excitations in metals and semi-conductors close to the roughening transition. STM and LEEM studies of step fluctuations on Si(001) [3–5] and x-ray reflections from Ge(001) [6] suggest a roughening transition at about  $T \simeq 0.8T_m$ , with  $T_m$  the bulk melting temperature. At such high temperatures, details such as the rather long-range tails of elastic interactions, are typically unimportant. They do not affect the universality class of the transitions.

A detailed microscopic description of the  $1 \times 2$  reconstruction in Si(001) can be found in the literature [1]. It suffices to present the essential aspects in cartoon style. The two-fold degenerate  $1 \times 2$  reconstructed state is uniaxial. The crystalline structure is such that at alternating surface heights the uniaxial direction switches by  $90^\circ$ , see figure 1. At even surface heights the reconstruction runs vertically, whereas at odd heights it runs horizontally; this is the essential point. If presented out of context, this property would be dismissed as quite esoteric. However, nature is not as limited as our imagination.



**Figure 1.** Schematic representation of the  $S_A$ - and  $S_B$ -type mono-atomic steps: (a) side-view and (b) top-view of up and down terraces.

Consider a surface with a structure like Si(001), but which does not reconstruct. Such a surface remains uniaxial, and the uniaxial direction still switches by  $90^\circ$  at each mono-atomic step. This inequivalence between surface structures at alternating heights creates a distinction between two types of mono-atomic step excitations, so-called  $S_A$ - and  $S_B$ -type steps (figure 1 illustrates this). The uniaxiality runs parallel to the step in the upper- or in the lower-terrace. These steps have peculiar topological properties: if two neighbouring parallel steps are of the same type, e.g. two  $S_A$ -type steps, one must be an up-step and the other be a down-step; if a step turns over  $90^\circ$  it must change type, e.g. from  $S_A$ - to  $S_B$ -type. The following restricted solid-on-solid (RSOS) model on a square lattice incorporates the essence of this

$$\mathcal{H} = \sum_{\mathbf{r}} \{ K[h(x, y) - h(x + 1, y)]^2 + \Delta \sin[\frac{1}{2}\pi(h(x, y) + h(x + 1, y))] + K[h(x, y) - h(x, y + 1)]^2 - \Delta \sin[\frac{1}{2}\pi(h(x, y) + h(x, y + 1))] \} \quad (1)$$

with  $\mathbf{r} = (x, y)$ . At  $\Delta = 0$  the model reduces to the conventional RSOS model [7], and only has a Kosterlitz–Thouless (KT)-type roughening transition. The  $\Delta$  operator describes the switching of the uniaxial direction in Si(001). The operator switches sign at alternating heights accordingly, such that  $E_A = K - \Delta$  and  $E_B = K + \Delta$ . Bi-atomic steps are neglected in this model and nearest-neighbour columns can only differ in height by  $dh = 0, \pm 1$ . Step-step and corner interactions are omitted as well, since they are not essential. At greater length scales the local surface height can be represented by a continuous variable,  $\psi$ , and the RSOS model renormalizes into a generalized sine–Gordon model of the form

$$\mathcal{H} = \int d\mathbf{r} \left\{ \frac{1}{2} K_G [\nabla \psi]^2 - u_2 \cos(2\pi \psi) - u_4 \cos(4\pi \psi) + u_\Delta \sin(\pi \psi) \left[ \frac{d^2 \psi}{dx^2} - \frac{d^2 \psi}{dy^2} \right] \right\}. \quad (2)$$

This is the same model as the one used to describe conventional roughening and PR transitions [7–10], except for the novel  $u_\Delta$  operator. The rough phase is described by

the Gaussian fixed line at  $u_2 = u_4 = u_\Delta = 0$ . Inside the rough phase the fluctuations associated with  $\Delta$  are irrelevant. They have a critical dimension  $x > 2$  for all values of  $K_G$ , because the  $u_\Delta$  interaction has the form of a gradient. Each derivative shifts the critical dimension of an operator by one,  $x \rightarrow x + 1$ . The roughening transition is driven by fluctuations associated with  $u_2$ , and takes place at  $K_G = \frac{1}{2}\pi$  where  $\cos(2\pi\psi)$  becomes relevant. Consequently,  $\Delta$  does not affect the scaling properties of the rough phase nor those of the roughening transition. It only renormalizes  $K_G$ , i.e. the roughening temperature.

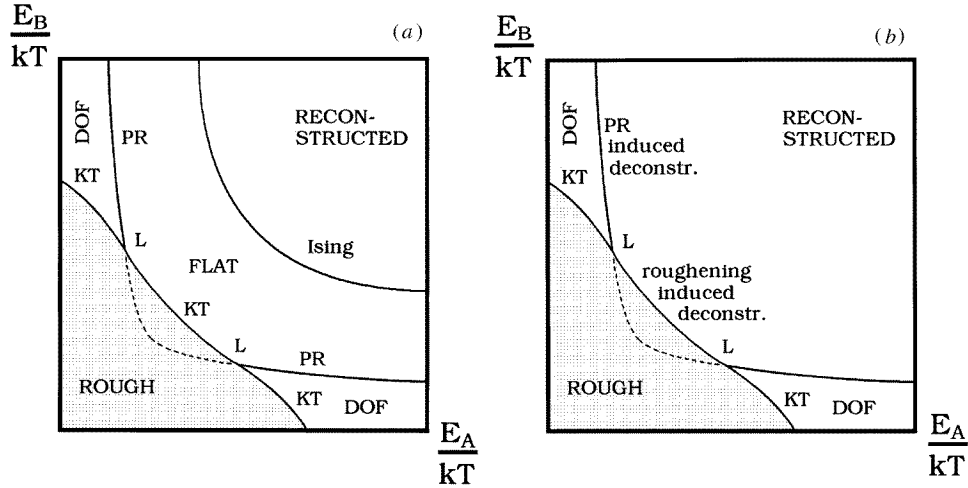
PR transitions and disordered flat (DOF) phases are associated with a change in sign of  $u_2$  [7–11]. The properties of the rough phase and the roughening transition remain the same, but the structure of the flat phase changes. At positive  $u_2$ , the local surface height prefers integer values commensurate with the microscopic height variables; this leads to the ordered flat phase. At negative  $u_2$ , the local surface height prefers half-integer values incommensurate with the microscopic height variables (integers). This leads to the DOF phase: a liquid of randomly placed steps, with long range alternating step-up step-down order, such that the surface remains flat at great length scales with on average a half-integer surface height. The PR transition line is the locus of  $u_2 = 0$  outside the rough phase.

The conventional mechanism for PR is by means of step–step interactions [7]. They can induce  $u_2$  to change sign. The central point of this letter is the observation that the  $\Delta$  interaction does this too. The peculiar topology of Si(001) type surfaces creates a DOF phase without the need for step–step interactions.

Before demonstrating the existence of the DOF phase in the RSOS model, I will present an intuitive argument. It is a generalization of the conventional argument that illustrates how step–step interactions give rise to an entropic preference for the DOF structure [7]. At low temperatures the ordered flat phase contains defects in the form of small ellipsoid shaped terraces. Up-terraces are oriented one way and down-terraces the other way. The characteristic size of these terraces increases with temperature, because meander entropy (kink excitations) decrease the step free energy. (Pre-) roughening comes into play when the terraces become large and sufficiently plentiful that they begin to hinder each other. Consider two neighbouring terraces. Their axes are orthogonal if one is an up-terrace and the other a down-terrace. Their axes are aligned if both are down-terraces or both are up-terraces. A structure with aligned ellipsoids has less excluded area and therefore more positional entropy than one with perpendicular ellipsoids. We might expect therefore a phase-separation into macroscopic domains of parallel oriented terraces in a (narrow) temperature interval close to the onset of roughening. These domains are droplets of DOF phase. The average surface height inside each domain is a half-integer.

The study of a specific model, the RSOS model of equation (1), will tell us whether this entropic preference is strong enough to actually stabilize the DOF structure. Figure 2(a) shows the structure of its phase diagram, as expected from the intuitive arguments presented above and the more detailed analysis below. Ignore the low temperature Ising type reconstruction transition line in figure 2(a), because equation (1) does not incorporate surface reconstruction. The DOF phase exists in the  $E_A \simeq 0$  corner of the phase diagram. At  $E_A = 0$  and zero temperature, steps of type  $S_A$  cost no energy while steps of type  $S_B$  are frozen out. A typical configuration consists of a set of randomly placed  $S_A$ -steps. These are straight lines, because kinks are segments of  $S_B$ -steps. Each vertical line in the lattice (or each horizontal one) contains with equal probability, an  $S_A$ -step or no step. The topological rules require that these are alternating up- and down-steps. This is a zero temperature DOF structure.

This DOF phase is stable against fluctuations. The low temperature part of the phase diagram close to the line  $E_A = 0$  can be described by a fermion expansion, similar to



**Figure 2.** Phase diagram of the RSOS model, equation (1): (a) the surface reconstruction vanishes prior to surface (pre-) roughening when wall excitations are cheaper than steps; (b) (pre-)roughening induces a simultaneous deconstruction transition when steps are cheaper.

the one for striped incommensurate phases in adsorbed monolayers [12,9]. The vertical  $S_A$ -type steps represent the world lines of the fermions. At low temperatures they can hop (meander excitations) and be created and annihilated in pairs (dislocations). Each of these events requires only one single  $S_B$ -step type segment. The conventional fermion analysis yields that the dislocation operator is relevant [12]. Therefore the low temperature phase is not a floating phase with an infinite correlation length but a DOF fluid phase. Roughening does not take place until much higher temperatures. This requires the nucleation of terrace excitations in the DOF where the surface height changes from  $h + \frac{1}{2}$  to e.g.  $h - \frac{1}{2}$ . The edges of such terraces require a number of  $S_B$ -type segments proportional to the size of the terrace. The step energy is approximately equal to  $\frac{1}{2}E_B$  per unit length (with respect to the DOF phase free energy as described by the fermion model).

One of the conventional methods to estimate the location of critical lines is to use the formula for the critical temperature in the Ising model,

$$\sinh(E_x/k_B T_c) \sinh(E_y/k_B T_c) = 1 \quad (3)$$

with approximate values of the step and wall energies,  $E_x$  and  $E_y$ , in the two directions [10]. Equation (3) with  $E_x = E_y = \frac{1}{2}(E_A + E_B)$  gives an estimate for the roughening transition line in figure 2(a) at general  $E_A$ , because roughness is created by perpendicular terrace excitations and the excluded area associated with them is approximately isotropic. Equation (3) with  $E_x = E_A$  and  $E_y = E_B$  gives an estimate for the PR line, because the DOF structure arises from the ordered flat phase by means of aligned ellipsoid shaped terrace excitations.

The DOF phase disappears at point  $L$  where the roughening and PR lines merge. The precise location of  $L$ , the value of  $\Delta_L$ , is difficult to estimate, but actually not essential, since this is a non-universal aspect that varies with details such as the strength of step-step interactions. It is important that we can exclude the two extreme cases,  $\Delta_L \neq \infty$  and  $\Delta_L \neq 0$ . The above stability analysis of the DOF structure along  $E_A = 0$  demonstrates that  $\Delta_L \neq \infty$ , while the following results along  $E_A = E_B$  imply that  $\Delta_L \neq 0$ . The PR line is the locus of  $u_2 = 0$  in equation (2) outside the rough phase. The broken curve in

figure 2(a) is its continuation inside the rough phase (where it does not represent a phase transition). It intersects with  $E_A = E_B$  at exactly  $E_A/k_B T_D = \log[\frac{1}{2}(\sqrt{5} + 1)] = 0.4812$  [13]. The roughening temperature is known accurately along  $E_A = E_B$  as well,  $E_A/k_B T_R = 0.633 \pm 0.003$  [13]. The DOF phase requires a non-zero threshold value  $\Delta_L \neq 0$ , because  $T_D > T_R$ .

The scaling properties of the PR transition in figure 2(a) are identical to those of conventional PR transitions (those where the DOF phase is stabilized by step–step interactions) [7–11]. Their scaling theories, equation (2), are identical, since the  $\Delta$  operator is irrelevant. Only the mechanism that causes  $u_2$  to change sign is different. The PR transition has continuously varying critical exponents that depend on where the experimental system crosses the PR line. The transition is infinite-order at point  $L$  and becomes stronger at larger  $|\Delta|$ .

At the PR transition the surface height changes spontaneously by one half unit. A facet with macroscopic surface height  $h$  phase separates into DOF regions with  $h + \frac{1}{2}$  and  $h - \frac{1}{2}$ . This requires macroscopic particle transport along the surface. Experimental systems follow particular paths through the phase diagram; approximately straight lines at specific  $E_A/E_B$  ratios. In surfaces where PR takes place close to roughening, the surface mobility will be large enough to achieve the DOF structure, because roughening requires particle transport along the surface as well. In surfaces with large  $E_A/E_B$ , PR might take place at temperatures where the surface mobility is too small to reach equilibrium.

Experimental signatures to distinguish between ordered flat, DOF, and rough phases, have been reviewed in detail elsewhere [7–11]. Here it suffices to point-out how uniaxiality behaves in each phase. There are two sources for uniaxiality: the crystalline structure at the microscopic level; and the ellipsoid shape of the terraces at intermediate length scales. The crystalline structure is responsible for macroscopic uniaxiality in the ordered flat phase, since there exists one macroscopic terrace, and its microscopic crystalline structure persists at large length scales (limited only by quenched surface defects). This source for uniaxiality vanishes at the PR transition, because in the DOF phase terraces of height  $h$  and  $h + 1$  form an equal mixture and they have alternating crystalline uniaxiality. However, the terraces have an ellipsoid shape and collectively they are oriented in the  $x$  or  $y$  direction. Therefore both phases are macroscopically uniaxial, but the length scale at which this is established is different and provides an additional experimental signature for the PR transition. Uniaxiality vanishes completely at macroscopic length scales above the roughening transition.

Si(001) and Ge(001) facets are  $1 \times 2$  reconstructed due to the pairing of dangling bonds. The low-lying excitations are terraces where the reconstruction order switches and/or the surface height changes by one unit. The edges of these are line defects, walls and steps [7–11]. At walls, the surface height does not change, only the reconstruction order switches. At steps, the surface height changes and the reconstruction order can switch as well. Deconstruction is a separate phenomenon taking place at lower temperatures if walls are cheaper than steps, as in figure 2(a). Deconstruction competes with (pre-)roughening if steps are cheaper than walls. The latter is the more interesting case. Two possibilities exist: (pre-)roughening simultaneously destroys the reconstruction, or the reconstruction persists inside a so-called reconstructed rough phase. The topological rules about how the step excitations couple to the reconstruction order determine this [9, 10]. In unreconstructed surfaces steps carry only one topological charge:  $dh = \pm 1$ , to denote the change in surface height between the two domains at either side of the step. In reconstructed surfaces they carry an additional topological charge  $(dh, d\theta)$ , to denote a switch between degenerate reconstructed structures across the step. For a review see [10]. Surprisingly, the topological charges of steps in Si(001) type surfaces are identical to those in missing-row (MR) reconstructed

FCC(110) facets like Pt(110) [9–11]. A 4-state clock variable,  $\theta = 0, \pm\frac{1}{2}\pi, \pi$  denotes the two  $1 \times 2$  states ( $\theta = 0, \pi$ ) and two  $2 \times 1$  states ( $\theta = \pm\frac{1}{2}\pi$ ) at alternating heights. Anti-phase domain walls have topological charge  $(0, \pi)$ . At each mono-atomic step  $\theta$  rotates over  $\pm\frac{1}{2}\pi$ ; their topological charges are  $(dh, d\theta) = (\pm 1, \pm\frac{1}{2}\pi)$ . The competition between surface (pre-) roughening and reconstruction in Si(001) can therefore be modelled by the same 4-state clock-step model introduced before for Pt(110) and Au(110) [9], with two important differences: we can ignore chirality, which represents the energy difference between clockwise ( $d\theta = \frac{1}{2}\pi$ ) and anti-clockwise ( $d\theta = -\frac{1}{2}\pi$ ) steps in Pt(110) because those have the same energy in Si(001) by symmetry; we need to introduce the  $\Delta$ -type interaction to describe the energy difference between  $S_A$  and  $S_B$  type steps in Si(001).

Figure 2(b) shows the structure of the phase diagram for surfaces where step excitations are cheaper than anti-phase walls. The essential feature is the absence of a reconstructed rough phase. Steps create surface roughness and destroy the reconstruction order simultaneously. Topology requires this, but this is not obvious. For example, MR reconstructed simple-cubic (110) facets lead to reconstructed rough phases, but MR reconstructed FCC(110) facets do not. In the SC case it is always possible to redefine the reconstruction order parameter in such a way that the steps responsible for the roughening do not couple to the reconstruction. In the FCC case this is impossible. For a detailed discussion see [9, 10]. Reconstructed rough phases are impossible in Si(001) facets because the steps in the  $1 \times 2$  reconstruction have the same topological properties as in MR reconstructed FCC(110) facets.

In figure 2(b) roughening induces a simultaneous deconstruction transition at small values of  $\Delta$ . Numerical results at  $\Delta = 0$  [9] show that this transition has a central charge  $c = 1.5$ , with both Ising-type critical exponents and the conventional universal surface roughness value  $K_G = \frac{1}{2}\pi$ . It is unlikely that  $\Delta$  modifies these scaling properties, but this needs to be tested in a future numerical study of this model. At large values of  $\Delta$  PR induces a simultaneous deconstruction transition. This is something new. The reconstruction order vanishes in the DOF phase for the same reason as in the rough phase. The proliferation of steps destroys the reconstruction order, irrespective of the presence (DOF phase) or absence (rough phase) of long range step-up step-down order. The scaling properties of this PR induced simultaneous deconstruction transition are as yet unknown. Maybe its scaling properties are a simple superposition of a KT roughening and PR transition (with a central charge  $c = 2$ ).

A deconstruction transition has been reported in an x-ray study of Ge(001) [6]. The reconstruction order parameter was determined from the  $(0, 1.5)$  reflection and vanishes with a critical exponent  $\beta$  close to the Ising value  $\beta = \frac{1}{8}$  at  $T = 955 \pm 7$  K. The authors report the onset of surface roughness at about the same temperature,  $T = 900 \pm 100$  K, from a drop in specular intensity. These results are not detailed enough to determine whether the path Ge(001) follows through figure 2. It might be a realization of the roughening induced simultaneous deconstruction transition at small  $\Delta$ . It might be like Au(110) [10, 11], in which the reconstruction is believed to vanish just before roughening (at  $(T_R - T_D)/T_R \simeq 0.05$ ). In both cases the reconstruction order parameter should vanish with the Ising exponent  $\beta = \frac{1}{8}$ . A PR induced deconstruction transition is also still possible, because the integrated intensity of the specular peak does not characterize nor quantify the onset of surface roughening very well.

Si(001) undergoes a similar transition at approximately the same reduced temperature,  $T/T_m \simeq 0.8$  [5]. The step energies in Si have been determined from observation of step fluctuations in STM and LEEM [4]:  $E_A \simeq 325$  K and  $E_B \simeq 1045$  K. The aspect ratio is of order  $E_B/E_A \simeq 3$ . Total energy calculations predict larger ratios of order 10 [1, 2].  $\Delta$  is

large enough that a path through the DOF phase seems unavoidable.

Two more generalizations might prove to be important in these experimental systems: bi-atomic steps and corner energies in mono-atomic steps. Bi-atomic steps are important in Si(001) at low temperatures. Those known as  $D_B$ -steps are cheaper than mono-atomic  $S_B$ -steps,  $E_{D_B} \simeq 600 \pm 230$  K [1–3]. This is responsible for the step-doubling phenomena in vicinal surfaces [1–4] at temperatures much lower than those considered here. Bi-atomic steps can probably be ignored near  $T = 0.8T_m$ , because kink excitations in  $S_B$ -steps cost much less energy than kinks in  $D_B$ -steps, and therefore the  $S_B$ -step free energy should become smaller than the  $D_B$ -step free energy well before  $T_R$ . If not, bi-atomic steps can easily be incorporated in the above models, although the phase diagram becomes quite complex (e.g. it includes a reconstructed-rough phase, since double steps do not couple to the reconstruction order).

STM and LEEM observations [4] yield large corner energies,  $E_c \simeq 925$  K. They need to be included in the estimates of (pre-) roughening temperatures<sup>†</sup>. More importantly, there is no intrinsic reason why inside and outside corners of terraces should have the same energy. Such an energy difference,  $dE_c \neq 0$ , does not affect the scaling properties of the roughening transition, but is known to wash-out PR transitions [8]. Instead of a sharp discontinuity by an half-atomic height, the average surface height changes continuously with temperature, slowly at low and high temperatures but quickly close to  $T_{PR}$  (see [8, 10] for more details).  $dE_c$  washes out the PR transition in figure 2(a), but the PR induced simultaneous deconstruction in figure 2(b) must remain a sharp phase transition. Such effects are unobservable and academic when the energy difference between inside and outside corners is small, but since  $E_c$  is large in Si(001),  $dE_c$  might be large as well.  $dE_c$  has not been calculated yet by total energy calculations nor measured in STM- or LEEM-type experiments.

In this paper I discussed the ramifications of the switch in the uniaxial direction at each mono-atomic step in Si(001)-type surfaces from the perspective of the theory of equilibrium surface roughening and reconstruction transitions. This aspect gives rise to a new mechanism for PR and a new type of PR induced simultaneous deconstruction. These transitions are likely realized in Si(001) and Ge(001). This research is supported by NSF grant DMR-9205125.

## References

- [1] Chadi D J 1987 *Phys. Rev. Lett.* **59** 1691
- [2] See e.g. Alerhand O L *et al* 1991 *Phys. Rev. Lett.* **64** 2406; 1991 *Phys. Rev. Lett.* **66** 962  
Bartelt N C *et al* 1991 *Phys. Rev. Lett.* **66** 961; 1992 *J. Vac. Sci. Technol. A* **10** 2600  
Pehlke E and Tersoff J 1991 *Phys. Rev. Lett.* **67** 465
- [3] For a review see Webb B 1994 *Surf. Sci.* **299/300** 45
- [4] Bartelt N C *et al* 1994 *Phys. Rev. Lett.* **73** 1656
- [5] Mochrie S and Yoon M 1996 Private communication
- [6] Johnson A D *et al* 1991 *Phys. Rev. B* **44** 1134
- [7] den Nijs M and Rommelse K 1989 *Phys. Rev. B* **40** 4709
- [8] den Nijs M 1990 *Phys. Rev. Lett.* **64** 435
- [9] den Nijs M 1992 *Phys. Rev. B* **46** 10386
- [10] den Nijs M 1994 Phase transitions and adsorbate restructuring at metal surfaces *The Chemical Physics of Solid Surfaces* vol 7, ed D King (Amsterdam: Elsevier) ch 4

<sup>†</sup> Corner interactions are similar to next nearest-neighbour interactions,  $L$ , in the Ising model ( $E_A = K_x + 2L$ ,  $E_B = K_y + 2L$ , and  $E_c = -L$ ). The shift in the critical temperatures is easy to estimate since the critical plane in the Ising model is almost flat. This moves the estimates for  $T_{PR}$  and  $T_R$  in Si(001) from about  $T = 700$ – $800$  K (when we ignore  $E_c$ ) up into the  $T \simeq 1200$ – $1400$  K range, in agreement with observations in [4–6].



- [11] Bernasconi M and Tosatti E 1993 *Surf. Sci. Rep.* **17** 365
- [12] den Nijs M 1988 *Phase Transitions and Critical Phenomena* vol 12, ed C Domb and J L Lebowitz (New York: Academic)
- [13] den Nijs M 1985 *J. Phys. A: Math. Gen.* **18** L549