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Pre-roughening dynamics on Si(100) stepped surfaces: a study by molecular dynamics

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Abstract. The thermal evolution of steps on Si(100) is well studied and experiment indicates that at temperatures below the roughening transition (*i.e.* $T \leq 1000$ K) the displacements of atoms at the step-edge are the basic factor of this evolution. However the evaluation of the nature and participants of these displacements is beyond experimental observations and a theoretical approach is therefore needed. The problem addressed by this study is the identification of the properties of atomic motions of step-edge atoms and this investigation is performed applying an isothermal Molecular Dynamics simulation method to simple stepped configurations on Si(100). The calculations describe the functional dependence of the motions of step-edge atoms on the step type, size and temperature and on the nature of the interatomic forces. Possible mechanisms of kink formations are suggested.

PACS. 64.60.-i General studies of phase transitions

1 Introduction

Stepped surfaces have attracted much interest as they provide a valuable tool for investigating, both theoretically and experimentally, the dynamics of various types of defects, notably steps and kinks. In this field the Si(100) surface is an ideal model system and has been widely studied by a variety of experimental and theoretical techniques. The study of steps on that particular surface is generally performed assuming the absence of deposition flux and of thermal motions of the step atoms. These stationary conditions, adopted to analyze the relative stability of steps [1–4], are used also for the evaluation of the structural changes due to a temperature increase as methods of the statistical mechanics are generally adopted to this purpose. In fact, the so-called terrace-step kink model (TSK, detailed references on TSK can be found in [5]) offers the fermion approach to this problem and is based on the evaluation of the total energy of kinks at the edge of the steps. Owing to the functional dependence of this energy on the temperature T and on the step structure, the TSK energy leads to a phase diagram which is a function of the miscut angle and of T [2,6–8].

The essential agreement of this phase diagram with experiments indicates that the dynamics of kinks is the central factor of the structural evolution of steps in the pre-roughening stage. However, in spite of their importance, the mechanisms of formation and migration of kinks, are unspecified both in theory and experiment. In fact, theoretical studies center on the kink formation energy E_k [9–11]. This quantity, together with some characteristic times, is also the main result obtained from experimental observations [12–14]. In addition, experiments are limited to the properties of 'forced' kinks caused by the azimuthal misorientation. The correlation between these observations and the dynamics of atoms on straight steps is an open question. In fact, the studies on E_k indicate that the energy cost for the formation of a kink containing n atoms is given by $E_k{=}n\epsilon + C$ where C is the larger term and represents the corner contribution. For 'forced' kinks the C contribution is not needed and they are therefore more prone to fluctuate than kinks on a straight step.

The problem addressed by this study is the atomistic description of thermal motions of step-edge atoms and the following calculations, based on isothermal Molecular Dynamics, illustrate the correlation of these motions with the step type and size and with the nature of the interatomic forces. To obtain this correlation the step must maintain an ordered structure and only a limited temperature increase can be allowed. The maximum temperature used in the calculations, *i.e.* 800 K, is, on the basis of the experimental values of C [12–14], too low to observe the formation of kinks. However the calculations offer useful suggestions on this formation and open some perspective on the possibilities offered by high-temperature experiments.

2 Molecular dynamics method

The simulation method, already adopted in [3], is based on the use of the correct zero-temperature structure of the isolated step [1]. According to theory and experiment, the silicon surface with a miscut angle $\theta \leq 1°$ with respect

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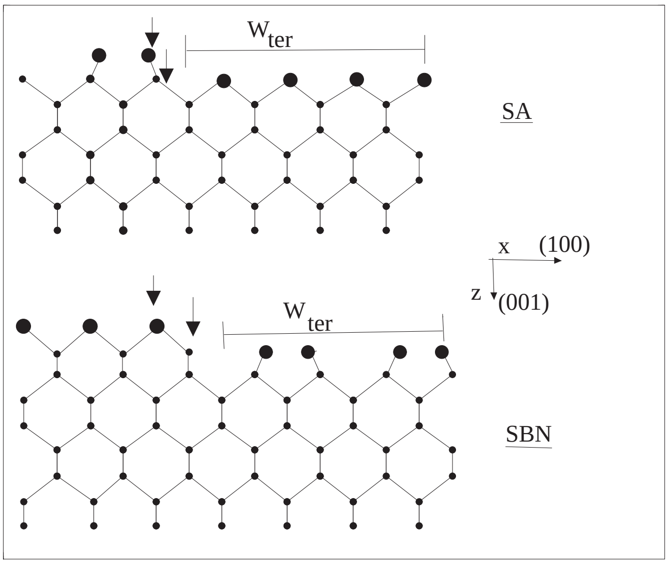

Fig. 1. Structure of the SA and SBN steps (from top to bottom, respectively) used in the simulation. Dimers are marked by the large circles and arrows indicate step-edge atoms used for the evaluation of d_{st}. The figure shows the coordinate axes used for the evaluation of the in-plane and perpendicular component of d_{st}.

to (100) is formed by trains of steps of monoatomic height with a SA or SB edge, *i.e.* with the dimer rows in the upper terrace parallel or perpendicular to the step-edge, whereas in the lower terrace the dimerization axis rotates 90°. On SA second layers atoms are fully bonded while on SB they may have a dangling bond (in the following this structure is indicated as SB non-rebonded or SBN).

A sketchy view of the SA and SBN steps used in the simulation is shown in Figure 1. The upper terrace has a length along x equal to 30 Å while the one of the lower terrace, W_{ter} is variable with a value in a range from 10 Å to 300 Å. In the following W_{ter} is normalized to the surface lattice constant a_o (*i.e.* 3.84 Å). The y boundaries of the simulation cell are held fix by applying toroidal conditions. Atoms up to a depth W_z (whose dimensioning has been taken from [2]) are movable while the ones in the four basal planes underneath are immobile to simulate the attachment of the step to a rigid substrate.

Atomic motions at the wanted temperature T are generated by using Andersen isothermal dynamics, and in order to find a stable step configuration, the duration of each calculation at a given T was approximately 10–20 ps till a stable step configuration was found.

In calculations of surface properties the evaluation of interatomic forces is critical. In fact, in addition to the interactions at the normal nearest neighbor distance in silicon (*i.e.* 2.3 Å), also interactions between dimer rows at a distance $\geq$ 3.8 Å and the change of the dimerization pattern across the step-edge have to be accounted for. The state of the art in computations indicates that these interactions can be correctly described using classical interatomic potentials. Silicon potentials of common use are the Stillinger-Weber and Tersoff potential [15,16] (indicated below as SWP and TP, respectively). Though SWP and TP have been designed to account for the fourfold correlation typical of bulk, they give reasonable results for the reconstruction of the singular Si(100) and for steps on Si(100) [2]. In addition, a potential of a promising versatility has been presented by Mistriotis, Flytzanis and Farantos [17] (indicated as MFFP). The MFFP potential, which derives from studies of clustered structures, has been developed from SWP with the purpose of offering a correct description of multiple interactions with a coordination larger than four though retaining the correct description of the bulk phase of silicon. In MFFP the dependence of these multiple interactions on the interatomic distances and angles has the same analytical formulation as the three-body interaction in SWP but contains two parameters, *i.e.* a decay length and an angular coefficient. By a proper adjustment of these two parameters the potential can be made representative of the multiple interactions of a different type, such as the ones occurring on surfaces as well as on small clusters.

In this study three potentials, *i.e.* SWP, TP and MFFP, have been used. In MFFP the parameters of the four body interactions have been matched to the interaction energies between dimers rows reported in [18]. Calculations for the flat (100) surface showed that both SWP and MFFP lead to a surface energy in the range 1.4 eV (1×1) which is consistent with *ab initio* calculations [19]. In the context of this study, the use of different potentials has a twofold purpose. On one side, it illustrates which features are dependent on the formulation of the potential. On the other side, the comparison of the bulk potentials and MFFP allows to clearly distinguish between effects arising from fourfold, bulklike interactions and from distant, multiple interactions.

3 Dynamics of step-edge atoms

An important issue in kink dynamics is if these atoms originate from mass transport along the step-edge or an exchange of atoms between adjacent terraces is needed. Studies on vicinal surfaces in metals [20,21] indicate, in fact, that kink production occurs with the one or the other of these two modes. Accordingly, our calculations have been focused toward the understanding of whether, or upon which conditions, the motions of step-edge atoms are preferentially oriented perpendicular or parallel to the plane of the step. The occurrence of motions of the first type will favor exchange with lower terrace atoms while in the second case displacements parallel to the step-edge are more probable.

The parameter used to describe these events is the displacement d_{st} of the step-edge atoms given by $d_{st} = (1/N_{st})(\sum_{i,k}(x_{i,k}(t) - x_{i,k}(0))^2)^{1/2}$ where $x_{i,k}(0)$ indicates the 0 K location of the step-edge atom, N_{st} is the number of these atoms and $k = 1, 2, 3$ are the Cartesian components x, y and z. The 'perpendicular' and 'in-plane' component of d_{st} indicate the z component of d_{st} and the one obtained by averaging between its x and y components,

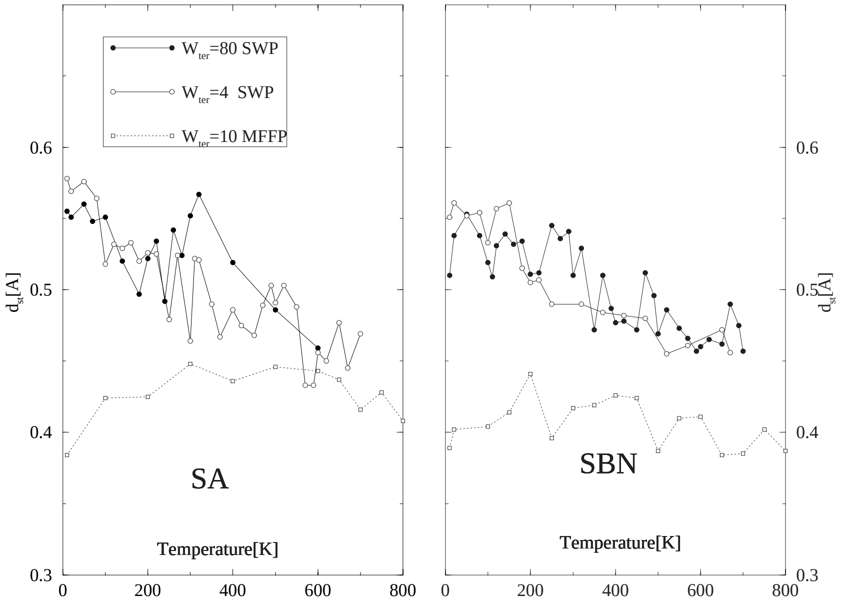

Fig. 2. Temperature dependence of d_{st} on both steps using SWP (circles and continuous lines) and MFFP (squares and dotted lines). W_{ter} is normalized to the surface lattice constant, *i.e.* 3.84 Å.

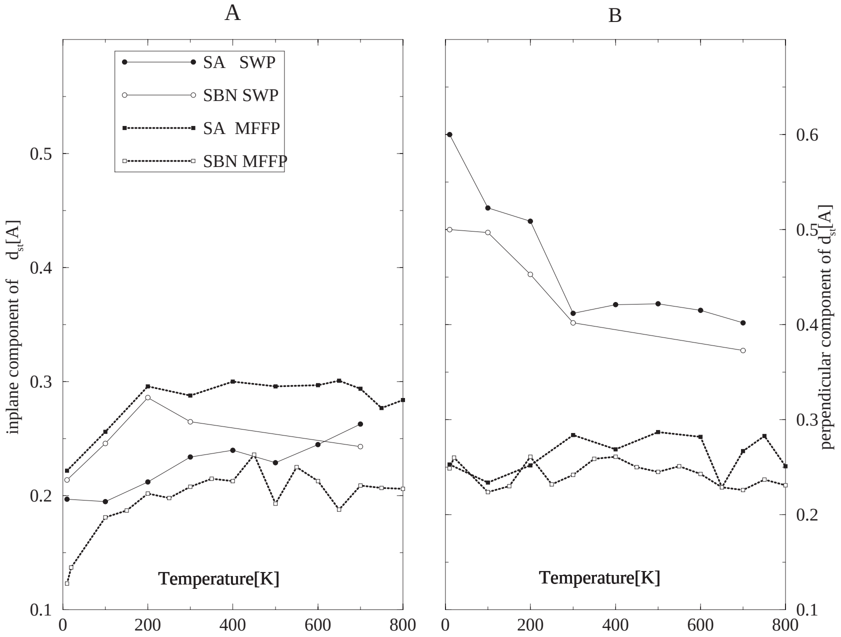

Fig. 3. Projection of d_{st} in the plane of the step (A) and perpendicular to it (B). Symbols as in Figure 2.

respectively. The coordinate axes used for the evaluation of these components is shown in Figure 1 and the atoms used for the d_{st} evaluation are marked by the arrow in the figure.

The temperature response of d_{st} is illustrated in Figure 2. These calculations refer to steps SA and SBN with a variable terrace width W_{ter} in the range from 30 to 300 Å and are obtained using both SWP and MFFP. The in-plane and perpendicular components of these displacements are reported in Figure 3.

In all cases the d_{st} values are limited and well below the amplitude of the surface lattice constant a_o, which is the displacement generally required for the observation of kinks [12–14]. There are, however, three important aspects in these calculations related, respectively, to the method of calculation and to the formation of kinks. First, the atomistic description obtained from MD and a macroscopic formulation obtained, for instance, from Langevin dynamic [22] lead to noticeably different results. The macroscopic theory predicts, in fact, step fluctuations with $1/W_{ter}^4$ dependence while the effect of W_{ter} on d_{st} (Figs. 2 and 3) is inappreciable. This scarce dependence on W_{ter} indicates a weak interaction of atoms at the step-edge with the ones on terrace which is also observed in MD simulations for Si(111) [9]. Second, remarkably different features arise from interatomic forces of a different type. When using SWP the temperature response of d_{st} has a jumping behavior (Fig. 2) which is peculiar of structures of small size and has been often reported, for instance, for small clusters undergoing a melting transition. Similar features have been also found when using TP (omitted for the sake of concision). In comparison with SWP, MFFP enhances the interactions of step-edge atoms with distant surface atoms located on the terrace. This stabilizes the step-edge whose motions are smaller than for SWP and almost independent on T (Fig. 2). A feature, which is however shared by both SWP and MFFP, is that the effects of the dimerization pattern are scarcely perceptible and no clear effect arises from the step type.

The orientation of the components of d_{st}, and the consequence of the values of this parameters on the kink formation mechanisms, represents the third noticeable aspect of the calculations. As shown by Figure 3B, the displacements generated by SWP are prevalently oriented perpendicular to the step-edge (Figs. 3A and B) and the sign of this component (not reported) shows that the step-edge atoms move toward the bulk. This indicates that SWP generates a back-bonding of the step atoms with the bulk stronger than the coupling among them and thus leads to a compressive stress at the step-edge. The stability of the entire step is maintained by the antagonist dilatation in the other atoms on the terrace. On the basis of the z direction of the displacements kink formation requires a complex mechanism possibly involving a vacant lattice site at the step-edge, due to the recession into the bulk of a step-edge atom, and an atom on the lower terrace occupying this vacant site. At variance with this model, the in-plane and perpendicular components of d_{st} calculated with MFFP are almost equal (Figs. 3A, B). This suggests a simple dynamics of kink formation by displacements of step-edge atoms along the step. It is added that the decrease of the z component calculated with SWP for increasing T (Fig. 3B) does not rule out a partial occurrence of these events also when using SWP.

4 Conclusion

In conclusion, this study has investigated the dynamics of step-edge atoms in the temperature regime below the roughening transition. A noticeable result of this study is a critical dependence of these displacements on the potentials. From this dependence different mechanisms of kink formation, involving atom exchange and displacements along the step-edge, are postulated. According to kinetic theories [20,21] these two mechanisms should occur on a macroscopic scale with a time dependence $t^{1/4}$ and $t^{1/2}$, respectively. The correlation of the observation of these characteristic times with our MD calculations should lead to the identification of the kink formation mechanism and of the forces beyond them.

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