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Initial stage of in-phase step wandering on Si(1 1 1) vicinal surfaces

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Abstract

Initial stages of the in-phase step wandering instability on Si(111) were studied by optical microscopy and scanning tunneling microscopy. Initiation of in-phase step wandering induced by surface defects such as pinning sites was observed, although initiation through thermal fluctuations may also occur as an additional pathway. Growth is faster along the steps than along the current direction. Possible mechanisms for the subsequent growth of this unusual instability are suggested. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The kinetic instabilities of the Si(111) surface steps first reported by Latyshev et al. [1] are now widely accepted to arise from an anisotropy in surface diffusion induced by direct current (DC) electric heating of the sample [2–7]. Step bunching instabilities arise during sublimation from a net downhill flux of material, which in the simplest picture would occur due to a downhill electromigration force on diffusing atoms. This simple picture of the electromigration force with positive effective charge is widely accepted above the phase

transition temperature between the 7×7 and 1×1 phases, where a step-down current induces step bunching and serves well in describing many aspects of the instability [3,8–10]. However, on Si(111), there are several reversals of the current direction that causes step instability as temperature increases, which suggests either a temperature-variable origin for the electromigration force direction [11], or a mechanism that may be different than the simplest Schwoebel type instability [12–15].

The physical evidence that sheds light on the origin of the instabilities has been expanded by the recent observation of an in-phase step wandering instability on vicinal Si(111) surfaces [16–18]. After extended heating, the instability appears as a roughly sinusoidal wandering of steps, with all steps having the same period and phase. This in-phase step wandering takes place under heating conditions with a step-down current in the

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temperature range between 1000°C and 1180°C, which will be referred to as range II. While an instability for an isolated step [19,20] would be expected under the same conditions that cause step bunching during sublimation, the in-phase step wandering instability occurs in the temperature range at which a step-up current induces step bunching. The notable fact is that the instability occurs under conditions that oppose step bunching, e.g. either by a net uphill flux, presumably due to a net uphill electromigration force (a negative effective charge) or by a different instability mechanism with a positive effective charge.

Previous theoretical studies on in-phase step wandering under a drift force on the adatoms on terraces do not correspond to the observation of wandering under conditions that stabilize uniform steps [19–21]. A recent study [14,15] however, has shown a mechanism by which this could occur. To address what types of physical models might give rise to the observations, it is very important to know the initial stages of the in-phase step wandering processes.

A short summary of previous studies on inphase step wandering is as follows [16–18]:

- (1) In-phase step wandering takes place on Si(111) vicinal surfaces under a step-down current at temperatures in range II (where a step-down current is stabilizing with respect to step bunching).
- (2) The wandering instability takes places on vicinal surfaces with off-angles up to 13–14° and with any azimuthal direction of the step-down direction [16].
- (3) The period of the wandering steps is in a range of 7–8.5 μ m and depends on the heating temperature with a maximum value in the middle of range II. This is in the case of 5° off specimens after annealing for 24 h [17].
- (4) In range II, in-phase step wandering is also seen in step-up current regions [18], after extended application of the current causes formation of step band and subsequent formation of anti-bands [3,9] followed by the wandering of steps in the anti-bands [18].
- (5) The period of in-phase step wandering of the anti-bands is shorter than 7 μ m [17], suggesting

a perturbation of the formation mechanism that occurs in the step-down current regions.

In the present paper optical microscope and scanning tunneling microscope (STM) studies of initial stages of in-phase step wandering in the step-down current regions are reported. It is also important to observe directly individual wandering steps by STM, because the wandering step images recorded previously using reflection electron microscopy (REM) are severely distorted due to the foreshortening inherent in the technique [1,16,18].

2. Experimental

Specimens with three different off-angles from the Si(111) plane were used in the present study. One, with a off-angle of 5° (here after denoted as the 5° off specimen) was measured using both optical and ultra high vacuum (UHV) STM. The other two, with off-angles of about 0.35° and 0.1° (0.35° off and 0.1° off specimens), were studied with UHV-STM. The optical microscope measurements were made after heating the specimen at 1100°C by DC in the step-down and -up direction in an UHV chamber as has been reported previously [16-18]. For the STM measurements, the specimens $(9 \times$ $2 \times 0.5 \text{ mm}^3$) were chemically cleaned, out-gassed in an UHV chamber of the STM, flash cleaned at 1200°C and annealed at 1050°C by a step-down DC. STM observations were carried out a few hours after quenching from the heating temperature. The STM can scan surface areas as large as $30 \times 30 \ \mu m^2$ [9]. The capability of wide area observations as well as high resolution zoom-in imaging of step structures is indispensable in the present study, because in-phase step wandering involves a few to 10 µm scale arrangements of atomic steps. The temperatures given here were measured by optical pyrometers without correction of emissivity, as in the previous reports [10–18].

3. Results

Fig. 1 reproduces optical microscope images of surfaces of 5° off specimens annealed for (a) 2 h,

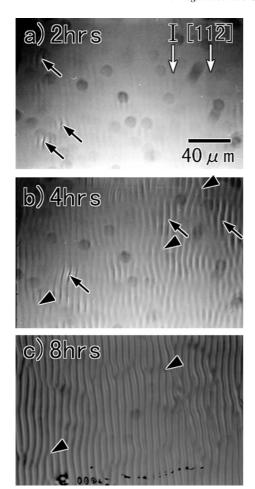


Fig. 1. Optical microscope images of surfaces of 5° off specimens annealed for (a) 2 h, (b) 4 h and (c) 8 h at 1100°C. Arrows in (a) indicate the short vertical line regions of nucleation of inphase step wandering, arrows in (b) indicate local high line contrast on the surface. Arrow heads in (b) and (c) indicate the position of branches. Current direction, and the direction of the net down-hill step staircase of the vicinal surface are both oriented downward in the images.

(b) 4 h and (c) 8 h at 1100°C. The white vertical arrows in Fig. 1(a) show the step-down direction and the current direction. Arrows in (a) indicate positions where short vertical line features are seen. These are local areas of in-phase step wandering as schematically shown in Fig. 2(a). (Circular images in Fig. 1(a), also seen in (b) and (c), are artifacts due to contamination of the microscope lens.) In (b) the surface is almost covered by the vertical line features. A notable fact is that the

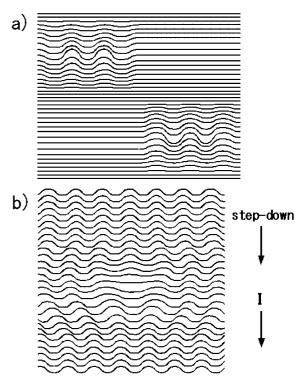


Fig. 2. Schematic drawing of (a) nucleation of in-phase step wandering and (b) a branch of the wandering structure due to a collision between regions nucleated with different phases.

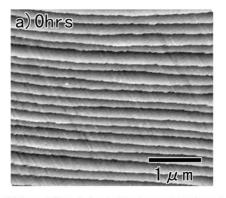
contrast of the line images is not uniform on the surface, and high line contrast is locally seen as indicated by arrows in (b). Differences in line contrast are the result of height differences between the top of the ridges and bottom of the valleys of the in-phase step wandering. These features indicate the growth of the in-phase step wandering, following the initiation events shown in (a). Another notable observation is that following the initiation events, the wandering of steps expands perpendicular to the heating current as well as parallel to the current.

In-phase step wandering starts to form locally on the surface (nucleation), followed by growth of the wandering areas and increase of the height difference of in-phase step wandering in the nucleated areas. The expanding areas meet each other to coalesce. However, the phases of the in-phase step wandering from different nucleation sites may be different, so the directions of ridges or

valleys bend to meet each other or to form branches at the boundaries as indicated by arrowheads in Fig. 1(b) and (c). Such a branch is schematically shown in Fig. 2(b). At later stages seen in (c), the contrast of the line images of ridges, hence the wandering amplitude, is uniform all over the surface suggesting that the height difference of the wandering is almost saturated in (c). The wandering periods seen in Fig. 1 are slightly less than 7 μm, which is smaller than the value (about 8.2–8.3 μm) reported in Ref. [17]. This suggests a smaller period at the nucleation stage. Details of the growth of the amplitude and period of wandering will be described in a separate paper [23]. This growth process of the step wandering areas does not depend on the off-angle.

An example of an STM observation of an initial stage of in-phase step wandering on an 0.1° off specimen is shown in Fig. 3. Fig. 3(a) shows the nearly regular array of straight steps on the surface just after flash heat cleaning. Only small fluctuations of the terrace width parallel and perpendicular to the step lines are seen. Fig. 3(b) shows step arrangements after annealing at 1050°C (lower than that in Fig. 1) for 4 h by DC in the step-down direction. The image is reproduced in such a way that the vertical scale is a half of the horizontal scale. Now, the steps show areas of local in-phase wandering with periods about 7–10 μm, in agreement with the optical microscope study. The wandering amplitude of an individual step decreases from A to B.

The long annealing required for nucleation of in-phase step wandering suggests the need for a rare nucleation event. Possibilities are a trigger due to defects on the surface, which cause bending of steps during sublimation, or an anomalously large thermal fluctuation of a step driving its neighbors to fluctuate in-phase. An inset in Fig. 3(b) shows an enlarged image of the area indicated by a rectangle (the scale difference is corrected), where terminations of steps at emergent points of screw dislocations at the surface are seen. Although a role of screw dislocation in formation of antibands has been pointed out [3,9,18], it is not clear that screw dislocations are the trigger for in-phase step wandering, because the image does not show a strong spatial correlation between the screw dis-



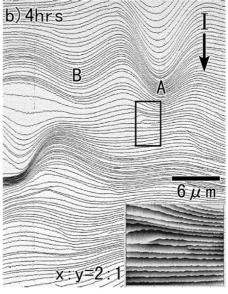
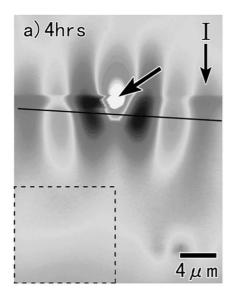


Fig. 3. STM images of an initial stage of in-phase step wandering on the 0.1° off sample: (a) shows a nearly regular array of straight steps before annealing and (b) shows the sample after DC heating at 1050°C for 4 h. A indicates a place where the wandering amplitude is large and B small. The inset is an enlarged image of the area indicated by the rectangle, showing the presence of screw dislocation.

location positions and positions with large wandering amplitude of steps. In the image no contamination is observed around the nucleation site.

Fig. 4(a) reproduces an STM image of a 5° off specimen surface after DC heating for 4 h. A nucleus of in-phase step wandering is seen. A bright dot indicated by an arrow is a contaminant (probably a SiC pinning site). There are seen subside deep valleys along the current direction and shallower valleys in outer areas. A profile of



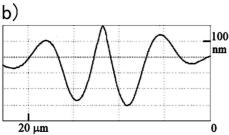


Fig. 4. (a) STM image of a 5° off specimen surface after DC heating for 4 h. The elevated feature indicated by the arrow is a contaminant. (b) Profile of the cross-section of the specimen along the line in the image. The dashed rectangle indicates a place where the surface is nearly flat and maintains the original 5° miscut morphology of the specimen.

the cross-section of the specimen along the line in the image is shown in Fig. 4(b). The profile shows that the wandering period is 6.3 μ m and the height difference is large (70–90 nm) at the nucleation site. In this case the contaminant is considered to have acted as a trigger to initiate the step wandering, yielding isolated structures similar to those observed in Fig. 1(a). Far from the pinning site along the current direction, the surface remained remarkably flat, keeping the original 5° off orientation. The rms roughness ρ of the 11.1 \times 10.6 μ m² area surrounded by the dashed rectangle amounts to only $\rho = 0.75$ nm. On the other hand in an area the same distance from the pinning site but in

the direction parallel to the steps, a larger rms roughness is noted. This shows that growth along the steps is faster than that along the current direction. Initiation of the wandering instability at sublimation pits on the surface has also been observed.

Fig. 5 reproduces an optical micrograph of inphase step wandering formed by step-up (up direction in the image) current heating at 1100°C for 24 h. Dark horizontal bands are step bands formed by step bunching with a step-up current in range II. Bright horizontal bands between the step bands indicated by large arrows are anti-bands. Antibands are formed at the upper side of the step bands [3,9,18] and are straight as indicated by the lower large arrow. However, during the DC heating most of them have changed to short segmented vertical line images, indicated by small arrows. These patterns are due to in-phase step wandering of the anti-bands. The wandering anti-band indicated by a large arrow at the top of the image has covered almost all of the terrace between the neighboring step bands. Thus, it is clear that inphase step wandering of anti-bands grows in the upper direction in the image. This growth process

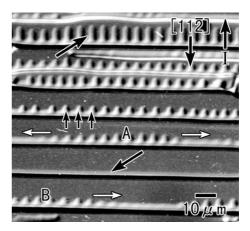


Fig. 5. An optical micrograph of in-phase step wandering of anti-bands formed by a step-up current at 1100°C for 24 h. A lower large arrow indicates a straight anti-band. Short arrows indicate the shortly segmented in-phase step wandering of anti-bands. Large arrow at the top indicates a fully developed in-phase step wandering anti-band that almost covers the surface between step bands. A and B indicate nucleation positions and the horizontal white lines indicate the growth direction of the wandering anti-band.

has been directly observed by REM [18]. On some terraces the vertical lines are not uniform in length, longer, for example at A and B, and shorter at both sides. This means that in-phase step wandering on those terraces nucleated at A and B and grew in the direction indicated by the horizontal white arrows. Growth in this direction has also been studied in-situ by REM [18].

Fig. 5 also indicates that initiation of the instability by nucleation is also the case for in-phase step wandering of anti-bands. The fact that some anti-bands retain their original form without showing wandering suggests that rare defects (as in Fig. 4) may also be nucleation sites. Growth seems to be more rapid along the steps than along the current direction as also has been pointed out in Fig. 4. A similar anisotropy in growth is also present in Fig. 7 of Ref. [18] after an analysis with a correction of foreshortening, although it was not discussed there.

Fig. 6 shows an AFM image (a) and STM images of the 5° off specimen surface (b) and (c) after heating at 1050°C for 15 h. In-phase step wandering is uniformly seen all over the imaging areas with branches in (a) and (b). The bright spots show the positions of very large height difference between ridges and valley and have the same characteristic structure as the contaminant sites in Fig. 4(a). Away from the few contaminant sites, a sinusoidal profile seen in the right of (b) suggests wandering of steps in the sinusoidal form illustrated in Fig. 2. The zoom-in STM image in (c), taken in the valley of the wandering pattern, shows the gentle overall curvature of the steps within the wandering pattern. The profile along a line seen in (c) shows that steps are not single steps, but have undergone the well-known faceting phase transition that occurs simultaneously with the phase transition from 1×1 to 7×7 phase [24,25] near 850°C. This phase separation would have occurred during the quench from the annealing temperature, thus adding the topography of the phase separation to the original pattern of single height steps that existed at the annealing temperature.

The AFM-image shows a representative area of this sample, from which the average distance between pinning sites is found to be approximately $50 \mu m/\sqrt{2}$ –35 μm , that is \sim 5 periods. Previous

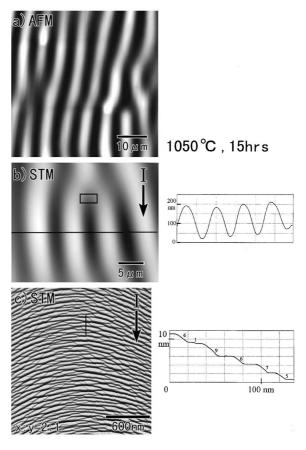


Fig. 6. (a) An AFM image (b, c) STM images of 5° off specimen surfaces after heating at 1100°C for 15 h. Profiles of the cross-section of the specimens along the lines in (b) and (c) are given in the right.

measurements have shown that the same wandering pattern is obtained with a much smaller pinning site density [16–18]. This means that the period of in-phase step wandering is an intrinsic value that depends on temperature and the physical properties of the Si(111) vicinal surface. This may also suggest that faster growth along the steps corresponds to propagation of waves of this period.

4. Discussion

A mechanism of nucleation followed by growth for the in-phase step wandering was deduced from observations at different step densities and annealing times. It was shown that some defects can act as a trigger for nucleation. Growth occurs through spreading of the wandering areas following nucleation. The expansion is not only parallel to the current direction but also perpendicular to it. The expansion seems to be more rapid along the step direction than along the current direction. These features are also the case for in-phase step wandering of anti-bands. When two domains of step wandering meet, branches are formed when the phases of wandering do not match.

The fact that step pinning defects can act as a trigger of nucleation might suggest that the wandering period depends on the density of defects. However, the observations of the period of inphase step wandering on the surface (Fig. 5) that was contaminated by SiC particles showed the same period found on clean surfaces. This means that the period is not governed by the density of defects and instead depends on the physical parameters (temperature and annealing time, and to a lesser extent on off-angle [23]) and an intrinsic basis for the instability. This also suggests that thermal fluctuation itself may also be a trigger of nucleation of in-phase step wandering.

A direct correlation between the screw dislocations and the nucleation sites observed on the 0.1° off samples was not evident. A terminating step at an emergent point of a screw dislocation at the surface may perturb the neighboring steps away from parallel arrangement (see Fig. 3 (b)), which may in turn cause further in-phase step fluctuations. However, it is also possible that on vicinal surfaces with such a low density of steps, nucleation may be caused by thermal fluctuation of steps in-phase. Results of subsequent studies, to be reported elsewhere [23,26] support this possibility: A study on a 1° off specimen showed that after only 1 h annealing at 1100°C, almost all of the surface is covered by in-phase step wandering regions [23]. It is expected that the fluctuation of steps is small on large off-angle specimen surfaces, and is large on small off-angle specimen surfaces. Thus, the nucleation probability may be larger on low off-angle samples.

It is clear that an initiation event consisting of a substantive perturbation of the equilibrium step structure can nucleate the step wandering instability. Following the initial perturbation the instability grows with a well-defined period dependent only on the physical parameters of the system (e.g. not the defect density). In the framework of standard models of step motion in the presence of externally driven atomic drift, the origin of this instability is quite puzzling. In standard models, fluctuations of an isolated step will be damped out, and step bunching will be unstable (e.g. spontaneously return to equilibrium) under the same condition, e.g. when the net adatom flux on the terraces is in the same direction as the step motion [19-22]. In contrast to this standard picture, in range II the two types of instability occur on opposite slopes on the surface with respect to the direction of the external force.

At least two approaches to understanding the instability are possible. One is to consider a reversal of the effective charge in range II, as in ranges I and III, to a negative effective charge as has been suggested [11]. In this case, the question is only to understand the in-phase step wandering instability under an uphill drift force, where a wind force [27] may play an important role. The other possibility is to consider the same positive effective charge in range II as has been suggested experimentally [28]. In this case the question is to understand how step bunching can arise due to an uphill drift force and in-phase step wandering due to a downhill drift force.

For the first approach, to obtain the instability with the force in the uphill direction requires some modification to the usual picture of the electromigration force. In the framework of the simple attachment/detachment limited BCF-type picture, in the presence of the uphill wind force, there is a concentration gradient of adatoms maintained on each terrace in balance with the opposing wind force [9,19,20,29]. When there is a step with a perturbation, the gradient mentioned above induces a lower concentration of adatoms near the ridge than the valley, on both sides of the step (lower and upper sides). This is illustrated in Fig. 7(a), where equiconcentration lines are indicated by thin lines and the concentration gradient is the same all along the step, as is shown on the righthand side of (a). The lower concentration near the

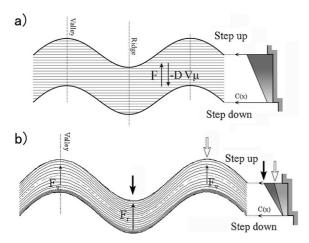


Fig. 7. Schematic drawings, (a) concentration of adatoms in the simple BCF-type picture with uniform wind forces on the terrace and wandering steps, (b) concentration of adatoms under the two assumptions; the step edge shadowing and stronger wind forces at the ridges than at the valleys (for details see text).

ridge will cause this part of the step to retreat faster than the valley part and the step will straighten. This is a well understood stabilizing mechanism [20,21]. Thus, to induce a step wandering instability we must introduce some mechanisms that reverse the above mentioned relative concentrations at the ridge and valley positions. First, we use Rous's [30] recent demonstration that the step-up wind force can be shielded by electron scattering from the step edge over a distance of several lattice constants away from the step edge. Under this assumption the wind force is removed in this small area along the step indicated by a dotted line in (b) and the concentrations on the upper terrace at the ridge and valley position will be equal except for variations due to the Gibbs-Thompson effect. Thus the stabilizing contribution from the upper terrace is reduced. Next, we assume that the wind force is stronger at the ridges than the valleys of the wandering steps. In this case the concentration gradient is larger at the ridge and smaller at the valley as indicated by black and white arrows, respectively in the right-hand side of (b). As a result, the relative concentration on the lower terrace near the ridge and valley position can reverse as illustrated in (b). This will not only remove the stabilizing contribution but also induce an opposing contribution and make the steps wander. However, the growth of the instability will only occur when the original magnitude of the fluctuation is large enough to create a concentration difference that exceeds the Gibbs–Thompson effect of the step curvature, which would cause the fluctuation to decay. Roughly this will occur when the period T and amplitude A of the fluctuation are large enough that $A^2 + T^2 > \Omega \tilde{\beta}/\Delta F$ where Ω is the atomic area, $\tilde{\beta}$ is the step stiffness, and ΔF is the difference in electromigration force between the tops and bottoms of the ridges. With reasonable physical values, this instability should become feasible for micron-scale deviations of the step wandering.

For the second approach, a step bunching instability can occur in the presence of an uphill drift force, if the steps are perfectly permeable [12,14,15]. In this case, there can be no concentration differential across a step edge, and the bunching instability is driven by concentration variations induced by step-step repulsions in regions of variable step separation [14,15]. If the steps are partially permeable, and step interactions are unimportant Liu and Weeks have shown that the bunching instability will occur with the normal (downhill) direction of drift force [29]. Simulations have recently been done by Sato et al. [14,15] for the case of permeable steps and by Suga et al. [31]. In the latter case a drift force along the step in addition to the step-down drift force was assumed, which enhances the ridges. For their prediction of step wandering, the simple estimate of the period Tmentioned above is believed to hold also. Sato et al. showed that an in-phase step wandering instability occurs above a critical threshold of the magnitude of the drift force.

Here it should be noted that in both theoretical approaches diffusion along wandering steps is indispensable for the growth of the in-phase step wandering. This diffusion may cause an expansion of in-phase step wandering along the step direction with the wandering period selected above. The expansion speed along the step direction may be affected by the diffusion along the step direction. On the other hand the expansion speed along the current direction is affected by the diffusion of adatoms crossing narrowly spaced steps and by the

step-step interactions. At present, however, no quantitative result from either theory predicts the observed anisotropic expansion of in-phase step wandering.

More extended stability analyses of the type of mechanisms discussed here, to determine the criteria for their effectiveness, would be of great interest in understanding the observations of the unusual in-phase wandering instability of Si(111). Additionally, thorough evaluation of the range of physical parameters consistent with each mechanism would be extremely valuable in understanding the larger problem of the reversals in step stability with temperature.

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