Onset of Step Antiband Instability due to Surface Electromigration

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Heating by a direct electric current can produce step bunches on vicinal semiconductor surfaces. Under extreme conditions, steps crossing from one bunch to another bend sufficiently to create bands of opposite sign (antibands). Unusual large scale scanning tunneling microscopy images reveal a mechanism where field-induced concentration gradients produce a spatially variable step velocity that drives the antiband formation. A continuum step model allows quantitative analysis of crossing step shapes, yielding an effective charge for the diffusing adatoms of \( q_{\text{eff}} = 0.13 \) electron units at 1270 °C.

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A direct electric current can directionally bias the diffusion of atoms in a solid. This effect, called electromigration, has long been observed in metals and more recently on semiconductor surfaces [1,2]. The pioneering studies of Latyshev et al. [3,4] showed that distinct step patterns arise during sublimation when the direction of the current normal to the steps on a vicinal Si(111) surface is varied. Current in one direction stabilizes the usual uniform step spacing, while current in the opposite direction produces a step bunching instability. Characteristic step patterns arise, with regions of much higher step density (step bands) separated by large flat terraces on which arrays of crossing steps pass from one band to another, often oriented at large angles to the original step direction.

Latyshev et al. [4] also showed that extended application of direct current at high temperature can lead to qualitatively different patterns, where the crossing steps bend through a full 180° to form bands of steps of the opposite sign (antibands). A typical surface revealing these features is shown in Fig. 1. Latyshev identified two mechanisms for antiband formation, one involving sublimation spirals as shown on the center terrace in Fig. 1, and another involving an apparently spontaneous evolution of the shapes of crossing steps as shown in Fig. 2. While many aspects of the initial step bunching are reasonably well understood [5–8], much less is known about the antibands, though there are theoretical predictions of step-bending instabilities arising when the current is parallel to the steps [9,10]. In this study we examine the antiband formation through the evolution of the structure of the crossing steps in detail, taking advantage of the inherent high resolution of the STM. Tracing the shape changes of crossing steps provides much insight into electromigration in the presence of strong sublimation and the physical processes governing this elusive high temperature regime.

The experiments were performed in an ultrahigh vacuum chamber (base pressure \( 7 \times 10^{-11} \) mbar) equipped with a commercial STM and LEED. The 9 × 2 × 0.4 mm³ samples, cut from a nominally flat 0.01 Ω cm Si(111) wafer, were cleaned by flashing for 1 min to 1250 °C. For the preparation of the electromigration-induced surface structures, the samples were heated to 1270 °C by passing an electric current directly through the sample in the direction approximately perpendicular to the step edges. Typical sample current was 4.5–5.1 A, with a corresponding voltage drop of 4.7–5.2 V. The temperature of 1270 °C was chosen because it is high enough to allow antibands to develop in a conveniently short time [3]. At this temperature, the rate of sublimation is approximately \( R_e = 2 \) ML/s (ML is monolayer) [11,12]. At higher temperatures (1300 °C), formation of vacancy islands disturbs the crossing step trains and renders the situation too complex for quantitative analysis.

![STM image of Si(111) after 15 min cumulative heating at 1270°C with direct current in the step-down direction.](image)

**FIG. 1.** 12.2 μm × 12.2 μm STM image of Si(111) after 15 min cumulative heating at 1270°C with direct current in the step-down direction. The formation of widely spaced step bunches, which are composed of single steps with approximately 10 nm spacing, leaves wide terraces, here on the order of 2–3 μm across. The average bunch separation L increases slowly, according to \( L \propto t^{1/2} \) [18,24,25]. (\( dL/dt \approx 2 \) nm/s for \( L = 4000 \) nm at this temperature). A strong sublimation of 2 ML/s [11,12] generates monostep spirals at dislocation sites (center terrace). Trains of crossing steps, as visible on the rightmost terrace, retract quickly in step flow, with a typical velocity of 3000–5000 nm/s, which is determined by \( \nu_0 = R_e d \), with \( d \) the spacing between the steps.
The set of STM images in Fig. 2 illustrates the evolution of structure due to electric current in the downhill direction of 1270 °C. Step bunches appear as black and antibands as white vertical ribbons. The initially straight and parallel steps bunch together (Figs. 2a and 2b) via the formation of step loops which cross the terrace and join a neighboring bunch [4,13]. As the average bunch separation $L$ increases, crossing steps become fewer and bend away from the bunches (Figs. 2c and 2d). The center segments of the crossing steps rotate further and shift to the downhill side, giving the steps an asymmetric shape (Figs. 2e and 2f). The center segments continue to expand and eventually merge into continuous antibands (Figs. 2g and 2i).

Previous studies [14,15] have shown that the migration of atoms to and from steps on Si(111) is limited by a competition between the rate of attachment/detachment at the step edges [16], and the rate of diffusion on the intervening terraces [17]. In the presence of an electromigration force $F_{\text{eff}}E$, with $q_{\text{eff}}$ the effective charge and $E$ the electric field, even a small barrier to attachment at the step edges results in the formation of a concentration gradient of adatoms across the large terrace between the bunches [18]. The electromigration force pushes adatoms from the uphill side of the terrace (left side in images) to the downhill side, where they pile up in front of the bunch. The associated adatom chemical potential $\mu^t$ on the terraces between the straight step bunches assumes a sawtooth profile as shown in the inset in Fig. 3.

$$\mu^t(x) = Fx + \mu^t_0,$$

with $x = 0$ defined as the midpoint between the bunches.

What is the fate of a crossing step in this environment? In the attachment/detachment limited case [19] the step velocity $\nu_{\text{cr}}$ is proportional, via a rate parameter $\kappa$, to the difference between the step chemical potential $\mu^s$ and the terrace chemical potential on both sides of the step. The step chemical potential is determined by the step curvature $K$ and the step stiffness $\tilde{b}$ (the free energy cost of bending the step) via $\mu^s = -\beta a^2 K$, where $a$ is the atomic spacing. We make here the simplest possible approximation for the terrace chemical potential $\mu^t(x)$, assuming that the linear variation shown in Eq. (1) continues to hold even in the presence of a crossing step. Thus the crossing step velocity is given by

$$\nu_{\text{cr}} = \kappa c_0 a^2 [\mu^s(x) - \mu^t(x)]/kT,$$

where $\mu^t(x)$ is given by Eq. (1).

In the description above, the steps are implicitly assumed to be impermeable [20]. Given this assumption, the concentration gradient defined in Eq. (1) will have a discontinuity at a step edge that moves in the direction of the electromigration force.
FIG. 3. Comparison of measured step shapes to the prediction of the simple model of Eqs. (1)–(2), with the value of the effective charge as the only undetermined fit parameter. All images are from different regions on the same sample, which was heated with direct current at 1270 °C. Heating times were 5 min for (a) and (b), and 8 min, for (c). The images are shown at the same scale. (a) $L = 1800$ nm, $d = 1100$ nm; (b) $L = 2780$ nm, $d = 2530$ nm; (c) $L = 3100$ nm, $d = 2000$ nm. The three fit curves superimposed on each image correspond to $q_{\text{eff}} = 0.10$ (dotted line), $q_{\text{eff}} = 0.13$ (solid line), $q_{\text{eff}} = 0.16$ (dashed line). The inset drawing shows schematically the variation of the chemical potential and retraction velocity of a straight step across the terraces between straight step bands. The electric field, as is the case for normal step bunching. Here we consider arrays of crossing steps moving in the $y$ direction, perpendicular to the direction of the applied field. If diffusion on the terraces is rapid enough, then the concentration can adjust to the external applied electric field so that there is no concentration gradient in the $y$ direction, and thus no discontinuity at the crossing steps. In Eq. (2), we explicitly make the assumption that this is the case in the early stage of shape evolution, where the crossing steps move with a constant velocity. The key physical consideration governing this approximation is that a non-negligible fraction of the adatoms on a terrace separating adjacent crossing steps can diffuse between any two points on the terrace before either step attachment or sublimation takes place. Because the diffusion length at 1270 °C is known from step flow measurements [3] and from measurements of vacancy nucleation [21] to be much greater than 2 $\mu$m, and possibly as large as 25 $\mu$m, it is reasonable to assume that this criterion will be met under steady state conditions. We expect the breakdown of these assumptions as the evolution of the step shape progresses. In the late stages the effects of concentration gradients in the $y$ direction on the steeply inclined crossing steps, which we call step shadowing, must be taken into account. Experimental observations of the shape evolution just prior to the onset of the instability (not shown) suggest that Eq. (2) does break down in this way.

The effect of the chemical potential gradient on the evolution of step shape can be illustrated by considering an initially straight crossing step, for which the chemical potential $\mu^S$ due to step bending vanishes. Under sublimation, the gradient of the terrace chemical potential will cause the left side of the step to retract more quickly than the right side (see Fig. 2d, where motion of the crossing step is from top to bottom). This driving force becomes larger as the system coarsens and the average terrace width increases. This leads to an S-shaped deformation which, for small enough concentration gradients (e.g., small $F$ or small terrace width $L$), can evolve into a steady-state shape where the terrace chemical potential $\mu^S(x)$ is compensated by the balancing step chemical potential $\mu^S(x)$ generated by the step curvature. At larger gradients (produced experimentally as the terrace width increases and theoretically when $FL^2/\beta a^2 > 2$), a steady state cannot be achieved. Then the S-shape grows without limit, which we define as the onset of the instability. Experimentally, this onset progresses through a rotation of the direction of retraction as shown in Figs. 2e and 2f, ultimately leading to overlap of the regions of reverse step direction, as shown in Figs. 2g and 2h. The 3D representation in Fig. 2i displays two fully developed antibands on the right sides of the wide terraces. The left terrace of the image shows an abrupt transition to an antiband, which might have been triggered by the abrupt change in terrace width at that point. These later stages of shape evolution cannot be described using the simple model of Eq. (2). The most likely origin of this discrepancy is step shadowing.

A quantitative test of this picture can be performed for the early stages of step bending by numerically integrating Eq. (2). The reference value for the terrace chemical potential $\mu^S(x = 0) = \mu^0$ was estimated by equating the rate of step flow under the known sublimation conditions, $v_0 = R_c d$ (where $d$ is the distance between crossing steps and $R_c$ is the sublimation rate), with the expression for a straight step in the absence of an external field, $v_0 = -\kappa c_0 a^2 \mu^0 / kT$. The value of the step stiffness $\beta$ at 1270 °C was extrapolated from its experimentally determined value of 30–50 meV/Å at 900 °C [16,22] by using the analytical expression for step stiffness from the Ising model [23]. The value obtained was 28 meV/Å. The attachment/detachment rate parameter was also extrapolated from the value of $\kappa c_0 a^2 = 3.5 \times 10^5$ Å/s previously measured at $T = 900$ °C [16]. This extrapolation was performed by comparing the measured rates of electromigration-induced step bunching at
900 °C and 1270 °C [24,25] and yields \( \kappa c_{eq} a^2 F/kT = 6.5 \, \text{s}^{-1} - R_c \). Given these reasonable estimates of the parameters, the effective charge of an adatom \( q_{\text{eff}} \) enters the calculation as the only adjustable parameter, via the rate parameter \( \kappa c_{eq} a^2 F/kT \) and Eq. (1) with \( F = q_{\text{eff}} E \).

The numerical solution of the equation governing the step shape specifically assumes that the relaxation of the step shape can keep up with the slow increase of the terrace width between bunches as the system coarsens. For measured images with different values of the bunch separation \( L \) and crossing step separation \( d \) we fitted calculated steady state shapes to our experimental observations. This is illustrated in Fig. 3, where all the STM images were taken from the same Si sample to minimize the temperature variation. These three examples demonstrate how sensitively our model reacts to variations of the effective charge \( q_{\text{eff}} \). The best fit was achieved with \( q_{\text{eff}} = 0.13 \). Testing the sensitivity of the result to variation in the other parameters showed that the largest sensitivity in the determination of \( q_{\text{eff}} \) is to the value of the step stiffness, with lesser sensitivity to the rate parameter. For instance, a 10% increase in the value of the step stiffness changed the best fit value of the effective charge to 0.16, about a 15% change. The use of the attachment/detachment limit for determining the effective charge to the later stages of structural evolution, where step shadowing clearly becomes important, is in progress.

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