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Si(1 1 1) step fluctuations in reflection electron microscopy at 1100 °C: anomalous step–step repulsion

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Abstract

Using reflection electron microscopy (REM) we study step fluctuations of Si(1 1 1) at 1100 °C. Sublimation is compensated by flux from a nearby crystal. The fluctuation behavior is qualitatively like that at 900 °C (where evaporation is negligible), with unexplained quantitative differences. Regarding the three parameters of the step continuum model of vicinal surfaces, the step stiffness is about half that at 900 °C, in agreement with theory. Step repulsions are at least six times as strong as predicted from 900 °C, suggesting non-equilibrium effects probably due to electromigration from the heating current. Temporal correlations have a large initial offset (due to slow scanning relative to fluctuations) but show scaling behavior.

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In this conference paper, we summarize the highlights of a very recent analysis [1] of reflection electron microscopy (REM) [2] data on slightly vicinal Si(1 1 1) at high temperature, $T = 1100\,^{\circ}\mathrm{C}$, at which there is sizable sublimation, ~0.015 BL/s [1 bilayer (BL) $\equiv 1.56 \times 10^{19}$ atoms/m^2], including figures and some aspects of the results not included in that paper. Essentially this study is a follow-up on an earlier investigation of equilibrium fluctuations on this surface at 900 °C [3], in the narrow thermal range above the 7×7–"1×1" phase transition (~860 °C), where steps are very mobile, but sublimation is insignificant. A capillary-wave analysis of REM data taken at 900 °C showed that the fluctuations were not due to periphery diffusion and that (if 2D evaporation–condensation kinetics are assumed) $\sim 10^6$ attachment or detachment events per second occurred at step sites. In the present experiment, we estimate a net loss per step site of 11 atoms/s. To replenish this loss, a second Si "source" wafer is placed just ~100 μm away and heated independently to a comparable T, as described elsewhere [4,5].

In our experiment the Si sample is in "steady state": there is no net change in mass, i.e. steps on average neither advance nor retreat. The question of whether

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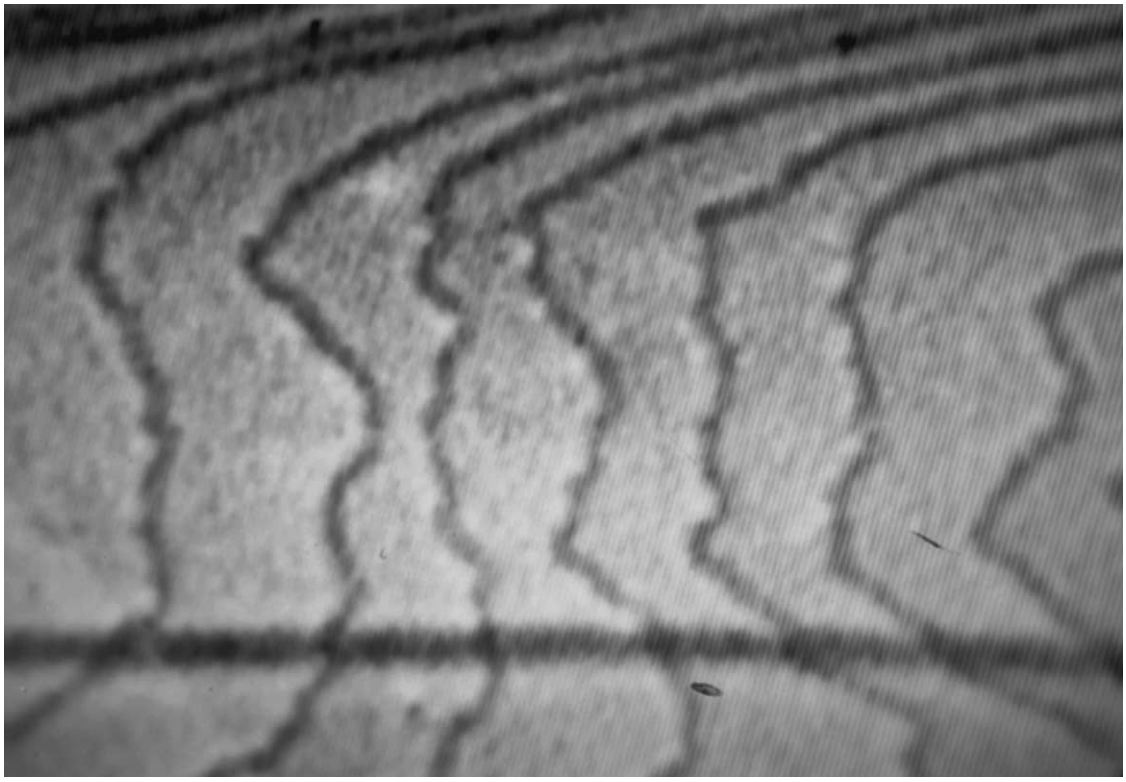

Fig. 1. Photo of a typical REM image frame, similar to Fig. 1 of [1]. The $\hat{x}$-axis is determined by the nearly-vertical line connecting the sequence of sharp crests of the steps (dark curves) on the near-right side of the image. There is a 35-fold compression along the perpendicular, nearly-horizontal $\hat{y}$-axis. Only data from the nearly-straight part of the steps nearly parallel to $\hat{y}$ were analyzed.

the system is in equilibrium is more subtle. In the earlier experiments at ∼900 °C, below significant sublimation, the system was in 2D equilibrium, with surface mass strictly conserved. Since here the adsorption/desorption events are orders of magnitude less frequent than attachment and detachment from steps onto terraces, one would expect little change from equilibrium. Seemingly the major reason that the system is not in equilibrium is that the dc heating current produces electromigration, leading to a net driving force on the steps (in this case in the unbunching direction).

In the REM [2] experiment, performed in Marseilles, frames were recorded in SECAM format (but at 24 frames/s) with a Sony video camera from a TV screen using helical scanning with 330 lines resolution. A sample frame is shown as Fig. 1. Several challenges impede quantitative analysis of this data. In addition to the usual foreshortening—by nearly 35:1 along an axis close to the mean step direction[3] (called $\hat{y}$ in "Maryland notation")—the double-crystal configuration created instabilities in the images, e.g. sporadic jumps due to electrostatic discharges. Furthermore, images of steps appeared and disappeared in successive frames, confounding extraction of terrace-width distributions or temporal correlation functions. We analyzed a range of 5000 frames out of 12,000 for which these effects were least problematic.

In the continuum step model [6], which has been remarkably successful in accounting for a broad range of morphological-evolution phenomena [6,7], the behavior of the step is described using parameters on an intermediate scale larger than atomic: specifically, one seeks the step stiffness, the strength of the interactions between steps, and a third parameter

[3] The 400 pixel diameter corresponds to 1.8 and 62 μm in the $\hat{x}$- and $\hat{y}$-directions, respectively.

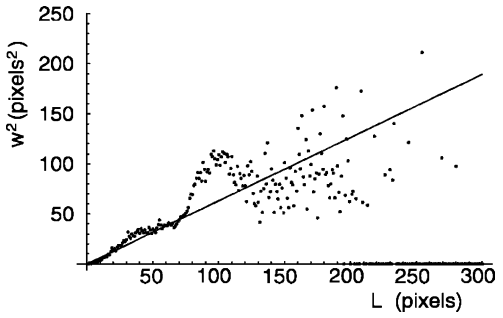

Fig. 2. Graph of spatial correlations using equipartition viewpoint of Eq. (1). L is the length along the step ($\hat{y}$), with a pixel representing 155 nm. The mean square width w^2 is in the perpendicular direction, for which a pixel is 4.5 nm. Most of the data lie below 80 pixels, so the hump and spread tail seem to be artifacts of the limited data. Fits of the first section of data produce similar results to fits of the entire range.

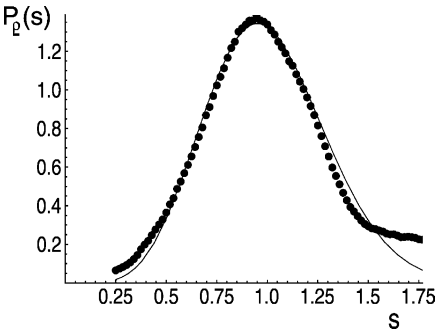

Fig. 3. Plot of the terrace width distribution $P_\varrho(s)$ vs. $s \equiv \ell/\langle\ell\rangle$, and the best fit using Eq. (2).

characterizing the dynamics. The first and third can be gleaned from the step fluctuations of individual steps and the second from the distribution of their spacings.

In [1], the step stiffness $\tilde{\beta}$ was extracted by finding the step diffusivity from the spatial correlation function $G(\Delta y) \equiv \langle[x(y+\Delta y) - x(y)]^2\rangle$, yielding the estimate $\tilde{\beta} = 16.3 \pm 1.8$ meV/Å. Thus, the value of $\tilde{\beta}$ is about half the $\tilde{\beta}$ of 30 meV/Å obtained at 900 °C [3], comparable to sophisticated estimates including the geometry of Si(1 1 1) [8].

Alternatively, as shown in Fig. 2, one can measure the mean square wandering of the entire step. From equipartition comes the expectation that $\tilde{\beta}$ can be deduced from the mean square deviation $\langle x^2\rangle$ [9]:

$$\langle x^2\rangle = \frac{k_B T L}{12\tilde{\beta}} \qquad \text{(free ends)} \tag{1}$$

where L is the length of step (along $\hat{y}$) over which $\langle x^2\rangle$ is found. In this way, REM measurements [2] led to [9,10] the estimate $\tilde{\beta} \approx 46$ meV/Å at 900 °C. (In that case, fixed ends in the model of the step as a vibrating string seemed appropriate, so that 6 replaces 12 in the denominator of Eq. (1).) For our data at 1100 °C (which seemingly corresponds to free ends), analysis based on Eq. (1) yields $\tilde{\beta} \approx 12$ meV/Å, about 3/4 the value from the diffusivity analysis. Also, as seen in Fig. 2, the fit is considerably noisier, particularly when step lengths are greater than 75 pixels (so ~12 μm). (Note that, perhaps coincidentally, if free ends are used in the analysis of the data at 900 °C, the estimate of $\tilde{\beta}$ is reduced to 23 meV/Å, about twice that obtained by our similar analysis at 1100 °C.)

The second parameter of the step continuum model describes the strength A of the elastic step–step repulsion, of the form A/ℓ^2, where ℓ is the step spacing. The standard way to estimate A is to analyze the terrace width distribution (TWD). Some of us have argued repeatedly [11–15] that the TWD can be well described by the "generalized Wigner distribution":

$$P_\varrho(s) = a_\varrho s^\varrho \exp(-b_\varrho s^2), \tag{2}$$

where $s \equiv \ell/\langle\ell\rangle$, while a_ϱ and b_ϱ are [ϱ-dependent] constants that assure normalization and unit mean, respectively [11,14]. The value of ϱ is obtained by optimizing the fit of Eq. (2) to the data (Fig. 3).[4] From ϱ one can quickly obtain[5] A [16]:

$$\tilde{A} = \frac{\varrho(\varrho-2)}{4} \quad \text{and} \quad A = \frac{\tilde{A}(k_B T)^2}{\tilde{\beta}}. \tag{3}$$

Using Eq. (2) we deduced $\varrho \approx 5$, so that $\tilde{A} \approx 4$. To check for possible bias toward broader TWD due to steps not appearing in some images, we also analyzed

[4] In studying physical (in contrast to Monte Carlo) data, it is usually best to perform a two-parameter fit in terms of ϱ and an effective mean step separation rather than a single-parameter fit just to ϱ [13].

[5] Alternatively, from the measured variance σ^2 of the TWD, one can obtain $\tilde{A}$ using the relation [13] $\tilde{A} \simeq (\sigma^{-4} - 7\sigma^{-2} + 27/4 + 35\sigma^2/6)/16$. (Conventionally, the TWD is fit to a Gaussian, from which the variance is gauged via the width at half maximum. This technique provides an adequate approximation if $\tilde{A}$ is not too weak [13]. However, since Eq. (2) is barely more complicated than a Gaussian, there is little to recommend the Gaussian method [except tradition].)

the pair correlation function of the half-dozen steps in the images. The novel procedure, described in detail in [1], shows $\tilde{A} \approx 6 \pm 1$, somewhat larger than deduced from the TWD. (Note also a fit of the first peak of $h_\varrho(S)$ yields $\tilde{A} \approx 4.8$.)

For comparison, at 900 °C $\tilde{A}$ is 1.7 [10].[6] Since $\tilde{A}$ is expected to *decrease* with increasing T (cf. Eq. (3); A is normally rather insensitive to T), our value for $\tilde{A}$ is strikingly large. Since $(k_B T)^2/\tilde{\beta}$ is 2.74 times as large at 1100 °C, A increases by at least a factor of ∼6 and perhaps up to ∼10. This remarkable finding says that the fluctuations in step spacings are strongly *suppressed* compared to the extrapolated equilibrium value at 1100 °C.

After considering several possibilities [1], we believe that the narrowing of the TWD comes from the dc heating current, leading to electromigration-induced biased diffusion similar to the asymmetry due to the Ehrlich–Schwoebel barrier [18]. Qualitatively, since bunching amounts to the effective $\tilde{A}$ becoming negative ($\tilde{A} < -1/4$) [14], current in the opposite direction might well *increase* the effective value of $\tilde{A}$ considerably. (However, electromigration should have a minute effect on the stress dipole at steps, which underlies the actual A.)

To see if the suppresssion of step wandering is correlated with any anomalous changes in the kinetic parameters, we evaluated the temporal correlations via a capillary-wave analysis [3]. By discrete Fourier transform along the step direction, one generates $x_q(t)$ from $x(y,t)$, choosing q as an integral multiple of $2\pi/N$, where N is a fixed number of pixels along the step, typically 64 in our analysis. The correlations of these Fourier components are expected to obey the relation [3]:

$$G_q(\Delta t) \equiv \langle |x_q(t+\Delta t) - x_q(t)|^2 \rangle = \frac{2k_B T/L}{\tilde{\beta} q^2 + c}(1 - e^{-\Delta t/\tau_q}). \quad (4)$$

The denominator in Eq. (4), $\tilde{\beta} q^2 + c$ can be well fit to a quadratic; the deduced coefficient $\tilde{\beta}$ is 12.6 ± 1.3 meV/Å with a miniscule value for c of $(-2.8 \pm 0.7) \times 10^{-8}$ meV/Å^3.

[6] For this $\tilde{A}$—in contrast to larger values [14]—the use of Eq. (2) makes negligible difference from the Gruber–Mullins analysis [2,6,10] formulated in [17].

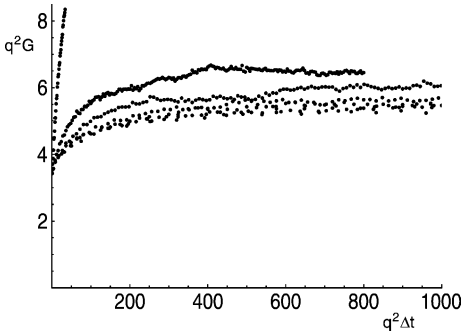

Fig. 4. Scaling plot of $q^2 G_q(\Delta t)$ vs. $q^2\,\Delta t$. Note that the offset scales like q^{-2}.

According to Eq. (4) $G_q(\Delta t)$ should start at the origin; however, the data has a finite positive offset, reminiscent of that seen for "frizzy" steps in STM experiments with inadequate scan rates [19–21]. Correspondingly, our times τ_q in Eq. (4) are comparable to the scan rate of the video camera. (The problem is confounded by the interleave nature of the scanning.) For c negligible, Eq. (4) suggests we collapse the results for various values of q onto a single curve by plotting $q^2 G_q(\Delta t)$ as a function of $\Delta t/\tau_q \to q^2 \Delta t$, as in Fig. 4. While the offset in the STM experiments is typically treated as a constant in the [real-space] repeated-scan correlation function, here the offset clearly scales like q^{-2}. However, the horizontal scaling is only fair, and is not improved substantially by using other powers of q in rescaling the abscissa. In the analysis giving the quoted results above, we ignored the offset. (When we tried subtracting it before analyzing the data [19,20], the fits were much poorer and the deduced parameters unreasonable.)

From the fit to Eq. (4) we simultaneously extract estimates of τ_q, which is expected to scale like q^{-z}, where for an isolated step $z = 2$, 3, 4 for EC, TD, or PD, respectively [6,19,22]; the best fit (shown in Fig. 4) value of z is 1.9 ± 0.2. The time correlation function in real space, i.e. the sum over q of $G_q(\Delta t)$ from Eq. (4), should vary like $(\Delta t)^{1/z}$. As discussed in [1], this analysis is particularly troublesome, but we estimate $z^{-1} = 0.58 \pm 0.06$.

In summary, we have studied Si(1 1 1) at 1100 °C in steady state, i.e. with no net flow of the steps, and compared results for the three parameters of the step continuum model with values extrapolated from equilibrium measurements at 900 °C. While the measured

step stiffness is consistent with expectations, the apparent step repulsion is much stronger than predicted, i.e. the TWD is unexpectedly narrower. Thus, the system seems away from equilibrium even though mass is conserved on average. Probably the surprisingly large deduced value of the step repulsion stems from electromigration due to the dc heating current. Finally, the dynamics seem consistent with the evaporation–condensation mechanism, but the analysis of the temporal correlation function is problematic due to inherent vagaries in the data and because of a large offset at the origin, which nonetheless shows interesting scaling behavior.

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