Bales-Zangwill meandering instability observed in homoepitaxial step-flow growth

Ludwig Schwenger,* Robert L. Folkerts,† and Hans-Joachim Ernst
CEA Saclay, DRECAM/SRSIM, 91191 Gif Sur Yvette, France
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The growth of Cu on vicinal Cu templates has been investigated with helium-atom beam scattering. Step flow on Cu (1,1,17) below room temperature forces steps to strongly meander collectively in phase, leading to the appearance of facets *parallel* to the average step direction. We identify this ''fingering'' with the meandering instability predicted by Bales and Zangwill, resulting from the presence of an adatom uphill current. In contrast to Cu (1,1,17), step flow above room temperature on Cu (1,1,5) leads to a destabilization of the step train *perpendicular* to the step direction. Conceivable origins of this type of faceting are discussed. [S0163-1829(97)50312-2]

The so-called ''step flow problem'' has numerous applications in supposedly unrelated fields, among them, for example, car traffic flow,[1] or the growth of crystals and interfaces.[2,3] In fact, epitaxial growth on vicinal surfaces provides an excellent example of step flow. Vicinals are surfaces with orientations tilted over a small angle with respect to a high symmetry (singular) direction; thus they consist of a regular arrangement of steps separated by terraces. In material science, molecular-beam epitaxy (MBE) on such surfaces has been recognized as a promising avenue to tailor atomically abrupt and smooth interfaces. In the step-flow growth mode, deposited atoms attach directly without island formation to preexisting steps. This advances the steps perpendicular to themselves, and, in steady state, a step train flows across the surface and growth proceeds in a layer-by-layer fashion.

This mechanism respects thus the self-replication property that is essential for ''good growth,'' provided that the flowing step train is stable. However, often, step bunching and faceting have been observed, an undesirable effect for most applications. Now there are many sources, for a destabilization of the step train, such as impurities acting as pinning centers,[4] or steric constraints due to surface reconstructions,[5] but also intrinsic origins related to the adatom attachment kinetics to ascending and descending steps.[6] In order to address the latter aspect, we have investigated the growth via step flow in homoepitaxial, nonreconstructed systems under ultrahigh vacuum conditions.

Burton, Cabrera, and Franck[2] studied step-flow growth a long time ago under the assumption that adatoms bond to ascending and descending steps with equal rates. However, there is by now solid experimental[7] and theoretical[8] evidence that an excess energy barrier (the Schwoebel-Ehrlich barrier) to migration over descending steps exists, which produces an adatom ''uphill current,''[9,10] and therefore directionally dependent capture rate coefficients to steps. Consequently, a terrace, which, for example, is larger than its upper neighbor, receives more adatoms from the incoming flux which then attach preferentially to the ascending step; this increases the velocity of the upper bounding step, and as a result the average terrace width is restored.[6] This ''negative feedback'' argument would imply that step flow should be stable in the presence of a Schwoebel-Ehrlich barrier. However, as pointed out by Bales and Zangwill,[11] the same situation can provoke a growth instability *along* step edges, which manifests itself by a transverse meandering of steps.

Here we provide unambiguous experimental evidence for the existence of the Bales-Zangwill instability. We find that homoepitaxial step flow on Cu (1,1,17) below room temperature produces a collective, in-phase meandering of steps and thus facets *parallel* to the average step direction (SD),[12] while the average terrace width is maintained. Step flow on Cu (1,1,5) in this temperature range leads as well to meandering of (1,1,5) steps, without, however, a specific phase selection. Above room temperature, growth on Cu (1,1,5) produces a faceting of the surface *perpendicular* to the average SD. In terms of the inherent kinetics of the step train, the latter observation asserts[6] the presence of a net adatom downhill current in this temperature range.

The experiments were performed with Ramses II Helium Atom Beam Scattering spectrometer at a base pressure of 2×10^{-10} Torr. Both Cu crystals, sparc cut within 0.2° to the (1,1,5) and (1,1,17) orientation, respectively, had been desulphurized under hydrogen flow for 2 months before being mounted into the apparatus. Cu was evaporated from a radiatively heated, desulphurized disk located about 10 cm in front of the sample. In most experiments, the incident Cu flux, estimated with a quartz balance, was about 3 Å per minute. The stability of both crystal surfaces under equilibrium conditions (thus without flux) for temperatures up to 680 K was checked prior to the step-flow experiments and no spurious diffraction peaks, indicating the presence of other than the nominal orientations, were detected.

The He diffraction pattern of both the Cu (1,1,5) and the Cu (1,1,17) surface parallel to the average SD prior to deposition is characterized by specular scattering only. Deposition of Cu below room temperature on Cu (1,1,5) leads to a slight decrease of the resolution limited part of the specular peak,[13] reflecting a small increase in the magnitude of the excursion of steps from their mean position. Surprisingly, step flow on the Cu (1,1,17) surface in this temperature range produces rapidly appearing additional peaks in the diffraction pattern, see Fig. 1, whose position then do not change on the time scale of our experiment, i.e., within about an hour. Measurements of angular distributions for different perpendicular momentum transfers reveal that these diffraction

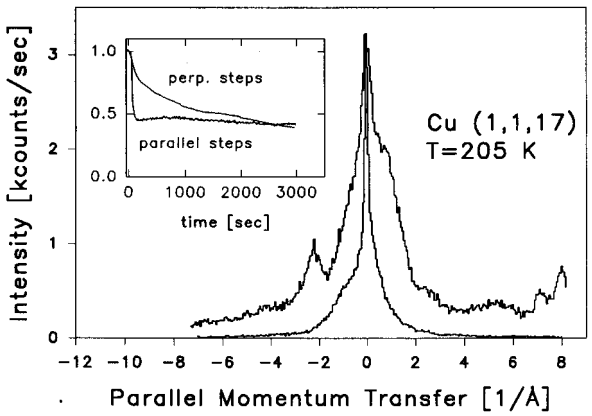

FIG. 1. Helium-atom beam diffraction pattern of the Cu (1,1,17) surface *parallel* to the average step direction (SD) prior to deposition (lower curve) and after growth of about 80 Å Cu in the step-flow mode below room temperature. The incident wave vector is 10.8 $Å^{-1}$, and the angle between source and detector is 92°. The appearance of additional diffraction peaks reveals a collective large amplitude meandering (fingering) of (1,1,17) steps. The inset shows the specular intensity parallel to the average SD and the intensity of a diffraction peak perpendicular to the average SD with deposition time.

peaks are associated with the formation of (−1,1,3) and (1, −1,3) facets; see Fig. 2. Since on both surfaces no additional diffraction peaks in the angular distribution perpendicular to the average SD are detected in this temperature range, we conclude that step flow below room temperature on these surfaces is stable with respect to the average terrace widths. However, (1,1,5) steps meander slightly stronger than thermally induced, and, in the case of Cu (1,1,17), step flow produces a collective (in phase), large-amplitude meandering of (1,1,17) steps, which leads to the appearance of ''fingers,'' or facets parallel to the average SD.

We interpret these findings as the manifestation of the meandering instability predicted by Bales and Zangwill.[11] Their analysis of the step-flow problem demonstrates that advancing steps can be subject to a transverse instability

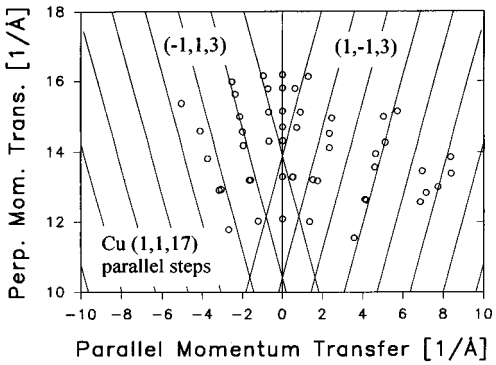

FIG. 2. Map of diffraction peaks parallel to the average SD of the Cu (1,1,17) surface appearing during step-flow growth. The Bales-Zangwill meandering instability leads to the formation of facets in this azimuthal direction. Note that the surface maintains its average distance between steps perpendicular to the average SD at this stage of the experiment.

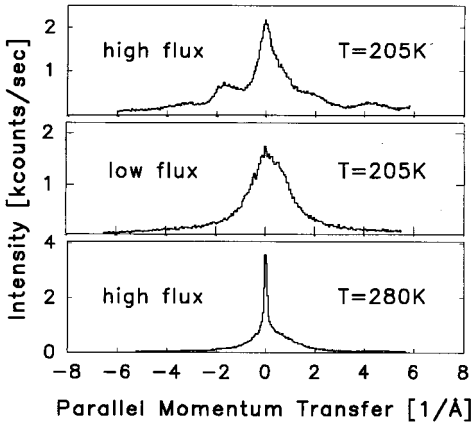

FIG. 3. Flux and temperature dependence of the diffraction peaks associated with the facet formation parallel to the average SD due to the meandering instability. ''Low'' flux refers to an incident Cu flux, which is about a factor 50 smaller.

when adatoms attach preferentially to ascending steps. The presence of the Schwoebel-Ehrlich barrier reduces the supply of atoms from the upper terrace, so that already advanced parts of a step receive more flux from the lower terrace and grow even faster. In this way, for example, a thermally excited modulation in the step is ''kinetically'' amplified.

This kinetically driven meandering is counterbalanced by the increase in the length and thus the line energy of the step. It has been shown before[14] that for Cu surfaces in the temperature range under consideration, the dominating smoothening mechanism is the diffusion of atoms along steps. Thus a prerequisite for the morphological instability to appear is that mass transport along steps is slow enough as compared to the mean rate of advance of the step.[11] Since diffusion along step edges is an activated process, and the step velocity proportional to the incident flux,[2] one should be able to cross over from unstable to stable step flow either by decreasing the incident flux or by increasing the surface temperature.[11] Figure 3 shows the result of these experiments. Indeed, in both cases the diffraction peaks associated with the facet formation along the average SD have largely disappeared, and step flow is nearly perfectly stable. Clearly, the outcome of these experiments strengthens our interpretation of the observed faceting on Cu (1,1,17) in terms of the Bales-Zangwill instability. Presumably, the smaller step velocity on Cu (1,1,5) (together with a strong step-step interaction) explains that the instability is less pronounced on this surface. However, it is unclear why in this case no specific phase is selected.

Recently, the linear stability analysis of Bales and Zangwill has been extended into the highly nonlinear regime by Rost, Smilauer, and Krug.[15] In fact, the observed morphology in our experiments on the Cu (1,1,17) surface resembles quite closely the pattern produced by their Monte Carlo simulation of step flow in the presence of an uphill current.[16] These authors predict that ultimately the vicinal surface will lose its anisotropy and large-scale mounds or pyramids will be formed, just as on singular surfaces.[17] The observation of diffraction peaks perpendicular to the average SD associated with the (1,1,17) lattice indicates that we have not yet reached this stage in our experiment. However, as seen in

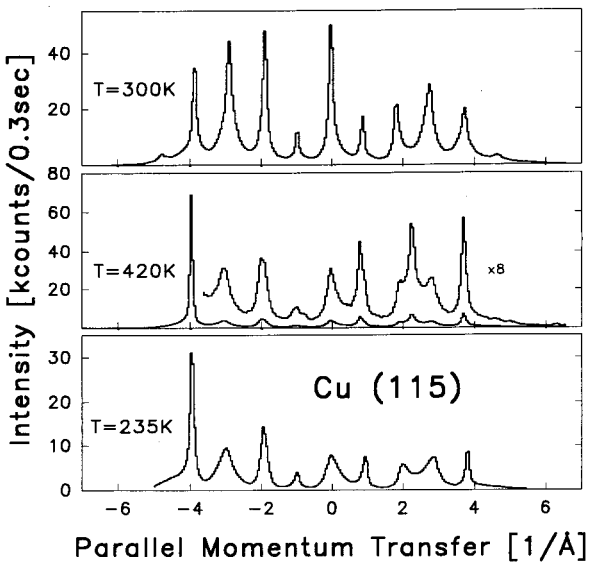

FIG. 4. Helium-atom beam diffraction pattern of the Cu (1,1,5) surface *perpendicular* to the average SD prior to deposition (upper panel), and after growth of about 150 Å in the step-flow mode. Growth above room temperature leads to the appearance of diffraction peaks, which are not associated with diffraction from the (1,1,5) lattice.

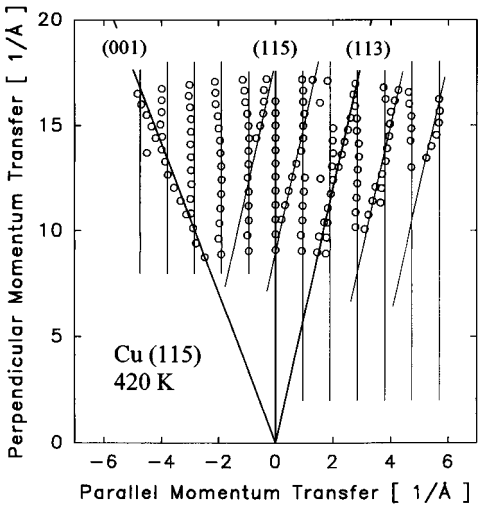

FIG. 5. Map of diffraction peaks perpendicular to the average SD during step flow on Cu (1,1,5) above room temperature. Step flow in this temperature range leads to a destabilization of the step train and to the formation of (001) and (113) facets.

Fig. 1 (inset), while the specular intensity stays nearly constant after a rapid decrease due to the meandering instability and the facet formation parallel to the average SD, the intensity of a diffraction peak perpendicular to the steps continues to decrease slowly with coverage. This is an indication (together with a slight broadening of certain diffraction peaks) that the terrace width distribution, although still peaked around the average width at this stage, gets larger with time, reflecting a tendency towards the loss of anisotropy. In fact, according to Rost, Smilauer, and Krug[15] the isotropic mound formation will start only after deposition of a very large number of layers, if the Schwoebel-Ehrlich barrier is small, this is certainly the case for fcc (001) lattices.[18]

We now turn to the results of our step-flow experiments above room temperature. Figure 4 shows the angular distribution of the (1,1,5) surface perpendicular to the average SD prior to deposition and after having deposited about 150 Å Cu at 420 K. Additional diffraction peaks appear that are not associated with diffraction from the (1,1,5) lattice. Angular distributions taken at different perpendicular momentum transfers reveal the formation of (001) and (113) facets; see Fig. 5. Moreover, from intensity measurements (not shown) of the diffraction peak associated with the (001) facet during deposition, we infer that the extension of this facet increases linearly with time—the faster, the higher the substrate temperature. Thus, for temperatures higher than room temperature, step flow on this surface is unstable with respect to the average terrace width. By contrast, we do not observe this type of faceting in the step flow on Cu (1,1,17) for substrate temperatures as high as 670 K.

These results require, within a description that argues exclusively in terms of the inherent kinetics of the flowing step train, that on the Cu (1,1,5) surface the uphill current is overcompensated[6] by a downhill current for temperatures greater than 300 K, while on Cu (1,1,17) the uphill current persists, up to 670 K at least. Since the strength of the uphill current is proportional to the width of the terraces separating steps[2,9] our findings suggest that the change in sign of the net surface current results from the thermal activation of kinetic processes in the immediate vicinity of descending steps only. One may anticipate that diffusional mechanisms, characterized not only by a higher energy barrier, but also by a higher preexponential factor[19] with respect to diffusion on terraces, become operative with temperature. This way, adatoms in the vicinity of a descending step may preferentially attach to it. Also, ''downward motion''[20] or ''knock out'' effects[21] of adatoms landing directly on top of a step-edge atom could contribute to the downhill current. In this respect, the Cu (1,1,5) surface, due to its short terraces (≈6.4 Å), could then be a marginal case. On the other hand, the small step separation on this surface may imply that in this case, a reasoning exclusively in terms of the adatom attachment kinetics fails, and that step-step interactions, presumably modified by the presence of the adatom diffusion field,[22] could play an essential role. A systematic study of step flow on intermediate vicinals between those investigated so far might help to identify unambiguously the nature of the driving force for this type of faceting. This work is in progress.

In conclusion, the step-flow growth in homoepitaxial, nonreconstructed systems has been investigated over a wide range of temperatures. We provide experimental evidence for the existence of the Bales-Zangwill meandering instability in such a system. The Cu (1,1,5) surface is found to lose its stability in the step-flow growth with respect to the average distance between steps above room temperatures, presumable due to the presence of a net adatom downhill current in this temperature range. Perceiving the exact nature of the driving force for this faceting will be an important step towards a comprehensive understanding of the stability of vicinal surfaces under both equilibrium and nonequilibrium conditions. On the basis of the phenomena encountered in the step-flow growth in homoepitaxial systems, one may antici-

pate that thermodynamics and kinetics compete generally on an equal footing in low-temperature, multicomponent MBE on vicinal surfaces. A controlled use and manipulation[15] of such kinetically driven growth instabilities offer perhaps a promising route for a lateral patterning of surfaces and interfaces on a nanometer scale.

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*Present address: Fraunhofer Institut IMS, Dresden, Germany.
†Present address: Cavendish Laboratory, University of Cambridge, Cambridge, England.

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