

Direct Observation of Misfit Dislocation Glide on Surfaces

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Using scanning tunneling microscopy we have observed thermally induced dislocation glide in monolayer Cu films on Ru(0001) at room temperature. The motion is governed by a Peierls barrier that depends on the detailed structure of the dislocations, in particular upon whether the threading dislocations that terminate them are dissociated or not. Calculations based on the Frenkel-Kontorova model reproduce the threading dislocation structure and provide estimates of the Peierls barrier and dislocation stiffness which are consistent with experiment.

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Thin metal films and close-packed surfaces of many metals are often observed to reconstruct by forming arrays of interfacial misfit dislocations. These dislocations affect many of the physical properties of the surfaces such as surface morphology [1], chemical reactivity [2,3], alloying properties [4], and epitaxial growth [5,6]. Understanding how these dislocations form and move is thus of fundamental importance. Unfortunately very little is known about dislocation motion at surfaces, in part because there are comparatively few cases where the atomic structure of the surface dislocations have been observed while the dislocations move.

In this letter, we report scanning tunneling microscopy (STM) observations of motion of dislocations in monolayer (ML) films of Cu on Ru(0001). In previous work on this system [7] we showed how dislocations can move by exchanging atoms with the thermally generated adatom gas over the film (i.e., by a climbing mechanism). Here we describe observations of thermally induced dislocation glide in which the number of atoms in the Cu monolayer stays constant. We find that, under certain conditions, this motion can be extremely rapid. Whether the glide motion can occur easily depends on the size of the Peierls barrier [8] to the motion. By comparing our observations of the structure and the motion of the dislocations to calculations based on the Frenkel-Kontorova (FK) model, we are able to understand the conditions required for a small Peierls barrier and rapid dislocation motion.

For films composed of less than a monolayer, the equilibrium structure of Cu on Ru(0001) is pseudomorphic. However, extra rows of Cu atoms can be readily inserted into the film by rapid Cu deposition. These added rows form misfit dislocations of pure edge character with the Burgers vector in the plane of the film. Because of the presence of energetically comparable fcc and hcp adsorption sites, each misfit dislocation relaxes into two parallel Shockley partial dislocations bounding a stacking fault, as observed in fcc(111) thin films and surfaces [4]. Figure 1a shows a STM image of such a structure. The bright lines in the STM image aligned with one of the three equiva-

lent second nearest-neighbor directions in the film consist of atoms close to bridge sites which separate the fcc and hcp regions and correspond to the positions of the partial dislocations. The Burgers vector associated with each bright line is the smallest vector that will connect fcc sites and hcp sites. A schematic of the dislocation structure is shown in Fig. 1b. As dislocations cannot end inside the film, the point where they terminate is the location of an edge dislocation that threads out of the film. This threading

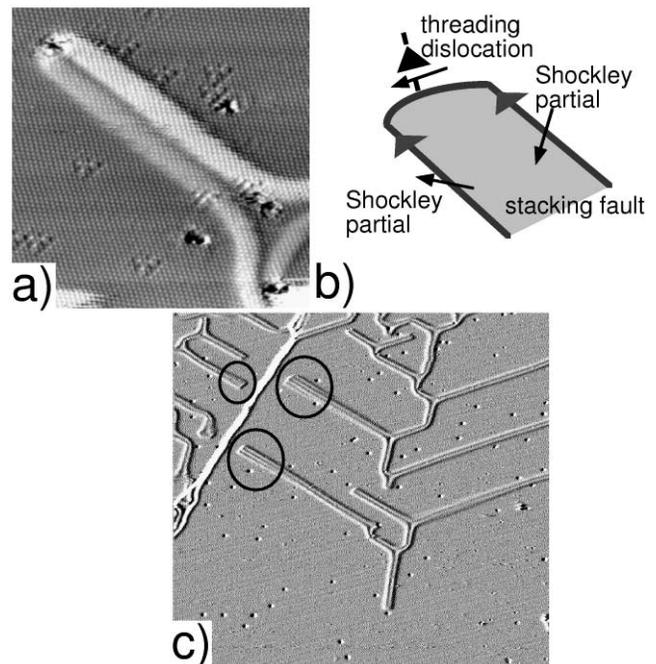


FIG. 1. Misfit dislocations in a Cu monolayer on Ru(0001). (a) STM image (15×15 nm) of a segment of a misfit dislocation terminating in a threading dislocation on the top left corner. (b) Schematic of the dislocation structure. The black arrows denote the Burgers vectors of the dislocations. (c) Lower magnification STM image (120×120 nm, differentiated to enhance the contrast) showing the mesoscopic arrangement of the dislocations ($I_{\text{tunnel}} = 1.0$ nA and $V_{\text{sample}} = 1.26$ V). Note the moving dislocations marked with black circles.

dislocation [9] in 1 ML Cu/Ru(0001) films is, of course, only one atom long [10].

At a larger scale, long segments of misfit dislocations (imaged as double lines) are oriented along three equivalent directions (Fig. 1c), where the segments are connected into Y-shaped superstructures. This image, taken at room temperature, provides evidence for dislocation motion. Some dislocation segments are imaged at a single position, while others are imaged as a superposition of segments slightly offset from one another. In this superposition, dislocations are simultaneously imaged in multiple positions within the same scan line. This multiple image effect is the result of dislocations moving during the imaging process. The dislocations appear most clearly in the regions where they spend the most time (much in the same way as one sees a superposition of a vibrating string at different positions in a time-lapsed photograph). We can rule out any multiple-tip imaging artifact because neighboring defects do not exhibit this behavior, even on the same scan line.

We have observed the motion of several types of dislocation configurations. In Fig. 2a, two segments of a “Y” move in the directions shown in Fig. 2b, while the node moves along the third segment keeping the total length of dislocations (and hence the energy) constant. These segments move distances of tens of nanometers during the time needed to measure a single scan line of the STM image (~ 0.1 s). The motion of the dislocations is in the glide plane for both the Shockley partials and the threading dislocations that compose the segments of misfit dislocations.

There are two obvious possible causes of the dislocation motion: tip effects and thermal excitations. The STM tip is known to modify adsorbate diffusion on surfaces [11–13]. This effect is used for atomic scale [14,15] ma-

nipulation. To check whether tip interactions with the dislocations cause the motion we have scanned with a wide range of speeds, directions, and tunneling conditions. The amplitude of the observed motion is independent of these parameters as long as the gap resistance is kept at or above a $G\Omega$. We thus conclude that tip effects are not the main contribution to the movement.

For thermal fluctuations to be responsible for the observed glide, the energy barrier for dislocation motion (i.e., the Peierls barrier) must be on the order of kT or smaller. To determine if this is reasonable, we have estimated the Peierls barrier within the FK model. The FK model can be remarkably accurate when describing monolayer metal films, as shown by the success in predicting the sequence of dislocation networks experimentally observed in Cu/Ru(0001) as a function of film thickness [16]. The substrate potential is estimated from *ab initio* calculations [17]. To estimate the Cu-Cu interatomic potential we initially follow the procedure of Narasimhan and Vanderbilt [18] and assume a harmonic potential connecting nearest-neighbor atoms, with the spring constant of the harmonic potential chosen to reproduce the experimentally observed distance between Shockley partial dislocations. The Peierls barrier was computed by rigidly displacing the atoms in the stacking-fault ribbon from their equilibrium positions in the close-packed direction perpendicular to the dislocation lines. A constant-energy molecular-dynamics (MD) simulation without threading dislocations showed a pair of Shockley partial dislocations moving sideways with uniform velocity with a periodically varying potential and kinetic energies. The Peierls barrier is given by the amplitude of the potential energy variations. For the movement of the extended dislocation without any terminating threading dislocation this barrier was extremely small, of the order of μeV per atom or less.

However, there is another component to the barrier for glide: the motion of the threading dislocation must be considered. If the Peierls barrier for the threading dislocation is calculated using the usual nearest-neighbor harmonic potential of the FK model, the barrier is large: approximately 1 eV/atom. This barrier would obviously fix the dislocations at room temperature. The reason for the high barrier can be found in Fig. 3b, where the result of a static FK calculation is shown. The core of the threading dislocation is very compact, concentrating into a small region all the mismatch between the stacking fault on one side and the perfect layer on the other. Since Peierls barriers typically decrease exponentially with the width of the dislocation [8], this narrow dislocation naturally has a high barrier. As noted above many dislocations are observed not to move. The structure of the static dislocations is similar to Fig. 3b. A STM image of such a threading dislocation is shown in Fig. 3a. Nevertheless, experimentally, we find that some of the dislocations do move. One possible reason for why there is a high Peierls barrier in the above calculation is that a harmonic potential between Cu atoms

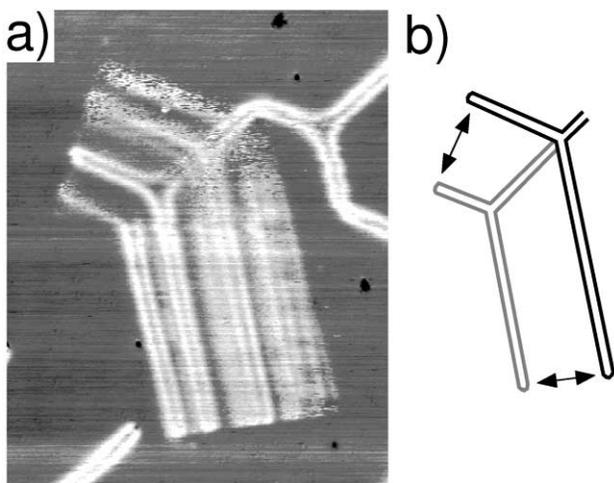


FIG. 2. (a) STM image (51×66 nm) showing moving dislocations. The tunneling conditions were $I = 0.4$ nA and $V = 0.82$ V. The scanning speed is 210 nm/s with the fast-scan direction along the horizontal axis. (b) Schematic of the dislocation in (a).

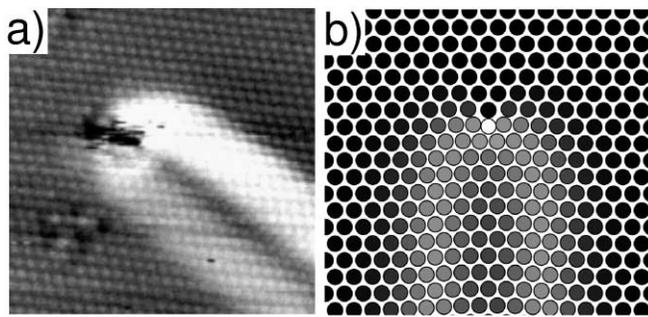


FIG. 3. Threading dislocation at the end of an extended misfit dislocation. (a) STM image (65×65 nm) showing the atomic arrangement close to the core of a threading dislocation. (b) Frenkel-Kontorova model of an extra row of atoms in the film (a misfit dislocation). The gray scale is related to the potential energy of each atom (higher is brighter).

was chosen. The assumption of a harmonic potential is valid only in the case of small interatomic displacements. Because of the presence of threading dislocations in the film, large lattice distortions may exist which require more realistic potentials [19]. The FK model does find a low Peierls barrier, if the interatomic Cu potential is improved by using a more realistic potential such as a Morse potential. The hard core of the Morse potential at small distances prevents the small interatomic separation characteristic of the core of the undissociated dislocations, as shown in Fig. 4c. The calculated Peierls barrier decreases to 10 meV/atom, a value that is consistent with the thermally driven dislocation glide. As shown by the comparison between Figs. 4a and 4c, the calculated structure matches the structure of the moving dislocations [20]. To check whether other interatomic potentials with a reasonable representation of the repulsive core interaction also give a low Peierls barrier, we have repeated the calculations using a Lennard-Jones potential. The results were quite similar.

This structure corresponds to a dissociated threading dislocation: as discussed above, edge dislocations usually relax into parallel partials bounding a stacking fault. The same arguments previously applied to the in-plane misfit dislocations also apply to threading dislocations because they are also edge dislocations, now in (111) planes intersecting the film plane [10]. The diagram of such a dissociated threading dislocation is shown in Fig. 4b (compare this with Fig. 1b). We propose that whether the dislocations are dissociated in the experiment or not is determined by contamination. Typically in images of the core of the undissociated, static threading dislocations there is a dark region which we interpret as an impurity atom: the atoms at the core of the threading dislocation are expected to often be reactive and have been shown to be preferential binding sites for adsorbates [2,3] and metal film growth [5]. Since moving dissociated dislocations lack this dark feature, they are presumably characteristic of the clean surface.

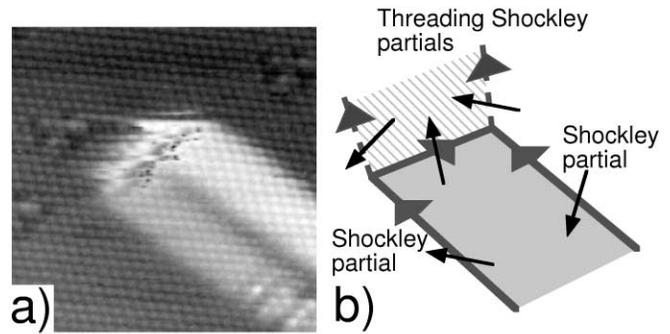


FIG. 4. Dissociated threading dislocation at the end of an extended misfit dislocation. (a) STM image (65×65 nm) showing the atomic arrangement close to the core of a dissociated threading dislocation. (b) Schematic of the partial dislocations terminating at a dissociated threading dislocation. (c) Frenkel-Kontorova calculation of the same starting structure of Fig. 3b but with a Morse Cu-Cu interatomic potential.

If a bent dislocation configuration is introduced as the starting configuration of a constant-energy MD simulation, the dislocations vibrate with a frequency of 45 MHz (see Fig. 5b). The threading dislocation stays dissociated while moving. Since this frequency is much greater than the STM scanning frequency, it provides an explanation for why STM images the dislocation in multiple positions in one scan line.

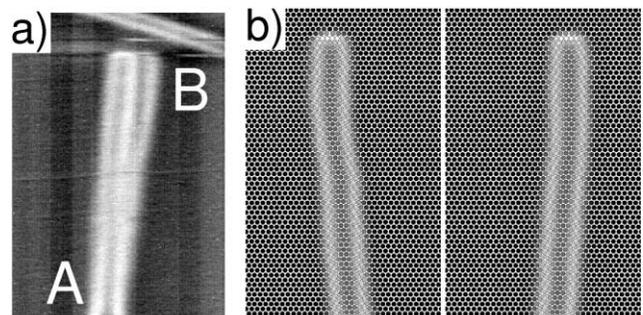


FIG. 5. (a) STM image (15×51.4 nm in size, with $V_{\text{sample}} = -1.85$ V, $I_{\text{tunnel}} = 0.99$ nA, and x -scan speed of 200 nm/s) of a misfit dislocation vibrating with a free end (marked with B) and a fixed end (marked with A). (b) Constant energy MD simulation of a vibrating dislocation. The time between the two frames shown is 70 ns.

From the experimental data we cannot determine whether the dislocation glide occurs by the generation and motion of kinks along the length of the dislocation (see Chap. 15 of [9]). The imaging rate of our STM is likely to be too slow to observe kink motion. However, in the FK simulation we find no evidence for kinks, as shown in Fig. 5, suggesting that the stiffness for bending the dislocations is large compared to the Peierls barrier. To determine the stiffness within the FK model, using the same Morse interatomic potential used previously, we calculated the energy of the dislocations as a function of their orientation. We find that the dislocations behave like strings under tension for small displacements from their equilibrium orientation. The potential energy is given by $V \sim \beta\theta^2$, where the effective stiffness of the dislocation is β , and θ is the angle of the dislocation away from the equilibrium orientation. The value for the stiffness given by our FK model is $\beta \approx 200$ meV/atom, much greater than the Peierls barrier, as expected.

To compare the calculated stiffness with our observations, we can analyze the vibrational amplitude of the segments of misfit dislocation shown in Fig. 5a. Dislocations with a length of ≈ 500 Å vibrate with an amplitude of ≈ 13 Å. Assuming a thermal origin of the vibrations and neglecting the Peierls barrier, we can estimate the stiffness by analyzing the movement of the dislocation in terms of normal modes of a vibrating string with one free end. Populating each normal mode with an average energy of $kT/2$ provides a relationship between the stiffness and the amplitude. Considering only the first mode, the stiffness is $\beta \sim kTl/w$, where the dislocation vibration amplitude is w and its length is l . The experimentally derived stiffness yields $\beta \approx 70$ meV/atom, in reasonable agreement with our FK calculations.

This analysis of the thermal vibrations is similar to the analysis of Treacy *et al.* [21] of images of vibrating carbon nanotubes obtained by transmission electron microscopy. In both studies, because of the large frequency of the vibrations, image intensities represent an average of the states of the systems over a period of time. In principle, this average is calculable by taking thermal averages over the normal modes of the system, as done by Treacy *et al.* This procedure does not work in our case, however. The problem with interpreting the dislocation images in terms of thermal averages is evident in Fig. 2a: there are preferred positions for the dislocations which are not predicted by simple theory. It is possible that defects in the Cu film are responsible for this behavior.

In summary, we have observed by STM the glide of segments of misfit dislocations in a monolayer Cu film on Ru. By comparing with FK model calculations we have shown that the observed movement at room temperature is consistent with a thermal origin. Finally, we point out that this dislocation motion is also a surface self-diffusion mechanism that, in principle, could be competitive with adatom diffusion on close-packed surfaces.

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