Glide and Climb of Dislocations in Ultra-Thin Metal Films

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Abstract. Near-surface misfit dislocations can be directly observed by Scanning Tunneling Microscopy through the distortion on the positions of the atoms of the surface layer. In this way, the behavior of misfit dislocations can be followed in detail. Experiments performed in monolayer films of Cu on Ru(0001) show both climb and glide of the misfit dislocations with atomic resolution. The motion of the dislocations represents a novel pathway for surface diffusion in ultra-thin films.

1 Introduction

Misfit dislocations are likely to appear whenever there is a significant lattice mismatch between a thin-film and a substrate. The misfit dislocations in turn determine many of the properties of the films, such as chemical reactivity [1, 2], elastic properties, etc. Furthermore, in many metal systems with hexagonal substrates the dislocations self-organize providing very well ordered patterns [3, 4] that can be used to grow additional material with a well defined nanoscale order.

The misfit dislocations considered in this work have their glide plane at the film/substrate interface, so they can move easily within the film by glide. In addition, the surface acts as a source of adatoms/vacancies in numbers which far exceed their bulk counterparts. Thus, as we will show in this paper, fast climb of dislocations can occur even at room temperature. An important consequence of this is that the motion of near-surface dislocations, through a combination of climb and glide, can be a pathway for mass transport close to the surface. Glide of near-surface dislocations as a mass transport mechanism has already been detected in nanoindentation experiments in Au(100) by means of a particular dislocation configuration, namely a dissociated dislocation loop [5]. But the lack of climb in that system limits the generality of the process to specific directions.

Many ultra-thin metal films with misfit dislocations present some variation of an hexagonal network of dislocations (moiré networks, bright star networks) with no threading dislocations in the film. Also very common is the presence of stacking fault ribbons or trigons (triangular areas) bounded by partial (misfit) dislocations with threading dislocations at points where different partial dislocations meet [6], shown for example in 2ML Cu/Ru(0001)[7], 1ML Ag,Au/Ru(0001)[8], Pt/Pt(111)[9], Au/Au(111)[3], and Co/Pt(111)[10] among others.

In this work, we will present observations of glide and climb of misfit dislocations in Cu films on Ru. The films have a thickness of one monolayer, and the experimental observations are performed by *in-situ* Scanning Tunneling Microscopy (STM) under Ultra-High-Vacuum conditions.

2 Experimental

The experiments have been performed in two STMs in different UHV systems. The base pressure of both systems is below 9×10^{-11} torr. Additional techniques on both chambers were electron spectrometers suitable for Auger Electron Spectroscopy (AES) and, in one system, a Low Energy Electron Diffraction apparatus. The Ru(0001) substrate has been cleaned by repeated cycles of exposure to oxygen (9 Langmuir of O_2) followed by flashing to 1500 °C. Cleanliness was checked by STM and AES. Terraces larger than 0.5 μm can be routinely found in the clean substrate. Cu films were grown by physical vapor deposition from a calibrated metal doser at rates of the order of 1 ML/min during which the pressure in the system remained below 2×10^{-10} torr. All the films have been annealed after growth to 600 °C in about 10s, and allowed to cool to room temperature before imaging with the STM. Additional Cu was deposited on the previously grown film to create the misfit dislocations. The STM images are presented in gray scale. When necessary to increase the contrast in the presence of steps, either the derivative of the image was added to the original data or the contrast was increased on each terrace individually.

3 Experimental Observations and Discussion

When a Cu monolayer is deposited on Ru(0001) and annealed to 600 °C, a perfect pseudomorphic film is observed. The adsorption of Cu takes place in hcp positions, following the substrate structure [7] and wetting the steps. The difference in lattice parameters (5.46%) is accommodated by elastic strain in the film.

The surface is under tensile stress: if small amounts (below 0.03ML) of additional Cu are deposited at RT, misfit dislocations are nucleated in the film (see Fig. 1). There is a barrier for such nucleation[11]. In addition to the incorporation of atoms into the copper films the adatoms can join the Cu steps. Depending on the width of the terrace and the presence of defects that enhance the nucleation of misfit dislocations, coexistence of terraces with dislocations and terraces where Cu is completely pseudomorphic can be observed (as seen in Fig. 1).

The misfit dislocations so nucleated are perfect edge dislocations oriented along next-nearest neighbors. Due to the hexagonal nature of the substrate with two different adsorption sites the edge dislocations dissociate into pairs of Shockley partial dislocations separated by stacked fault areas[12]. Given that the “correct” stacking sequence for 1ML Cu is hcp, the stacking fault areas have atoms in fcc adsorption sites[7].

When the dislocations are nucleated in the middle of a terrace “Y” shaped structures are formed by the initial nucleation of atoms in fcc positions followed by growth of edge dislocations in the three equivalent next-neighbors directions.

The ends of the dissociated edge dislocations are threading dislocations one atom high. The structure of a trigon is depicted if Fig. 2, where the dislocations are named using Thompson’s tetrahedron notation[12] as described in [6]. Although such notation is valid for fcc materials, we can apply it to Cu/Ru as long as we consider only one Ru terrace at a time: Cu grown on the next terrace would appear as a twin of Cu grown on the previous one due to the hcp nature of the substrate.

The Shockley partial dislocations are imaged in the STM images as bright lines: the distortion in the atomic positions close to their cores corresponds, disregarding distortions on the substrate, to atoms in bridge positions.

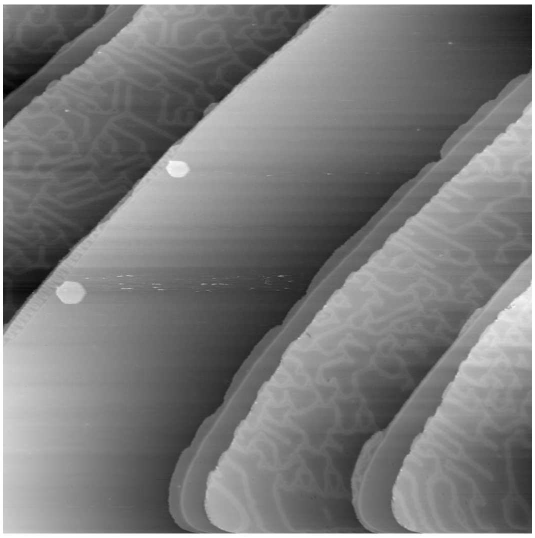

Fig. 1. STM image of a 1ML Cu film on Ru(0001). The image size is 284nm×284nm. Several substrate terraces, covered by the Cu monolayer, can be seen. On some, misfit dislocations are nucleated. Some of the terraces are bare of dislocations (and instead contain nucleated islands).

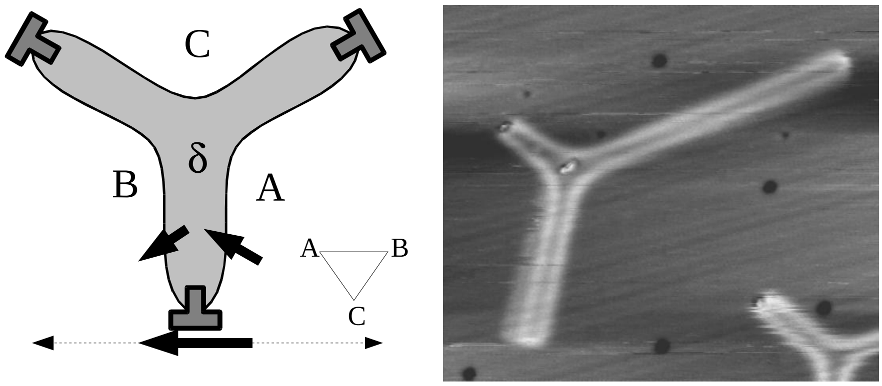

Fig. 2. Left: schematic of the structure of a trigon, showing the threading dislocations as small “T”s. The light grey area contains a stacking fault. The Thomson tetrahedron is also shown. The thick black arrows represent the Burgers vector of the dislocations. The glide direction for the BA dislocations is shown as a dotted line. Right: STM image (37.4nm ×32.4nm) of a trigon.

The dislocations on Cu/Ru are observed to move at RT. STM is a relatively slow technique: in our case, the STM system can obtain images in a time-scale of minutes for images of a hundred angstroms in size. As it is a scanning technique, each individual line takes around two orders of magnitude less time than the full image.

The observations of moving dislocations fall under two distinct categories. First, dislocations are imaged at different positions (see Fig. 3) within the time needed for acquiring one image. Second, a slow change in the length of the dislocations is detected when comparing images taken at the same spot of the surface (see Fig. 4).

In the first case, the effect is very much like an over-exposed photograph of a moving object [13]. Sometimes one end of a long dissociated dislocation is pinned, and the other end

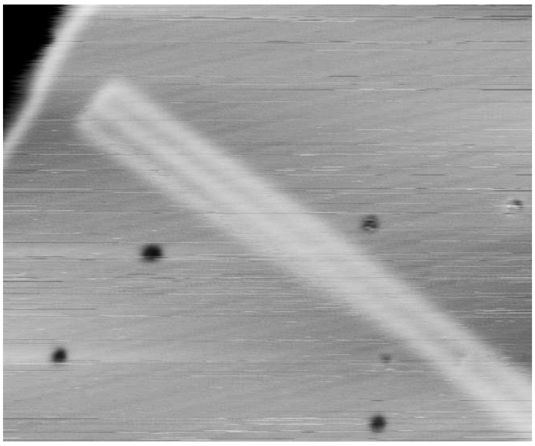

Fig. 3. STM image of dislocation glide. The image size is 32.8nm×27.1nm. One end of the misfit dislocation is observed at two different positions.

resembles a vibrating string (see Fig. 3). Other times, the full dissociated dislocation is moving perpendicular to its axis. In both cases, the motion is much faster that the time needed to obtain even a portion of a single scan. Imaging artifacts can be ruled out, as other features of the surface such as defects and steps are imaged normally. We will refer to this motion as "fast".

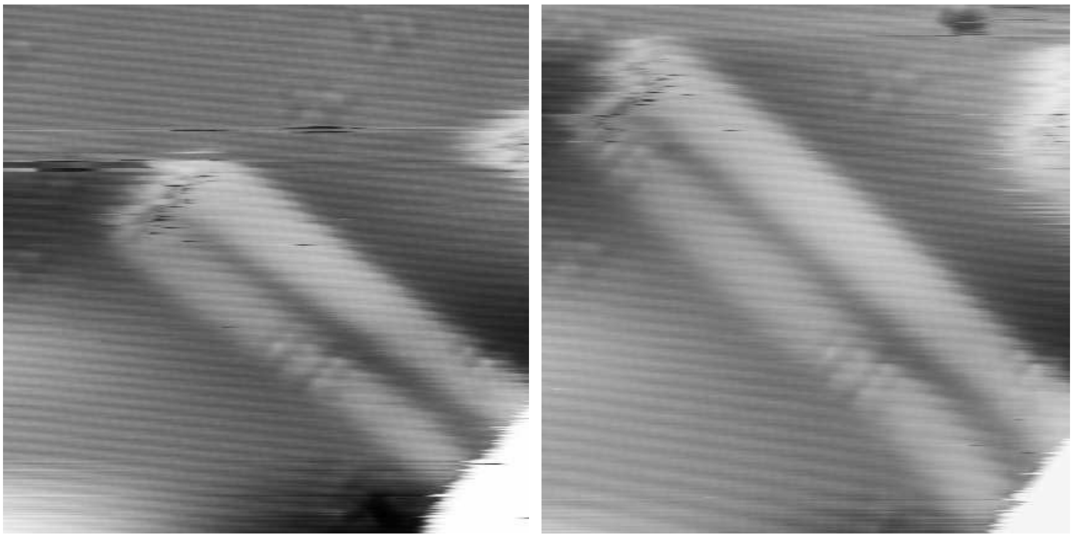

Fig. 4. Consecutive images showing the change in length (climb) of the dislocations. The image sizes are 10nm × 10nm. The time between images is 3 minutes.

The second type of motion (which we will refer to as "slow") is a displacement of a threading dislocation in the direction parallel to the axis of the dissociated misfit dislocation. This displacement implies that the length of the misfit dislocation must increase or decrease with time. This happens in a time scale of minutes for distances of a few angstroms. See Fig. 4 with the same dislocation observed after three minutes. Experimentally a random walk of the threading dislocation is observed[14].

The same dislocation can display both types of motion, producing a mass transport on the surface as observed in Fig. 5. The images shown are part of a movie, and they are separated by twenty minutes. The only way to go from the initial configuration to the final one is by climb *and* glide. Furthermore, unlike the random walk observed for single threading dislocations, in this case the total size of the trigon (the sum of the length of its dissociated misfit dislocations) is conserved within the experimental error.

The geometry of the dislocations needs to be considered to explain their motion. The Burgers vectors of the dislocations are shown in Fig. 2. The partial misfit dislocations Aδ, Bδ, or Cδ have their glide plane at the Cu/Ru interface (the d-plane). The threading dislocations have glide planes that intersect the d-plane. The intersection of their glide planes and the interface plane gives rise to glide lines. For example the dislocation BA attached to Bδ and δA can glide in the AB direction (as show in Fig. 2). The experimentally observed "fast"-motion directions are the glide directions just described. The speed of the motion is naturally explained by the conservative nature of the movement. Calculations which involve the detailed structure of the threading dislocations give estimates of tens of meV for the Peierls barrier for glide [13]. On the other hand, the "slow" motion of the threading dislocation is not in any glide plane. Thus, the motion requires the incorporation or extraction of Cu atoms from/to the Cu layer. In a bulk environment, the low density of vacancies or interstitials at room temperature would make such movement very slow. On the surface there is a supply and sink of Cu atoms in the form of a 2D adatom gas on top of the Cu layer. The motion of the threading dislocations corresponds experimentally to an average of one atom hopping in or out of the threading dislocation every 30s [14]. The density of the 2D gas of Cu atoms is much higher that bulk vacancies or interstitials, due to the lower barrier for atom detachment at surface steps.

A faster climb should be observed if the 2D adatom gas is increased (making the misfit dislocations grow) or decreased (tending to make the dislocations disappear). Experiments performed under exposure of S and O, which remove the Cu adatoms by reacting with them make the dislocations gradually disappear.

In the case of small isolated trigons (Fig. 5), we propose the following explanation for the constant total size of the trigon: the adatoms coming from one of the threading dislocations increase locally the adatom density. In turn this increases the probability that they get incorporated into any of the other two threading dislocations before moving away from the trigon. In this way, most of the time the trigon keeps a constant size, while moving around the surface. Since mass is transported across the surface during motion, it represents a (novel) surface diffusion mechanism.

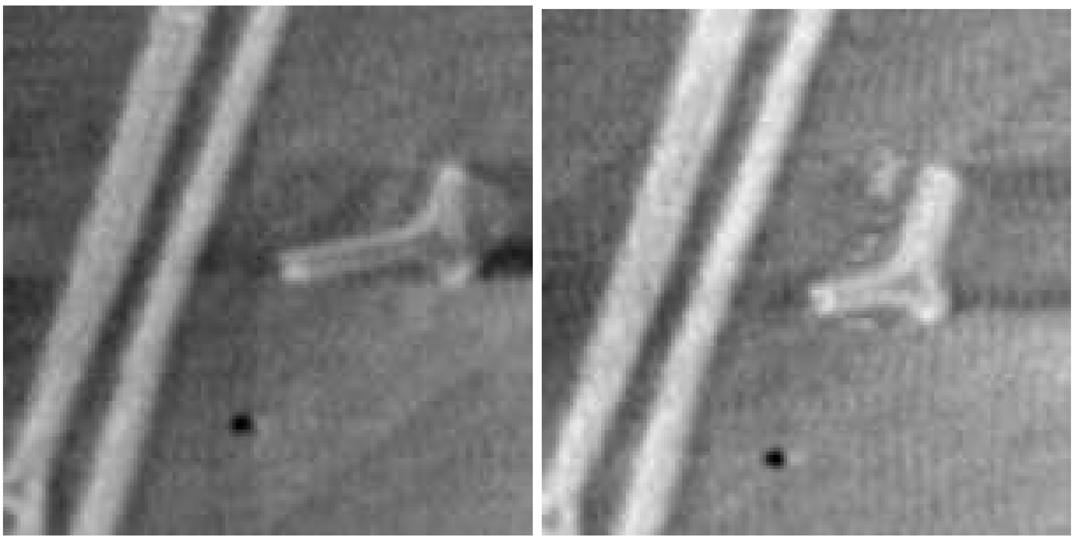

Fig. 5. STM images from a movie. A trigon which has undergone both climb and glide is shown in both images. The time interval between them is 20 minutes. The image size is 50nm$\times$50nm.

4 Conclusions

STM allows the observation of near surface dislocations with atomic accuracy. We have observed climb and glide of misfit and threading dislocations in 1ML Cu/Ru(0001) films. The combination of both climb and glide is a pathway for displacing mass parallel to the surface. Given the relatively high energy cost of forming adatoms and vacancies in close-packed metal surfaces, motion of these dislocations represent an important mode of surface diffusion on close-packed thin metal films.

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