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Study of ion bombardment induced vacancy islands on Au(100) by scanning tunneling microscopy

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The influence of the anisotropy of the $(5\times n)$ reconstructed (100) face of gold on the generation of vacancy islands by sputtering with 600 eV Ar^+ ions has been investigated by scanning tunneling microscopy. The shapes of these islands, bounded by monoatomic steps, are shown to be growth shapes, rather than equilibrium shapes. The growth mechanisms leading to these shapes are discussed.

I. INTRODUCTION

Low-energy (<1 keV) noble-gas ion sputtering is commonly used as a means to clean surfaces under ultrahigh vacuum (UHV) environment. Its use for modifying the surface microstructure in view of different applications, for instance, to modify the mode of epitaxial growth on the substrate¹ is also envisaged. It constitutes one of the reasons for the increasing interest in the understanding of the processes accompanying the interaction of these ions with surfaces. Furthermore, several recent investigations, using diffraction techniques² or scanning tunneling microscopy (STM)³⁻⁵ have shown that the abrasion process is closely analogous to epitaxial growth. The incident ions produce a supersaturation of mobile vacancies which results in the nucleation and growth of vacancy islands in the same way as adatom islands nucleate and grow from mobile adatoms in a growth experiment. Due to this similarity, the mechanisms revealed by these studies can be expected to apply to epitaxial growth.

This article extends a previous study of sputtering induced defects on Cu(100) by STM.⁶ In this work, monolayer deep vacancy as well as adatom islands were observed on the surface after an Ar^+ (600 eV) bombardment. The mobility on copper at room temperature is high enough for these islands to achieve their equilibrium shape, which can be described as a square with rounded corners, with the sides oriented along [011] equivalent crystallographic directions. We report in this paper, similar experiments on the (100) face of gold. Gold and copper are in many respects similar metals; they crystallize with the same structure and have similar melting points. Nevertheless, one striking difference is that low index faces of gold reconstruct. On gold (100), the reconstruction breaks the square symmetry of the (100) face, inducing an anisotropic structure at the surface. Our main motivation here is to investigate the influence of such an anisotropy on the formation of vacancy islands.

II. EXPERIMENT

The experiments were performed in an UHV chamber with an operating pressure in the low 10^{-10} Torr range. The tungsten tip was cleaned *in situ* by heating to about 1000 °C, prior to the measurements. The surface of the sample was oriented better than half a degree from the (100) plane. It was

cleaned by repeated cycles of Ar^+ sputtering at 700 K followed by annealing at 750 K. This cleaning stage was followed by a bombardment with 600 eV Ar^+ at room temperature controlling the time of exposure.⁶ The flux was 10^{13} ions $\text{s}^{-1} \text{cm}^{-2}$ for all the experiments discussed in this work. The images presented here were recorded in the constant current mode at room temperature with tunneling currents around 400 pA and biases of the order of 100 mV.

III. RESULTS AND DISCUSSION

We will first report on the interplay between the orientations of the reconstruction domains and the residual steps. Several conclusions in this part will be useful to understand the observed shapes of the islands which will be discussed in a second part.

A. The reconstruction and its orientation relative to steps

The Au(100) reconstruction has been investigated by a great number of different techniques,⁷⁻⁹ including STM.¹⁰ The following conclusions are now widely accepted: the reconstructed (100) surface can be described, in a first approximation, as a pseudohexagonal compact layer adsorbed on a square substrate. The interatomic distances in this layer are 4% shorter than bulk ones. X-ray diffraction experiments have shown that the dense rows of the surface layer make an angle of the order of 1° with the dense rows of the substrate.⁷ This leads to a nearly $(5\times n)$ unit cell, where the periodicity n , which seems to be very sensitive to defects of all kinds⁸ varies from 15 to 28. For brevity, we will, respectively, call the “ L ” and “ S ” directions the large and short period directions of the reconstruction.

The short period of the reconstruction appears clearly on the upper part of Fig. 1, while the longer one is not resolved.¹¹ The corrugation along this short period varies between 0.02 and 0.1 nm, depending on tip condition. Monoatomic steps which are observed are most often oriented near one of two orthogonal directions. One remarkable observation is that the “ L ” direction of the reconstruction is aligned on these steps. Knowing the model for the reconstruction,⁷ this identifies the two directions taken by the steps as [011] and [0-11]. We occasionally observe step bunches, like in the bottom-right side of Fig. 3, for which the preceding observation does not hold. Presumably, these macrosteps permit to



FIG. 1. 265 Å × 300 Å image of the Au(100) surface prepared by ion bombardment (Ar^+ , 600 eV, 10^{13} ions $\text{cm}^{-2} \text{s}^{-1}$, 40 s). A few vacancy islands slightly disturb the reconstruction which appears on the upper part of the image. Monoatomic kinked steps can be observed on the lower part. The "S" and "L" directions of the reconstruction are indicated.

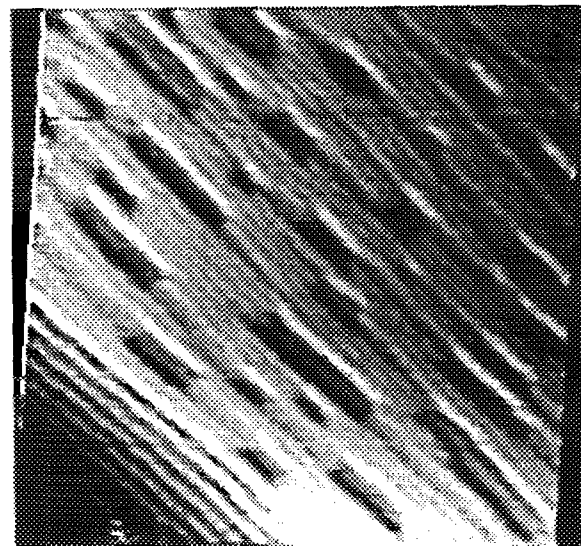
restore the macroscopic orientation of the surface, which probably cannot be maintained only by monoatomic steps in the two directions mentioned above. These observations indicate that domains with their "L" direction oriented along the monoatomic steps are favored.

On stepped parts of the surface like on the lower part of Fig. 1, we observe terrace widths taking discrete values, corresponding to an integral number of "S" periods plus about two interatomic distances and periodically arranged kinks. A model relating these observations to the rotation of the surface layer relative to the substrate,⁷ will be published elsewhere.¹²

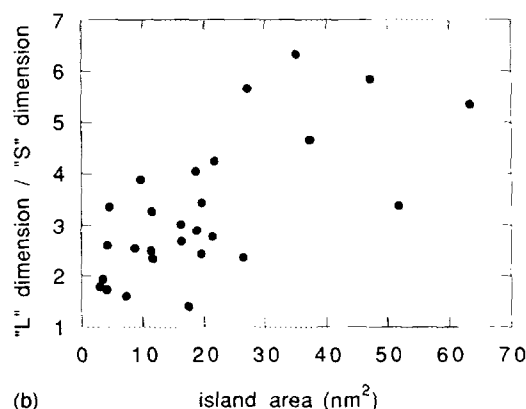
B. Shape of the islands

Vacancy islands appear in the images of Figs. 1, 2, and 3 as dark, one monolayer deep rectangular areas. They are aligned along the "L" direction. The corners of these depressions are much better defined than on Cu(100). These shapes clearly reflect the strong surface anisotropy induced by the reconstruction.

Another observation is that the islands perturb the stripped pattern of the reconstruction. The stripes are no more strictly parallel and can be curved locally. This is not surprising as the orientation of the pseudohexagonal surface layer relative to lower layers has been shown to vary with local stress fields, especially near atomic steps,⁹ such as those bordering the islands. The reconstruction stripes which have been observed by STM on the (111) face of gold are, in a similar manner, very easily perturbed by defects.¹³



(a)



(b)

FIG. 2. (a) 590 Å × 680 Å image of the Au(100) surface prepared by ion bombardment (Ar^+ , 600 eV, 10^{13} ions $\text{cm}^{-2} \text{s}^{-1}$, 120 s). Vacancy islands appear as darker rectangular areas. Steps occupies the lower-left-hand corner of the image. (b) Ratio of the "L" over the "S" dimensions as a function of the area for the islands of the image.

These observations raise the question of the origin of the shape of these structures. It seems clear that it is not the equilibrium shape. In fact, one expects an anisotropic equilibrium shape, as a consequence of the anisotropic structure of the surface, but this shape should be the same for all islands. This is not the case: indeed, the aspect ratio (ratio of the lengths of the long side to the short side) increases with the area, as shown in Fig. 2(b). This demonstrates that we are observing growth shapes, and that the islands grow mainly by their short-side ends. Annealing experiments, where a sample with a vacancy coverage similar to what is observed in Fig. 2(a) was maintained at 50 °C for 30 min, lead to a similar conclusion. The image of Fig. 3, obtained in these conditions, shows that the shapes of the islands are now characterized by a lower aspect ratio. This can be understood if one admits that during the annealing process islands tend toward their more compact equilibrium shape.

Three possible causes can be invoked to explain this growth by the short side ends. It can result from an anisotropy in (i) diffusion, (ii) sticking coefficient, or (iii) what we call "incorporation." (i) A strong anisotropic diffusion would

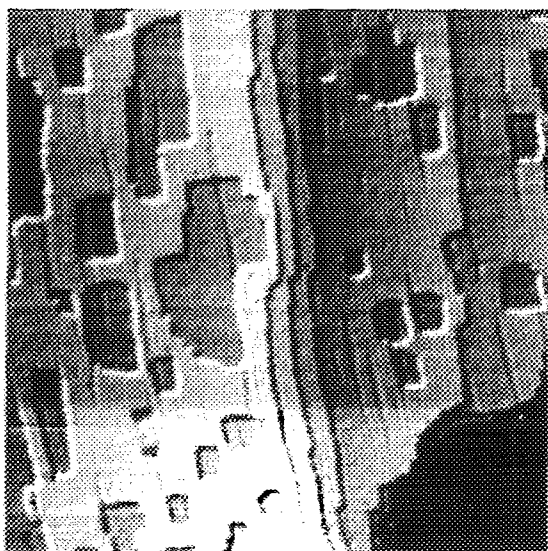


FIG. 3. $1200 \text{ \AA} \times 1300 \text{ \AA}$ image of the Au(100) surface prepared by ion bombardment (Ar^+ , 600 eV, $10^{13} \text{ ions cm}^{-2} \text{ s}^{-1}$, 60 s) and annealed at 50°C during 30 min.

lead to an anisotropy in the spatial distribution of the islands. A careful examination of our images shows that their anisotropic appearance lies mainly in the shape of the islands, but not in their spatial distribution on the surface. Anisotropic diffusion can then be excluded as the dominant contribution to the observed shapes. (ii) The probability of capture of a vacancy by an island certainly depends on the side on which it impinges. But on a metal, one does not expect a sticking coefficient much lower than unity, as it would be necessary on the long side of the islands to account for our observations. (iii) The remaining possibility is similar to "faceted growth," which happens frequently for three-dimensional crystals. A facet of a crystal, at a temperature below its roughening transition, generally grows more slowly than the neighboring high index planes. The reason is that the growth of such a facet requires the nucleation of a two-dimensional nucleus on the surface while sites of incorporation of atoms are already available on the steps of the high index faces. The situation here is closely analogous, the long and short sides of the islands corresponding to the facets and the high index faces, respectively. As previously mentioned, only widely spaced geometrical kinks are seen on the steps which are oriented along the "L" direction. The long sides of the islands are such steps, and thus do not provide sites of easy incorporation for a vacancy, such as kink sites. An island can grow in the "L" direction only by nucleating a one-dimensional vacancy nucleus on one of its long sides. Most of the time, a vacancy sticking the long side will diffuse along the step edge to incorporate on the short side, where kink sites are available.

These shapes are affected by the reconstruction in another way. The uncovered surface which constitutes the bottom of the islands is observed to be reconstructed and the widths of the islands are generally an integer number of "S" periods. So, the distance between steps of a different sign takes dis-

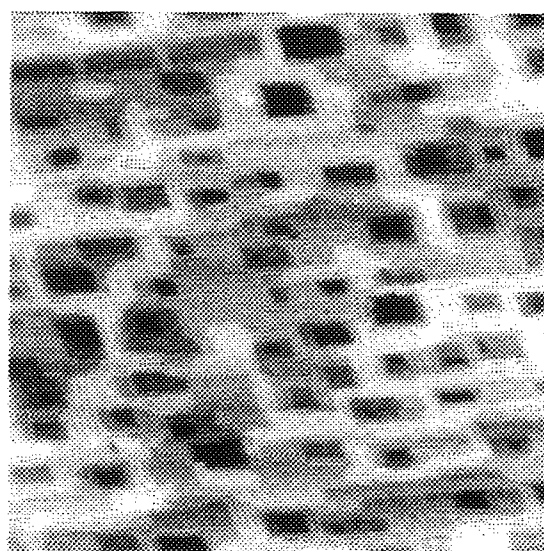


FIG. 4. $1200 \text{ \AA} \times 1300 \text{ \AA}$ image of the Au(100) surface prepared by ion bombardment (Ar^+ , 600 eV, $10^{13} \text{ ions cm}^{-2} \text{ s}^{-1}$, 1080 s). For higher fluences, small domains of the reconstruction oriented perpendicularly to the reconstruction of the surrounding terrace appear at the border of the islands.

crete values as on vicinal surfaces for steps of the same sign, as mentioned above.

A new phenomenon appears when the coverage in vacancies increases. Small domains of the reconstruction oriented perpendicularly to the reconstruction of the surrounding terrace are observed at the border of some islands. It seems that the steps which develop in the "S" direction as the width of the island increases forces the reconstruction to take the favorable orientation. This shows that the energy gained in this process is larger than that needed to create a domain boundary. This affects the shape of the island which now tends towards a square. At the highest investigated fluences, most of the islands exhibit square shapes, as illustrated in Fig. 4.

IV. CONCLUSION

Ion bombardment (Ar^+ , 600 eV) on Au(100) generates vacancy islands whose shapes are strongly influenced by the anisotropy induced by the reconstruction of this face. This shows that the reconstruction of a surface can play an essential role in determining its structure in the first stages of ionic abrasion. This conclusion is likely to apply more generally in the case of epitaxial growth.

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