PERSISTENT LAYER–BY–LAYER SPUTTERING OF Au(111)

M.V. Ramana Murty[(a),*], A.J. Couture[(a)], B.H. Cooper[(a),†], A.R. Woll[(b)], J.D. Brock[(b)], R.L. Headrick[(c)]

(a) Department of Physics and Cornell Center for Materials Research, Cornell University, Ithaca, NY 14853.

(b) School of Applied and Engineering Physics and Cornell Center for Materials Research, Cornell University, Ithaca, NY 14853.

(c) Cornell High Energy Synchrotron Source, Wilson Laboratory, Cornell University, Ithaca, NY 14853.

Abstract

Persistent layer-by-layer removal of Au(111) during Ar^+ ion irradiation was observed in a real-time X-ray scattering study. Over 100 specular beam intensity oscillations were measured. For a given ion energy, a smoother surface morphology is obtained when the ion flux is reduced. For a fixed erosion rate, ion energy in the range 70 – 500 eV does not have a strong influence on the evolution of surface morphology. Diffuse scattering measurements show the development of features with a characteristic lateral length scale on the surface during ion irradiation.

* Current Address: Argonne National Laboratory, Materials Science Division, IL 60439
Email: murty@anl.gov

† Barbara Cooper passed away prior to the submission of this paper. She was instrumental in setting up this collaboration to study low energy ion irradiation effects on metals.

Ion beam etching is widely used for patterning wafers during device fabrication, thin film composition analysis, and specimen preparation for electron microscopy. Sputtered surfaces are often rough exhibiting a pattern of ripples [1, 2] or mounds and pits [3, 4, 5], with lattice damage setting in at high incident ion energies [6]. Maintaining a smooth surface during sputtering can have several benefits. For example, a smooth surface morphology can improve depth resolution in secondary ion mass spectrometry [7] and reduce the amount of overetching that may occur while removing the top layer from a stack of layers on a substrate. Layer-by-layer removal allows precise control on the number of monolayers removed [8].

In a previous paper [5], we had reported the different regimes of surface morphology evolution on Au(111) during 500 eV Ar^+ ion irradiation. In this Letter, we report the influence of Ar^+ ion flux and ion energy on the evolution of surface morphology on Au(111) investigated using real–time X-ray scattering. Under appropriately chosen ion irradiation conditions, we observe persistent layer-by-layer sputtering. Diffuse scattering measurements show the development of approximately regularly arranged features on the surface during sputtering.

X-ray scattering experiments were carried out with 11 keV X-rays on the A2 station at the Cornell High Energy Synchrotron Source (CHESS). The surface of single crystal Au(111), with a miscut of less than 0.1°, was prepared by repeated sputtering and annealing at 450°C. The smooth starting surface showed the herringbone reconstruction characteristic of Au(111) [9, 10]. The base pressure in the ultrahigh vacuum (UHV) chamber was 5×10^{-10} mbar. An RF ion source was used for Ar^+ ion irradiation. In the experiments described below, the ions were incident at 2 – 7° with respect to the surface normal. The background pressure of Ar during ion irradiation was 3×10^{-4} mbar. The details of the X-ray scattering geometry and the UHV chamber are discussed elsewhere [11, 12]. In this paper, we show measurements of the specular reflectivity, which gives information about the surface roughness, and diffuse scattering, which gives information about lateral spatial correlations in the surface morphology.

Figure 1 shows the specular beam intensity at the $(0\ 0\ 1.5)_{\mathrm{hex}}$ position ($\equiv (0.5\ 0.5\ 0.5)_{\mathrm{cubic}}$) during 70 eV Ar^+ ion irradiation at 230°C [13]. In this geometry, the scattered waves from two successive (111) planes are 180° out of phase, thereby maximizing the sensitivity to surface roughness. Over 100 specular beam intensity oscillations were observed. Such persistent layer-by-layer removal was observed for ion energies in the range of 70 – 200 eV. Layer-by-layer sputtering has been reported previously [5, 14, 15, 16], but fewer than ten oscillations were observed in all cases. Extended (> 100) oscillations have been observed before, but only in the more commonly studied case of growth, rather than ion erosion, and only for a small handful of systems [8, 17, 18].

The erosion rate depends on both the ion flux and the ion energy. The evolution of the specular beam intensity for 200 eV Ar^+ ions at 205°C and different ion fluxes is shown in Figure 2. A lower ion flux leads to a smoother surface morphology, as indicated by the higher number of intensity oscillations. For a fixed temperature, a similar dependence on the ion flux was observed for ion energies in the range of 50 – 300 eV.

Figure 3 shows the specular beam intensity evolution for three different ion energies at 230°C. The ion flux was adjusted at each energy to achieve a fixed removal rate of 0.6 ML/s. It appears that in the 70 – 500 eV energy range, the ion energy does not have a strong influence on the specular beam intensity evolution. Of course, ion beam damage is expected to influence the surface morphology at higher (keV) energies.

Specular beam intensity oscillations occur when each monolayer is removed in sequence through the nucleation, growth, and coalescence of two-dimensional vacancy islands. The evolution of surface morphology during ion erosion mirrors that commonly observed in molecular beam epitaxy (MBE), suggesting that surface vacancies play a role analogous to that of adatoms during MBE. The smoother surface morphology obtained by lowering the ion flux is attributed to the increasing diffusion length of the surface defects (vacancies) with decreasing ion flux. However, compared to MBE, there are many more microscopic events during ion bombardment. This includes sub-surface vacancy creation and adatom-vacancy pair creation at the surface, both likely to increase with increasing energy [19]. How does this effect the simple scenario for the surface morphology evolution above? Since adatom diffusion is generally faster than vacancy diffusion, it is likely that adatoms created by ion bombardment quickly annihilate with the surface vacancies. At the temperatures investigated here, In a similar fashion, the vacancies created in the second layer and below can annihilate by atom detachment and diffusion from the upper layers. This leaves only the slower transport component, namely, the surface vacancy kinetics determining the evolution of surface morphology.

While the oscillations in Figs. 1–3 indicated persistent layer-by-layer removal, they eventually decay indicating that the surface roughness is increasing with time. Measurement of the specular beam intensity I close to a Bragg position on the truncation rod gives information about the evolution of surface roughness. The rms roughness w is obtained from $I \approx I_0 \exp(-q_z^2 w^2)$, where I_0 is the intensity from the smooth starting surface. Figure 4(a) shows the evolution of w during 200 eV ion irradiation for two different irradiation conditions and temperatures. The specular beam intensity was measured at $q_z = 0.243$ Å^{-1} at 180°C and $q_z = 0.291$ Å^{-1} at 205°C.

Diffuse scattering measurements, involving transverse scans through the $(0\ 0)_{\rm hex}$ truncation rod, give information about lateral correlations in the surface morphology. Figure 4(b) shows transverse scan in two orthogonal directions through the anti-Bragg position $(0\ 0\ 1.5)_{\rm hex}$ taken during 800 eV Ar^+ ion irradiation. The satellite peaks indicate the development of features with a well-defined lateral length scale. Satellite peaks were observed in such transverse scans for all ion energies (50 – 800 eV) and fluxes considered in this study. The average separation between features l can be estimated from $l \approx 4\pi/\Delta q_{||}$, where $\Delta q_{||}$ is the separation between satellite peaks. The length l is estimated as 85 nm under the conditions indicated in Fig. 4(b), being approximately the same in the two orthogonal directions. Such pattern formation was also observed during ion irradiation at low temperatures in the three-dimensional rough erosion regime [5]. Possible origins of such pattern formation include an Ehrlich-Schwoebel barrier for surface vacancies [20], step-vacancy attraction [21], and step edge diffusion of vacancies [22].

In conclusion, we have observed persistent layer-by-layer removal of Au(111) under 70 – 200 eV Ar^+ ion irradiation at 230°C. A smoother surface morphology results from the use of lower ion fluxes. Continued removal of materal results in increasing surface roughness and the development of features with a characteristic lateral length scale.

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Figure Captions

Figure 1. Specular beam intensity at the $(0\ 0\ 1.5)_{\mathrm{hex}}$ position during 70 eV Ar^+ ion bombardment of Au(111) at 230°C. Over 100 intensity oscillations were observed.

Figure 2. Influence of ion flux on the specular beam intensity evolution. The ion energy was 200 eV and the substrate temperature was 205°C.

Figure 3. Specular beam intensity evolution at three different ion energies. The ion flux was adjusted at each energy for a sputter rate of 0.6 ML/s. The substrate temperature was 230°C.

Figure 4. (a) Increase in the rms roughness w during etching with 200 eV Ar^+ ions. (b) Transverse X-ray scans in two orthogonal directions through the $(0\ 0\ 1.5)_{\mathrm{hex}}$ position taken during 800 eV Ar^+ ion irradiation. The characteristic length scale l is estimated as 85 nm.