SPUTTERING FROM ENERGETIC SELF-ION CASCADES

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SPUTTERING FROM ENERGETIC SELF-ION CASCADES*

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Abstract

The rather high primary recoil energies that occur under 14-MeV neutron irradiation can be simulated by self-ion bombardment. However, the primary ion always starts at the entrance surface when self-ion bombardment is used. To examine the effects of cascades in which the momentum of the primary atom is directed towards an exit surface, one has to introduce a second surface that will intersect the cascade in the forward direction. Self-ion transmission sputtering on very thin films has been used for this purpose. The analysis was done in situ by He^+ backscattering. A backspattering ratio of 120 was observed at 540 keV. At the same energy a forward sputtering ratio of 70 is obtained for a film of $660 \text{ \AA}$ in thickness. These ratios are in order of magnitude agreement with theoretical estimates, however, the results seem to indicate that collective and lattice effects play a significant role. It was observed that a tensile stress is built up in the films during irradiation, leading to flattening and rupturing along preexisting defects.

Introduction

The production of monoenergetic displacement cascades by self-ion bombardment has been used by a number of investigators to study defect structures and sputtering in metals. Defect structures have been examined using various

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forms of ion and electron microscopy(1-3) while sputtering information has been obtained from ejection patterns and material loss measurement.(4-6)

A correlation between effects from neutron induced cascades, generated by a primary knock on, with cascades from self ion irradiations, which simulate the primary knock on condition, has long been of interest. This interest arises from the fact that ion bombardment techniques afford a simple, efficient, and accurate method for producing energetic cascades in solids. Thus, ion irradiations represent a useful alternative to, and a complementary check on, cascade phenomena associated with high energy neutron collisions. For example a recent comparison of transmission electron microscopy (TEM) data obtained from self ion bombardments with data resulting from 14-MeV neutron irradiation has demonstrated good quantitative agreement.(7)

Recently sputtering from 14-MeV neutrons has received considerable attention.(8-11) In a number of such experiments the total sputtering yield was found to be significantly greater than that predicted by theory. In addition to single atom sputtering and the emission of small clusters of atoms, ejection of macroscopic particles has also been reported.(8)

Since most of the displacements produced by 14-MeV neutrons occur in very energetic cascades (~ 100 keV), it is of crucial importance to know the sputtering mechanisms for such cascades when attempting to understand high energy neutron ejection.

The purpose of the present investigation was to determine whether those aspects of 14-MeV neutron sputtering relating to the ejection from energetic displacement cascades could be studied by means of self ion bombardment. Of particular interest in this connection are those cascades that lead to very high sputtering ratios. In making these comparisons an important difference between the 14-MeV neutron recoil cascades and self ion cascades is that self ion cascades always start at the entrance surface. Recoil cascades,

however, will be initiated with equal probability at any depth from the surface and, most importantly, the recoil momentum of the primary particle can also be directed towards a surface. In the case of the self-ion cascade, the primary ion is initially always going into the entrance surface. Results from such experiments cannot, therefore, be directly related to neutron sputtering yields.

In the present experiment we were interested in duplicating the situation where a primary recoil initially moves towards a surface at a distance comparable to the mean damage depth. This can be done by self-ion bombardment of thin films where the transmission sputtering rate is measured from films whose thickness is comparable to the mean damage depth. As seen in Fig. 1, displacement cascades of some hundreds of keV split into subcascades. Each subcascade that intersects a surface can give rise to sputtering. Basically any atom that arrives at the surface with a kinetic energy greater than the surface binding energy will be sputtered. In a very energetic cascade more than one subcascade can be intersected by the surface. The number of sputtered atoms that are ejected from a subcascade will depend strongly on the position of the surface relative to the center of the subcascade. If the energy density in the core of a subcascade is very high, one can expect the influence of collective effects. As Anderson and Bay(12) have shown by their molecular ion experiment, an increase in the cascade density leads to a more than linear increase in the sputtering rate. In the case of very energetic cascades, where several subcascades are formed, there also is a finite probability of overlap between subcascades. An increase in the subcascade density results if two subcascades overlap, whereupon consequences similar to those observed in the Anderson-Bay experiment(12) can be expected. The core of a subcascade may have a very high displacement

density, in which case a large fraction of the atoms will be involved in the displacement process, allowing it perhaps to be described by a thermal spike model. In this case, a relatively large amount of material could be ejected if the core of the subcascade is intersected by an external surface.

Experimental

Specimen Preparation

Thin monocrystalline films of Au were grown on (001) cleavage surfaces of rocksalt at 350°C. During evaporation the ion-pumped evaporator was at a pressure of approximately 10^{-7} Torr. Single-crystalline films of (001) orientation were obtained in this way. Subsequently the films were annealed in air at 350°C for 30 minutes. Film thickness was determined by gravimetry. After dissolving the rocksalt in water the specimens were mounted on standard TEM copper specimen grids.

Transmission Sputtering

The grids were transferred to a UHV scattering chamber which was attached to a 300 kV heavy ion accelerator. As shown in Fig. 2, the film was mounted in an Al holder and the material sputtered in transmission was collected by an Al catcher. Several Al collectors could be sequentially rotated into position. The specimen holder could also be moved out of the path of the beam so that the Al collector could be examined in situ by He^+ backscattering. The self ion transmission sputtering was performed with a doubly charged Au^{++} beam of 540 keV. This energy, although nearly twice the maximum for elastic 14-MeV neutron collisions in Au, was used because thinner specimens were not available and because the same basic ejection mechanisms are expected in the region of subcascade formation. The Au-beam was 3.2 mm in diameter at the specimen and was swept in two dimensions to achieve uniformity of irradiation. The dose measurement was

obtained by means of a Faraday cage that intercepted an annular fraction of the beam in front of the target. During irradiation the pressure in the target chamber was on the order of 5×10^{-8} Torr.

Backscattering

The film, as well as the collectors were analyzed in situ by means of 270 keV He^+ Rutherford backscattering at $\theta = 150^\circ$. One can determine, by this method, the loss of material from the film(13) and also the amount of material that has accumulated on the collectors. These data result, therefore, in a direct measure of both the total yield (from specimen thickness reduction) and the forward yield, as observed on the collectors.

When analysing the film a beam of 1 mm in diameter was used while a swept 3.2 mm diameter beam was used to observe the sputtered Au on the collector. A laser beam whose path coincided with the ion beam greatly facilitated line up of target and collector. Figure 3 shows a composite of several backscattering spectra. The channel numbers given on the abscissa can, in the usual manner, be translated into an energy or depth scale. The solid line represents the scattered intensity from Au that has been sputtered and collected on the Al catcher. The open points indicate the background from the Al substrate. The position of the Au-peak coincides with the position at half maximum of the front edge of the spectrum from a gold film. The width at half maximum of the sputtered gold peak also coincides with the width of the edge of the gold film. This indicates that the sputtered Au is located directly at the surface of the Al catcher. If a significant fraction of the gold would have been implanted to a finite depth in the Al or if particles of a finite thickness, comparable to the film thickness would have been emitted, the sputtered Au spectrum would extend to lower energies.

For calibration purposes a 50 keV Au^+ implant was made into one of the Al collectors. The backscattering spectrum from this implant is also shown in Fig. 3. It can be seen that the most probable position of the implanted Au is at some finite depth in the Al. Range straggling is also seen from the increase in width of the spectrum. The calibration factor for the amount of Au present was derived from the integrated number of counts in each peak and from the known implant dose. This calibration factor was in good agreement with the one derived from the Rutherford backscattering cross section and the scattering geometry.

Results

Transmission Yields

The self ion transmission sputtering yield as a function of fluence is plotted in Fig. 4 for an Au film of 656 Å thickness. In determining the yield from the amount of Au present on the Al collector within the 3.2 mm diameter area sampled by the He^+ beam, we have assumed that the amount of sputtered gold is proportional to $\cos \psi$, where ψ is the angle to the surface normal. A sticking coefficient of 1 for Au on Al has also been assumed. It can be seen from Fig. 4 that the transmission yield increases linearly with dose. The straight line in Fig. 4 corresponds to 69 atoms sputtered in transmission for each incoming ion.

Effects of Radiation on Film Structure

Similar to the backspattering thickness measurement of film on a substrate⁽¹³⁾ one would expect to be able to determine the total sputtering (forward and backward) from examining the Rutherford backscattering spectra of non-backed films. For uniform sputtering one expects a reduction in the width of the film spectrum with increasing Au^{++} dose. In contrast to this we see in Fig. 5, after an incubation period corresponding to a dose of approximately $4 \times 10^{13} \text{ Au}^{++}/\text{cm}^2$,

a reduction in the height and also a widening rather than a decrease in width of the spectrum. A decrease in height is expected if holes are formed in the film. Figure 6 shows what actually has happened to the film. Rather large holes have appeared and in some instances the film has partially folded over. The latter would account for the apparent increase in width of the spectrum. The most striking feature is that the film has otherwise become completely flat during the self ion irradiation. The upper right hand corner of the film has not been irradiated. Apparently during irradiation a tensile stress has built up in the film straightening out the wrinkles and finally tearing the film apart. TEM observation clearly indicates that mechanical deformation is involved in forming the gaps. It can also be seen by TEM that the tears start at some weak points in the film. These weak points are mostly linear regions, characterized by a reduced film thickness and the presence of small holes. Those linear defects are introduced during evaporation of the films and come from the effects of cleavage steps or scratches in the rocksalt substrate. It is possible that the tears could be avoided if these preexisting defects could be eliminated.

Backward Sputtering Yield

To determine the backward sputtering yield we sputtered a $1200 \text{ \AA}$ thick film that was still attached to the rocksalt substrate. In this case the film did not form large holes as in the unsupported film. The reduction in thickness is clearly seen in the He-backscattering spectra after various irradiation doses as shown in Fig. 7. The backward sputtering ratio derived from this reduction in thickness is $S \sim 120$.

Comparison with Theory

The theory of sputtering from amorphous targets has been given comprehensive treatment by P. Sigmund.(14) The theory agrees rather well with a

number of sputtering measurements on polycrystalline targets(12,14) and it is also expected to give a good indication of the total sputtering yield for a beam incident under random direction on a single crystalline target. According to ref (14) the sputtering yield S as a function of energy E is given by

$$S(E) = \Lambda F(E)$$

where under the present conditions $\Lambda = 0.187 \text{ \AA}^{-2}$. In first approximation the function $F(E)$ is given by

$$F(E) = \frac{v(E)}{\langle \Delta x^2 \rangle^{1/2}} \frac{1}{\sqrt{2\pi}} e^{-\xi^2/2}$$

with $\xi = -\langle x \rangle / \langle \Delta x^2 \rangle^{1/2}$ for backward sputtering and $\xi = (d - \langle x \rangle) / \langle \Delta x^2 \rangle^{1/2}$ for transmission sputtering. Here $v(E)$ is the energy that is dissipated in nuclear collisions, d is the film thickness and $\langle x \rangle$ and $\langle \Delta x^2 \rangle$ are moments over the deposited energies given in ref (15) for various power law cross sections. With $m = 1/2$ (see ref.15) we get $\langle x \rangle = 378 \text{ \AA}$ and $\langle \Delta x^2 \rangle^{1/2} = 233 \text{ \AA}$. If we assume $v(E) = E$ we find $S_b = 50$ and $S_t = 85$ for the backward and transmission sputtering ratio respectively. Taking into account that about 1/3 of the energy is lost in electronic excitations(16) the above yields are reduced to $S_b = 33$ and $S_t = 56$. Sigmund et al.(14) have also considered the influence of electronic stopping on the moments $\langle x \rangle$ and $\langle \Delta x^2 \rangle$. For this case one gets $\langle x \rangle = 273 \text{ \AA}$ and $\langle \Delta x^2 \rangle^{1/2} = 150 \text{ \AA}$ which gives $S_b = 32$ and $S_t = 6.5$ for the backward and forward sputtering ratios respectively.

In comparing these theoretical values with our experimental results we find that the observed back sputtering ratio is a factor of 2 to 4 higher than predicted by theory. Similar increases above the theoretical yields have previously been found for Ag(12). The values for the transmission ratios are in rather good agreement with the experiment for the case where the influence of electronic losses on the deposited energy distribution is neglected. The rather low yield $S_t = 6.5$ which is obtained when electronic losses are taken

into account illustrates the strong dependence of the transmission yield on average damage depth or film thickness. TEM work has indicated that the size of the cascade is larger than indicated by the random theory.(18) This increased size is probably due to channeling effects. Larger yields at the present thickness and a smaller thickness dependence of the transmission sputtering yields would be expected from channeling effects.

Conclusions

1. The observed backspattering ratios from energetic self-ion cascades are somewhat higher than predicted by the random cascade theory. However, the deviations are within a factor of 4 of the theoretical value and are probably due to collective effects.
2. Transmission sputtering yields are in good agreement with the simple theory, however, thickness dependence measurements are essential for a more quantitative comparison with theory.
3. Tensile stresses in the films are built up during self ion irradiation, leading to film flattening and rupture along preexisting defects. The loss of interstitials from the interior of the film due to diffusion to the surface and sputtering, together with the spontaneous formation of vacancy clusters in energetic cascades causes internal stresses that lead to this effect. This "stretching effect" is opposite to the "dimpling" effect that has been observed in Si under He bombardment.(19)
4. The ejection of macroscopic particles has not been observed in the present experiment. The total material loss of the target as estimated from the backscattering spectra is of the same order as the sum of the transmission and backscattering yields.
5. The method introduced here for measuring thin film transmission yields seems to be well suited to more detailed studies. Within certain limits it is possible to study the thickness and energy dependence of self-ion sputtering.

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Figure Captions

- Fig. 1. TEM images of 540 keV self-ion displacement cascades in Au, exhibiting formation of subcascades. Each group of spots was produced by a single ion. The black spots represent vacancy clusters that are formed in the depleted zone of subcascades.
- Fig. 2. Specimen geometry for thin film sputtering in forward and back directions.
- Fig. 3. Composite of information obtainable from backscattering measurements.
- Fig. 4. Transmission sputtering yields as a function of bombardment fluence.
- Fig. 5. He^+ backscattering spectra from an Au foil mounted on a Cu grid for various Au^{++} bombardment fluences.
- Fig. 6. Optical micrograph of Au film after irradiation to a total fluence of $2.8 \times 10^{14} \text{ Au}^{++}/\text{cm}^2$ at 540 keV. Magnification 70x.
- Fig. 7. He^+ backscattering spectra of Au film on NaCl substrate after various fluences of Au^{++} at 540 keV.

Fig. 1

TRANSMISSION SPUTTERING

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7