THE ANGULAR DISTRIBUTION OF ATOMS SPUTTERED FROM MONOCRYSTALLINE GOLD

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Summary

The angular distribution of atoms sputtered from metal single crystals has been used to study atom ejection processes. By analysing this distribution into the sum of a Gaussian and a cosine contribution it is shown that the isotropic background can be removed and considerably improved agreement obtained between theory and experiment. This is applied to the temperature variation of the width of Wehner spots. New experimental results are presented for gold sputtered by 10 keV argon ions over a wide range of temperatures.

I. INTRODUCTION

Sputtering, which is the ejection of atoms from a solid as a result of particle bombardment, was first observed by Grove (1852) over a century ago. Intensive study of the sputtering process has been carried out only in the last decade, since Wehner (1955) discovered the anisotropic sputtering of single crystals. He found that atoms are preferentially ejected in directions that correspond to low-index crystallographic directions. The collected sputtered material reveals discrete spots. This phenomenon was explained by Silsbee (1957) in terms of focused correlated collision sequences that can propagate along close-packed crystallographic directions. The intersection of such a focused collision sequence with the surface of the crystal may cause the ejection of the last atom of the sequence, in the direction of the sequence. This last atom carries off the residual energy of the focused collision chain. Much experimental work has since been done to investigate the importance of focused collision sequences on the sputtering process (e.g. Garber and Fedorenko 1964; Thompson 1964).

The angular distribution of sputtered material is an important parameter of the sputtering process not only for the development of a complete sputtering theory. Knowledge of the angular distribution is necessary in many cases for the evaluation of the sputtering yield (i.e. the number of sputtered atoms per incident particle). An analysis of angular distribution may also help to distinguish atoms ejected by different mechanisms, for instance, channelling, evaporation from thermal spikes, focused collisions, and defocused and random collision cascades.

The material sputtered from polycrystalline metals is isotropically distributed (Seeliger and Sommermeyer 1935; Wehner and Rosenberg 1960; Patterson and Tomlin 1962; Ramer *et al.* 1964), approximately obeying Knudsen's (1909) cosine law (there being a tendency for "over cosine" sputtering at high bombarding ion

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energies and "under cosine" sputtering for low ion energies). The angular distribution of ejected atoms as a result of monocrystalline sputtering is usually expressed in terms of the variation of sputtering intensity about the close-packed preferential ejection direction, i.e. the angular distribution for individual spots in the ejection pattern. Spot profiles have been obtained for a number of metals: copper (Molchanov, Tel'kovskii, and Shakh-Melikova 1962; Yurasova and Bukhanov 1962; Yurasova and Murinson 1962; Endzheets *et al.* 1963); gold (Nelson, Thompson, and Montgomery 1962); aluminium (Nelson and Thompson 1962); nickel (Endzheets *et al.* 1963); tungsten (Yurasova and Sirotenko 1962; Molchanov, Soshka, and Faruk 1963); zinc (Yurasova and Murinson 1962; Molchanov, Soshka, and Faruk 1963). These profiles have a similar shape, with the angular distribution approximately Gaussian in the region of the spot maximum (i.e. for small angles from the preferential ejection direction). However, at larger angles the distribution does not exhibit the cutoff of a Gaussian function, it being much smoother with an extended tail. The departure from Gaussian is more pronounced for high temperature sputtering and for centre spots (i.e. preferential ejection direction normal to crystal surface). Endzheets *et al.* (1963) have suggested that this is due to the influence of background sputtering.

The anisotropic preferential ejection is accompanied by an isotropic ejection which produces a continuous background. This background sputtering, which presumably results from ejection of atoms not in their normal lattice positions and evaporation from thermal spikes, is present to varying degrees depending on the metal species and bombardment conditions. The background is very intense for lead and hardly visible for aluminium (Perovic 1961). The background is more pronounced for high energy bombardment and high crystal temperatures; this is evidence for a thermal spike background.

We assume here that the "natural" angular distribution of ejected atoms, as a result of preferential focuson ejection, follows a Gaussian function. From this assumption we are able to explain some quantitative features of the effect of temperature on the angular distribution.

II. The Angular Distribution

Since the spot patterns are always accompanied by a background of isotropically ejected atoms, the experimentally measured spot distribution $I(\theta)$ will be the resultant distribution of a superposition of a background distribution $I^{\mathrm{b}}(\theta)$ and the natural spot distribution $I^{\mathrm{s}}(\theta)$; that is, $I(\theta) = I^{\mathrm{b}}(\theta)+I^{\mathrm{s}}(\theta)$, $I(\theta)$ being the sputtering intensity at angle θ to the preferential ejection direction. For the purpose of comparison of experiment with a focuson-ejection theory, it is, then, the natural spot distribution $I^{\mathrm{s}}(\theta)$ that must be taken as the experimental distribution, for this represents the anisotropic preferential ejection contribution.

We shall now consider the functions $I^{\mathrm{b}}(\theta)$ and $I^{\mathrm{s}}(\theta)$. Since the background is an isotropic ejection from a finite-sized directed source, we assume that it is distributed according to Knudsen's (1909) cosine law, that is,

$$I^{\mathrm{b}}(\theta) = I_0^{\mathrm{b}}\cos\theta.$$

This seems a reasonable assumption, since both polycrystalline sputtering and evaporation processes obey this law. Nelson's (1965) measurements for copper show that the distribution of copper in the background approximates that expected from a mechanism of evaporation. Further, our measurements for gold indicate that the background distribution is approximately cosine.

For $I^s(\theta)$, as already mentioned, we have assumed a Gaussian function of the form $I^s(\theta) = I_0^s \exp(-\theta^2/2\Psi^2)$. This has been chosen because it is a good approximation to the form of $I^s(\theta)$ experimentally determined by subtracting the appropriate cosine background distribution from the experimentally measured $I(\theta)$ for gold $\langle 110 \rangle$

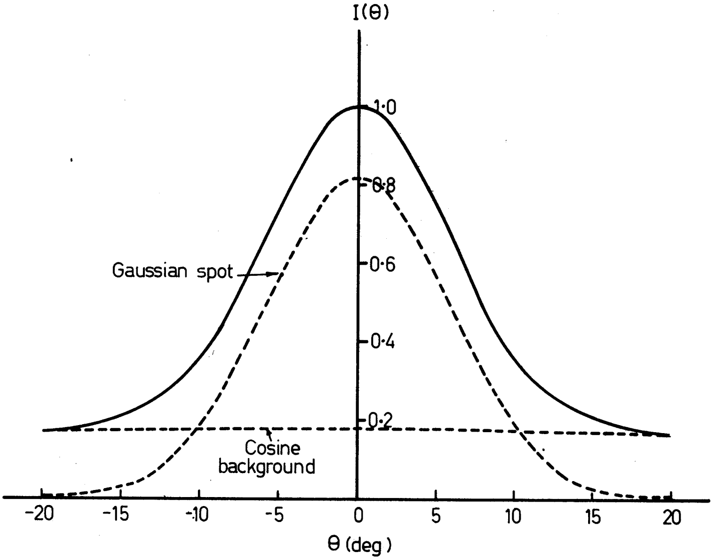

Fig. 1.—Angular distribution $I(\theta)$ for $\langle 110 \rangle$ gold spots represented by the superposition of a cosine background distribution and a Gaussian natural spot distribution (equation (1)).

spots. In other words, the experimentally measured spot angular distribution $I(\theta)$ is fitted well by the sum of a Gaussian and a cosine distribution (see Fig. 1)

$$I(\theta) = I_0^b \cos\theta + I_0^s \exp(-\theta^2/2\Psi^2). \tag{1}$$

Further evidence for support of a Gaussian distribution for $I^s(\theta)$ is that the experimentally measured spot distribution is approximately Gaussian for conditions under which the background is relatively small, namely, for small θ and low temperature sputtering. Under these conditions the first term in equation (1) is small compared with the second term.

Yurasova and Murinson (1962) have determined the following empirical expression for $I(\theta)$ from measurements of copper $\langle 110 \rangle$ and zinc $\langle 1120 \rangle$ spots:

$$I(\theta) = I_0 \cos\theta \exp(-p \sin^2\theta) \qquad \begin{cases} p = 3\cdot 3 \text{ for Cu} \\ p = 9\cdot 5 \text{ for Zn.} \end{cases}$$

This expression, as does equation (1), approaches Gaussian form for small θ; it is also well described by equation (1).

The constants in equation (1) (i.e. I_0^b, I_0^s, and Ψ) are adjusted to give the best fit to the experimentally measured spot distribution $I(\theta)$. I_0^b and I_0^s give an indication of the relative importance of background and preferential sputtering; Ψ^2 gives a measure of the mean squared angular deviation of ejected particles from the focusing axis. For large θ the cosine background term prevents the otherwise sharp cutoff of $I(\theta)$. The effect of background then is to broaden the spot width.

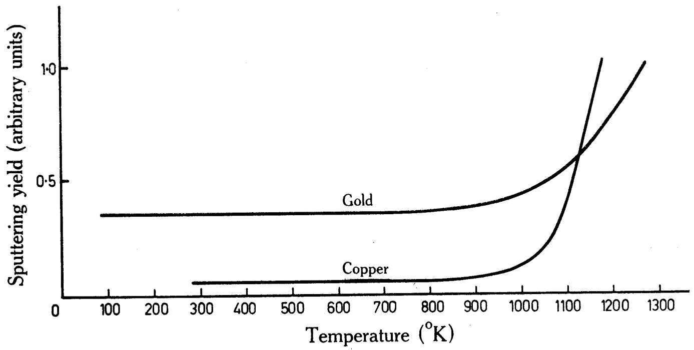

Fig. 2.—Sputtering yield for polycrystalline copper and gold as a function of target temperature (after Nelson 1965).

III. Effect of Crystal Temperature

The effect of crystal temperature on the angular distribution of sputtered material in a spot is particularly important to the testing of a preferential focuson-ejection theory (Nelson, Thompson, and Montgomery 1962; Sanders and Fluit 1964; Kurkin and Odintsov 1965). Thermal vibrations have a large influence on focused collision sequences, the most important mechanism being thermal scattering, which has been treated by Nelson, Thompson, and Montgomery. This effect renders focuson propagation inefficient and should be manifested experimentally as a reduction in the sputtering yield and a broadening of the ejection spots with increasing crystal temperature. This broadening has been reported for copper (Yurasova and Bukhanov 1962) and gold (Nelson, Thompson, and Montgomery 1962). However, to compare the broadening of the ejection spots with theory, the corrected angular distribution $I^s(\theta)$ must be used, since part of the broadening may be due to an increasing background.

Nelson (1965) has found that for copper the total quantity of material comprising the background increases with temperature in the same way as does the sputtering yield for polycrystalline copper (see Fig. 2). Thus, the background intensity is approximately constant up to 900°K and then shows an exponential increase for higher temperatures (this he attributes to evaporation from thermal spikes). Yurasova and Bukhanov (1962) have found that the copper ejection spots are especially well defined for temperatures below about 800°K. For higher temperatures the total background and spot width display a pronounced increase. This suggests that part of the observed increase in spot size could be due to an increasing background. If this same qualitative reasoning is applied to gold (Fig. 2) one would expect a substantial increase in spot width for temperatures above about 1000°K. This is in agreement with the measurements of Nelson, Thompson, and Montgomery (Fig. 3, curve (i)) of the mean squared angular deviation of ejected particles from the ⟨110⟩ axis. This curve shows a pronounced increase for temperatures above about 1000°K. Curve (ii) of Figure 3 shows the predictions of the thermal scattering theory (Nelson, Thompson, and Montgomery 1962). For low temperatures the discrepancy between theory and experiment is less. This could be due to a smaller background contribution, since the background intensity decreases with decreasing crystal temperature.

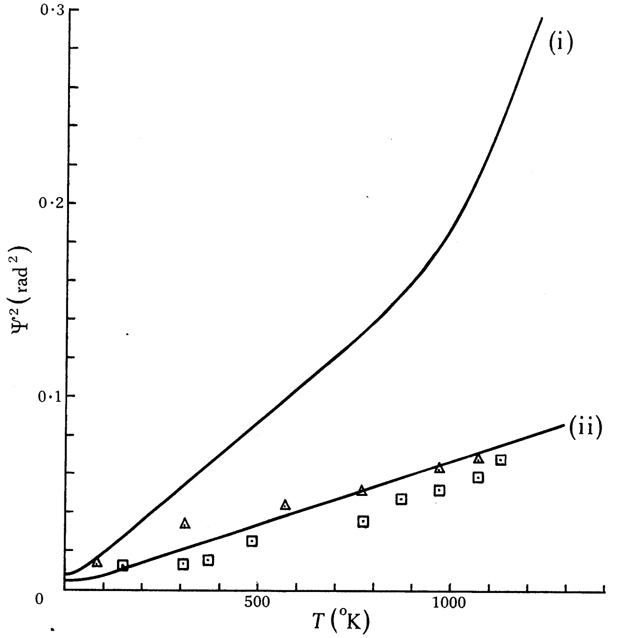

Fig. 3.—Temperature dependence of the mean squared angular deviation of ejected atoms from the ⟨110⟩ axis (for gold): (i) experimental results obtained by Nelson, Thompson, and Montgomery (1962); (ii) prediction of the thermal scattering theory due to Nelson, Thompson, and Montgomery. The points marked show the experimental results obtained after correcting for background sputtering, using our results (⊡) and those of Nelson, Thompson, and Montgomery (△).

The results of our measurements of the angular distribution $I(\theta)$ for ⟨110⟩ spots are presented in Figure 4. These were obtained by a method described elsewhere (Chapman and Kelly 1967). The sputtering was carried out by 10 keV argon ion bombardment of a [111] face of a gold crystal, maintained at temperatures between 100°K and 1100°K. The results have been analysed, by use of equation (1), to obtain the temperature dependence of Ψ^2 (Fig. 3, points enclosed in squares) and I_0^b/I_0 (Fig. 5). We note that I_0^b/I_0 shows a large increase for temperatures above 1000°K.

We have recalculated the experimental results of Nelson, Thompson, and Montgomery (which were obtained at 43 keV) allowing for the background. These corrected results (Fig. 3, points enclosed in triangles) are now much closer to the theoretical curve, lying slightly above our results. Both sets of experimental results are in good agreement with theory. It seems reasonable to conclude that the observed temperature broadening of the ejection spots is to a large extent due to the influence of background sputtering.

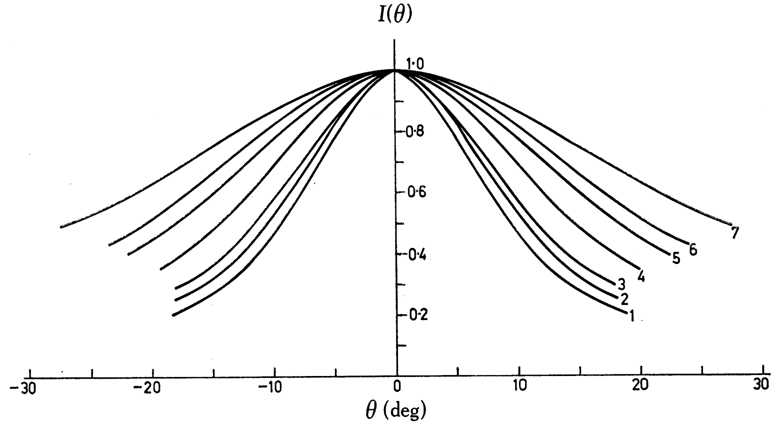

Fig. 4.—Angular distribution for ⟨110⟩ gold spots for various target temperatures. 1: 153°K; 2: 308°K; 3: 376°K; 4: 484°K; 5: 774°K; 6: 977°K; 7: 1077°K.

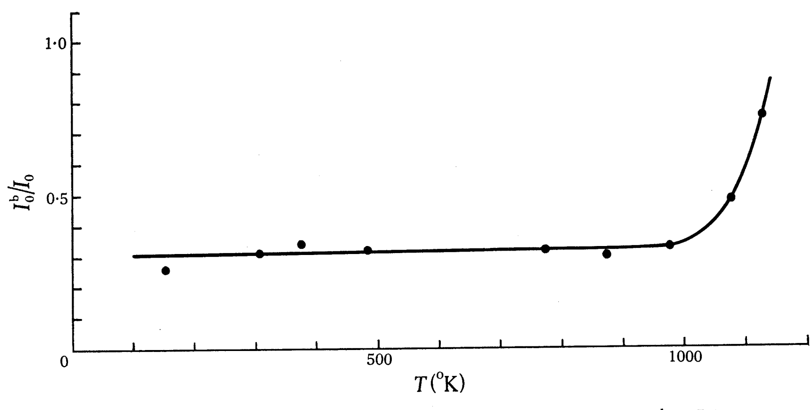

Fig. 5.—I_0^b/I_0 as a function of target temperature ($I_0 = I_0^b + I_0^s$).

IV. Discussion

The relative intensity of background sputtering is dependent on target temperature and also on projectile energy. Perovic (1961) reports that ejection spots are accompanied by a strong background when copper is sputtered with 1·2 MeV argon ions. The ⟨110⟩ and ⟨100⟩ spots are very diffuse and without clearly defined boundaries. Yurasova, Pleshivtev, and Orfanov (1960) have found that an increase in bombarding ion energy increases the relative intensity of centre to side spots. A similar intensification of centre spots relative to side spots occurs with increasing target temperature (Yurasova and Bukhanov 1962). These effects could in part be due to an increasing background contribution, since the background influence is larger on the centre spots (i.e. in the normal direction). The presence of a low energy background sputtering may also contribute to the observed increase in preferential ejection efficiency (Yurasova and Murinson 1962) and decrease in the average preferential ejection energy (Stuart and Wehner 1964), for small ejection angles to the surface normal, since the background contribution is largest in the normal direction.

The results given in the previous section support the thermal focuson scattering theory (Nelson, Thompson, and Montgomery 1962). This predicts well the temperature broadening of the ejection spots, when background effects are taken into account. A considerable proportion of the total sputtering is due to background sputtering, especially for high target temperatures and high energy bombardment. It is possible that a number of the apparent discrepancies between a focuson ejection theory and experiment, and between the experimental results of different workers, are due to neglect of a proper background correction.

V. References

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