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ENERGY DISTRIBUTIONS OF ATOMS SPUTTERED FROM POLYCRYSTALLINE SURFACES

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A new formula is presented for the energy and angular distributions of atoms ejected from polycrystalline solids due to keV particle bombardment. The formula predicts that the peak position in the energy distribution decreases with increasing polar angle from the surface normal. It also predicts that the angular distribution is $\sim \cos\theta$ for low energy particles and is $\sim \cos^2\theta$ as the energy of the particles increases.

The measured energy distributions of particles ejected from polycrystalline samples subjected to keV ion bombardment have been well described by a model originally developed by Thompson [1]. He assumed that i) in the solid there is an isotropic distribution of velocities, ii) the collision cascade has randomized so that the energy distribution inside the solid, denoted by a subscript i, is E_{i}^{-2}, and iii) the particles must overcome a planar surface binding energy of value U. If E is the energy of the ejected particle and θ is the exit angle as measured from the surface normal, then the yield Y as function of energy and polar angle is given as

$$Y(E, \theta) = \frac{CE \cos\theta}{(E+U)^{n+1}}, \tag{1}$$

where n usually equals 2, and C is a normalization constant. The predictions of eq. (1) are that i) the peak in the energy distribution occurs at U/n, ii) the peak position is *independent* of θ, and iii) the polar distribution is *independent* of E. Over the years this relationship has fitted the experimental data quite well. This agreement is remarkable since the underlying assumption in the Thompson model is that the atoms only undergo binary collisions. For particles that eject the energies $< 2U$, there are undoubtedly multiple collisions. In fact, an attractive interaction e.g., surface binding energy, is inconsistent with binary collisions as attractive interactions are long ranged and binary collisions are only valid for close encounters. Generally in any one experiment either the energy distribution at one polar angle is measured or the polar angle distribution for a large energy bandwidth is measured. In the first case, the constants U and sometimes n of eq. (1) are fitted to the data. In the case of the polar distributions, it has been observed [2–4] that the polar distribution is closer to $\cos^2\theta$ than $\cos\theta$.

Recently energy and angle resolved (EARN) distributions of neutral atoms ejected from In and Rh foils have been measured [5–8]. In these experiments polar distributions at several energies and energy distributions at different polar angles were obtained simultaneously [9]. In this case the primary particle was Ar^+ with 5 keV of energy oriented normal to the surface. There are two interesting deviations from the predictions of the Thompson model. 1) The peak position in the energy distribution shifts to lower angle as the polar angle from normal increases (fig. 1a). Each of the individual curves, however, if U and n are used as parameters, can be well represented by eq. (1). 2) The polar distribution becomes narrower at higher kinetic energies (fig. 2a), so that at low energies the distribution is $\cos\theta$ and at higher energies the distribution is approximately $\cos^2\theta$.

Other researchers have been concerned with corrections to the original Thompson model. Sigmund, Oliva, and Falcone [10] derived an expression where the explicit dependence of the depth of origin of the particles was included. Their resulting expression does not predict any shift in the peak position in the energy distribution with polar angle. Two groups [11,12] have included a dependence of the incident beam angle and energy. The resulting equation predicts that the polar distribution shifts with particle energy, however, it is in the wrong direction as compared with the results from ref. [9] and as shown in fig. 2a.

We have examined various reasons for the deviations from the Thompson model. As Jackson [13] has pointed out, particles that eject with more grazing exit angles should be blocked by neighboring atoms. We have examined [14] the effect of blocking on the energy distributions using a hard sphere model and found that blocking does predict qualitatively the correct shift in

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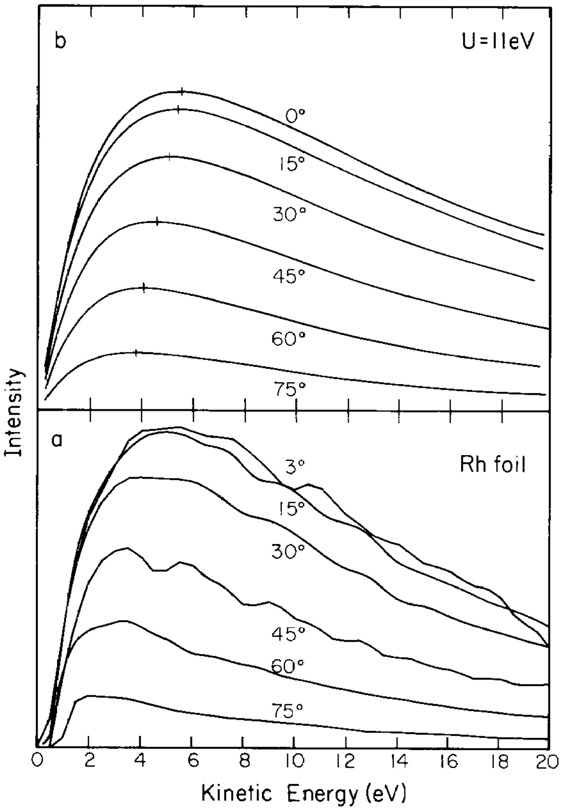

Fig. 1. Energy distributions of Rh atoms ejected from a Rh foil for various polar angles, θ, as measured from the surface normal. a) Experimental results from the authors of reference [9]. The primary ion beam was Ar^+ with a kinetic energy of 5 keV aimed perpendicular to the surface. b) Calculated from eq. (11). The value of U is 11 eV. The cross marks denote the positions of the maxima.

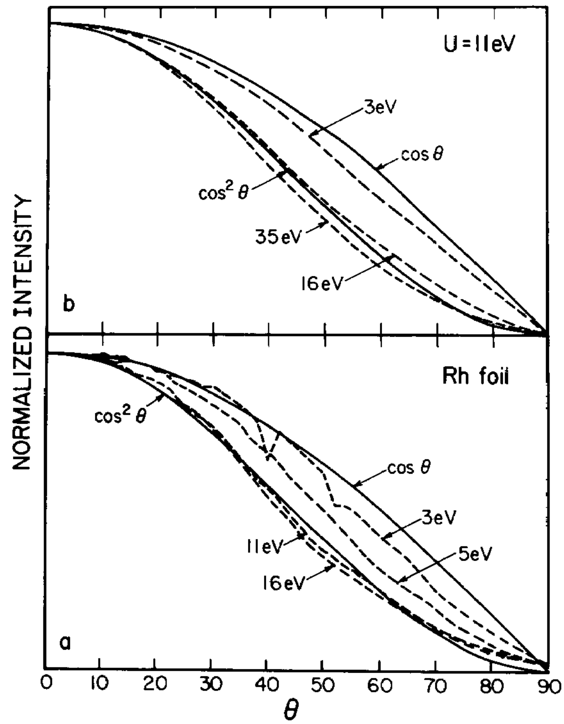

Fig. 2. Polar distributions of Rh atoms ejected from a Rh foil for various energies. All curves are peaked normalized at $\theta = 0°$. a) Experimental results from the authors of ref. [9]. The energy ranges are ± 1 eV. b) Calculated from eq. (11). The value of U is 11 eV.

the peak position in the energy distribution. However, blocking effects also cause the maximum intensity in the polar distribution to *not* be normal to the surface. In other words, blocking contributes to a maximum (at 15–30°) in the polar distribution; a feature important in single crystals and in the older literature termed "a spot". This feature is presumably averaged out for a polycrystalline sample.

We now approach the problem by analyzing the various assumptions in the Thompson model. A reasonable agreement with the experimental distributions can be made by assuming that the distribution in the solid is not isotropic. This is not completely unreasonable. In the Thompson model, conceptually a bulk solid is considered and then an imaginary plane is used to define the surface. In the real solid, the surface undoubtedly influences the distribution of velocities. In addition, there might be memory left of the incident beam direction thus causing additional anisotropies.

We start with the distribution of particle energies and directions inside the solid with

$$f_i(E_i, \theta_i) = \cos^m\theta_i / E_i^2, \tag{2}$$

where we have assumed that n of eq. (1) equals two. The distribution outside the surface is given by

$$Y(E, \theta) = f_i(E_i, \theta_i) J \begin{vmatrix} \cos\theta_i & E_i \\ \cos\theta & E \end{vmatrix} \cos\theta, \tag{3}$$

where J is the Jacobian of transformation and the final $\cos\theta$ term in eq. (3) is to account for the angular dependence of the flux of particles traversing the surface. As in the Thompson model we assume that the particles leaving the surface must overcome a planar surface binding energy, U, that only affects the prependicular energy or velocity. As in ref. [1], the relevant equations of transformation are

$$E_i = E + U, \tag{4}$$

$$\cos\theta_i = \frac{\sqrt{E\cos^2\theta + U}}{\sqrt{E+U}}, \tag{5}$$

$$\frac{\partial\cos\theta_i}{\partial\cos\theta} = \frac{E\cos\theta}{\sqrt{E+U}\sqrt{(E\cos^2\theta + U)}}, \tag{6}$$

$$\frac{\partial \cos \theta_i}{\partial E} = 0, \tag{7}$$

$$\frac{\partial E_i}{\partial \cos \theta} = 0, \text{ and} \tag{8}$$

$$\partial E_i / \partial E = 1. \tag{9}$$

Substituting eqs. (2), (4)–(9) into eq. (3) results in the final distribution of

$$Y(E, \theta) \propto \frac{E \cos \theta}{(E+U)^3} \frac{\sqrt{(E \cos^2\theta + U)^m}}{\sqrt{(E+U)^m}}. \tag{10}$$

In Thompson's case the value of m is 0, and eq.(10) reduces to eq. (1). The choice of $m = 2$ fits the experimental data quite well. In this case eq. (10) becomes

$$Y(E, \theta) \propto = \frac{E \cos \theta}{(E+U)^4}(E \cos^2\theta + U). \tag{11}$$

The energy and polar distributions as predicted by eq. (11) are shown in figs. 1b and 2b respectively. A value of $U = 11$ eV was chosen to obtain a reasonable fit to the experimental data. The agreement between the calculated and experimental curves is remarkable. The energy peak position shifts to lower energy as the polar angle increases. The expression for the energy peak position is given as follows:

$$\frac{E_{peak}}{U} = \frac{\left(2 \cos^2\theta - 3 + \sqrt{4 \cos^4\theta - 4 \cos^2\theta + 9}\right)}{4 \cos^2\theta}. \tag{12}$$

For $\theta = 0°$, $E_{peak} = U/2$ and for $\theta = 90°$, $E_{peak} = U/3$. The angle integrated peak occurs at $(\sqrt{2} - 1)U$.

We have treated U as a fitting parameter and its physical significance is not clear at this stage. Of importance here is that the trend of the energy peak position shifting to a lower value as θ increases is independent of the choice of U. Investigations are underway to understand more fully the factors which determine the peak position in the energy distribution.

In the case of the polar distributions, at very low energies the distribution is nearly cos θ. As the energy of the particles increases, the polar distribution becomes narrower (fig. 2). In the limit of very high energies the predicted distribution becomes $\cos^2\theta$. If all energies are averaged over the polar distribution is

$$Y(\theta) \propto \cos \theta (2 \cos^2\theta + 1)/3, \tag{13}$$

where $Y(\theta = 0°) = 1$. The difference between eq. (13) and $\cos^2 \theta$ is less than 0.03.

The angle integrated energy distribution is

$$Y(E) \propto \frac{E}{(E+U)^3} + \frac{EU}{(E+U)^4}. \tag{14}$$

The first term is the same as from eq. (1) and dominates for large values of E. Eq. (14) also gives that the average peak position should occur at $(\sqrt{2} - 1)U$.

In conclusion, we have presented a new analytic model for the energy and angular distributions of atoms ejected due to keV particle bombardment at normal incidence from polycrystalline solids. The main modification from the Thompson model is to assume that the velocity distribution near the surface region is *not* isotropic. The model presented here predicts that the peak in the energy distribution shifts to lower energies as the polar angle increases and that the polar distribution becomes narrower as the energy of the particles increases.

The modifications of the Thompson model presented here are qualitative at best. The experimental feature that precipitated this study is the shift in peak position in the energy distribution with polar angle. This occurs in a low collisional energy regime where attractive interactions are important. The fundamental assumptions of binary collisions and a planar surface binding energy are suspect. What we have hoped to accomplish is to provide an analytic formula of energy and angular distributions that better fits the experimental data. Efforts are underway to assess via computer simulations [15] the assumptions leading to eqs. (1) and (11).

Inclusion of effects such as non-normal angle of incidence of the primary beam needs to wait until we have a fundamental understanding of the dominant cause of the anisotropy. For example, if the anisotropy is due to the presence of the real surface instead of a planar surface then there might be minimal changes in the distributions with the primary particle angle of incidence. On the other hand, if the anisotropy is inherent in the collision cascade, then possibly angle of incidence effects could be important.

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References

[1] M.W. Thompson, Phil. Mag. 18 (1968) 377.
[2] M.F. Dumke, T.A. Tombrello, R.A. Weller, R.M. Housley, and E.H. Cirlin, Surf. Sci. 124 (1983) 407.
[3] H.H. Andersen, B. Stenum, T. Sørensen, H.J. Whitlow, Nucl. Instr. and Meth. B6 (1985) 459.
[4] S. Kundu, D. Ghose, D. Basu, and S.B. Karmohapatro, Nucl. Instr. and Meth. B12 (1985) 352.
[5] J.P. Baxter, G.A. Schick, J. Singh, P.H. Kobrin and N. Winograd, J. Vac. Sci. Technol. 4 (1986) 1218.

[6] G.A. Schick, J.P. Baxter, J. Singh, P.H. Kobrin and N. Winograd, in: Secondary Ion Mass Spectrometer – SIMS V, Springer Series in Chemical Physics 44 (1986) 90.

[7] P.H. Kobrin, G.A. Schick, J.P. Baxter and N. Winograd, Rev. Sci. Instrum., in press.

[8] N. Winograd, P.H. Kobrin, G.A. Schick, J. Singh, J.P. Baxter and B.J. Garrison, Surf. Sci. Lett., in press.

[9] J.P. Baxter, J. Singh, G.A. Schick, P.H. Kobrin, and N. Winograd, Nucl. Instr. and Meth. B17 (1986) 300.

[10] P. Sigmund, A. Oliva, and G. Falcone, Nucl. Instr. and Meth. 194 (1982) 541.

[11] H.E. Roosendaal and J.B. Sanders, Proc. Symp. on Sputtering, eds., P. Varga, G. Betz, and F.P. Viehbock, Perchtoldsdorf, Vienna (1980).

[12] Y. Yamamura, Nucl. Instr. and Meth. 194 (1982) 515.

[13] D.P. Jackson, Can. J. Phys. 53 (1975) 1513.

[14] B.J. Garrison, unpublished results.

[15] D. Lo, M. Sharpiro, T. Tombrello, B.J. Garrison, and N. Winograd, unpublished results.