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ENERGY AND ANGULAR DISTRIBUTIONS OF ATOMS SPUTTERED FROM POLYCRYSTALLINE SURFACES: THOMPSON AND BEYOND

Barbara J. GARRISON *

Department of Chemistry, The Pennsylvania State University, University Park, PA 16802, USA

We present 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. We have shown from computer simulations that the anisotropy in the surface region is due to the inherent asymmetry of the surface–vacuum interface. Finally, we and others have shown that the energy cost to remove an atom from the solid is greater than the heat of sublimation.

The measured energy distributions of particles ejected from polycrystalline samples subjected to keV ion bombardment have been reasonably 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^{-n} and iii) the particles must overcome a planar surface binding energy of value U, a quantity which is often equated to the heat of sublimation. 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

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

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 fit 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 with 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 fit to the data. In the case of the polar distributions, it has been observed [2–4] that the polar distribution is often 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 three interesting deviations from the predictions of the Thompson model. 1) The peak position in the energy distribution shifts to a lower value 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. 1c), so that at high energies the distribution is approximately $\cos^2\theta$. 3) The value of U needed to fit the data is larger than the heat of sublimation.

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. 1c.

* Camille and Henry Dreyfus Teacher Scholar.

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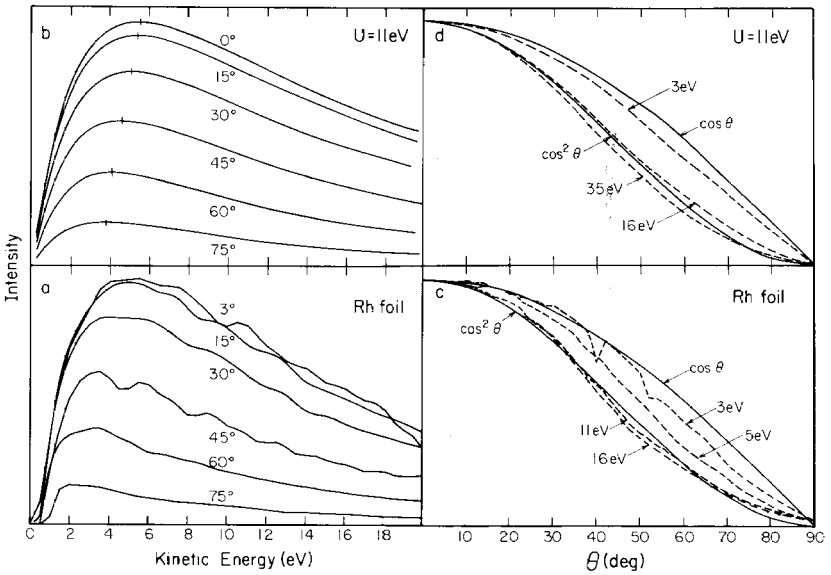

Fig. 1. Energy distributions of Rh atoms ejected from a Rh foil for various polar angles, θ, as measured from the surface normal. a) Experimental results from the authors of ref. [9]. The primary ion beam was Ar^+ with a kinetic energy of 5 keV aimed perpendicular to the surface. b) Calculated from eq. (4). The value of U is 11 eV. The cross marks denote the positions of the maxima. Polar distributions of Rh atoms ejected from a Rh foil for various energies. All curves are peaked normalized at $\theta = 0°$. c) Experimental results from the authors of ref. [9]. The energy ranges are ± 1 eV. d) Calculated from eq. (4). Modified from ref. [13].

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 velocity distribution in the solid is *not isotropic* [13]. This is not completely unreasonable. In the Thompson model, conceptually an infinite 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.

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

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

where we have assumed that n of eq. (1) equals 2. Applying the same transformations as Thompson in order to get the distribution outside the solid results in the final distribution of

$$Y(E, \theta) \propto \frac{E\cos\theta}{(E+U)^3}\left(\frac{(E\cos^2\theta + U)^m}{(E+U)^m}\right)^{1/2}. \tag{3}$$

In Thompson's case the value of m is 0, and eq. (3) reduces to eq. (1). The choice of $m = 2$ fits the experimental data reasonably well and simplifies the mathematics. In this case eq. (3) becomes

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

The energy and polar distributions as predicted by eq. (4) for rhodium are shown in figs. 1b and 1d 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. 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. 1d). In the limit of extremely high energies the predicted distribution becomes $\cos^3\theta$. If all energies are averaged over the polar distribution then

$$Y(\theta) \propto \cos\theta(2\cos^2\theta + 1)/3, \tag{5}$$

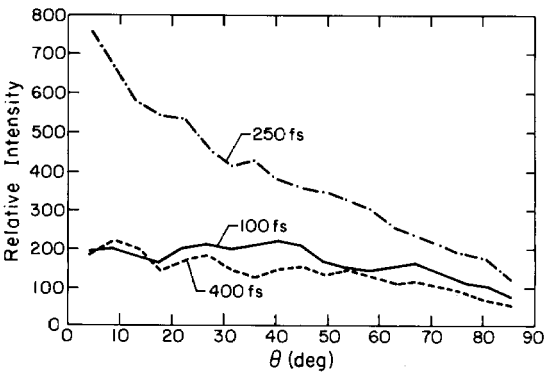

Fig. 2. Time development of the angular distributions in the first surface layer [14]. The majority of particles eject between 100 and 250 fs (1 fs = 10^{-15} s).

where $Y(\theta = 0°) = 1$. The difference between eq. (5) and $\cos^2\theta$ is less than 0.03.

Computer simulations of the keV particle bombardment of an amorphous indium sample were performed by Lo et al. [14] in order to verify the assumption of anisotropy in the velocity distributions. The angular velocity distributions were determined for the surface layer as a function of time in the collision cascade (fig. 2). Their results show that the presence of the free surface (neglected in the Thompson model) causes significant anisotropy in the outermost surface region. The distribution quickly became isotropic in the subsurface region [14]. Of note is that the simulations reproduced the shift in peak position in the energy distribution with polar angle and the narrowing of the polar distribution with increasing sputtered particle energy.

The final issue is the choice of a value for U. As a matter of convenience (and lacking a better choice) the value of the surface binding energy is often equated to the heat of sublimation of the solid, ΔH_s. The choice of ΔH_s for the energy parameter in the prediction of the sputtering yields gives a reasonable agreement with the measured quantities [5]. However, Husinsky [16] has pointed out that the values of U fit to experimental energy distributions are sometimes larger than heats of sublimation of the materials.

Our contention is that the energy cost to remove an atom from a solid is actually greater than the heat of sublimation [17]. The roots of this idea lie in two virtually unnoticed papers by Jackson [18,19]. He simulated the ejection of a surface atom by giving it an initial kinetic energy. The final kinetic energy (for a pairwise additive potential approximation) corresponds to an energy loss greater by 30–40% than the bulk heat of sublimation. One would think that this would imply that by ripping the entire solid apart it would take more energy than it had in the first place.

This dilemma is best described by examining a diatomic molecule which has a bond strength of D_e. Under the rules used for solids, the binding energy of each atom is $D_e/2$. If, however, one atom is clamped fixed and one asks how much kinetic energy must be supplied to the other atom so that the bond can be ruptured, then the answer is D_e – twice the "binding energy". (Note that it now "costs" nothing to remove the second atom.) An analogous situation occurs in the solid. In one extreme in the ion bombardment process all atoms but one are fixed. The energy cost for this atom to escape the solid is greater than ΔH_s in line with recent experimental results. It is not clear either theoretically or experimentally precisely what is the value of the energy cost to remove an atom from a solid. It is also not clear what influences the magnitude of this energy cost. Should it be the same for amorphous Rh, Rh{111}, Rh{110} and Rh{100}, for example?

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. We have shown from computer simulations that the anisotropy in the surface region is due to the inherent asymmetry of the surface–vacuum interface. Finally, we and others [20] have shown that the energy cost to remove an atom from the solid is greater than the heat of sublimation.

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.

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