MANY-BODY INTERACTIONS FOR THEORETICAL STUDIES OF KEV PARTICLE BOMBARDMENT

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The sequence of events initiated in a solid by bombardment with energetic heavy particles are highly complex. Over the years there have been numerous attempts to describe the microscopic atomic motions and the macroscopic experimental observables in both a qualitative and quantative manner. The most detailed of the theoretical approaches is that of computer simulations. Over the years we have used a model that incorporates simultaneous interactions among the atoms and utilizes a pairwise additive interaction potential that includes an attractive portion[1-4].

These potentials have been quite successful at elucidating mechanisms of particle ejection, and at allowing calculations of semi-quantitative aspects of the sputtering process[5]. However, a quantitative comparison between theory and experiment has been hindered by a scarcity of detailed experimental data. A new method based on the multiphoton resonance ionization of neutral atoms after they have desorbed from the target surface has been developed recently. The technique is capable of measuring the energy and angle resolved neutral (EARN) distributions of sputtered atoms[6-8] with sensitivity sufficient to avoid surface damage. These data provide the best trajectory data yet available and force a critical test of assumed interaction potentials and scattering dynamics.

In a previous study we described the EARN distributions of Rh atoms ejected from Rh{111} with a computer simulation using pair-wise additive interaction potentials[4]. Although the overall trends of azimuthal anisotropies and relative intensities of angular peaks were well-described, the position of the peaks and the peak widths in both the polar angle and energy distributions differed between the experimental and calculated results. Variation of the parameters in the potential within physically reasonable bounds did not significantly improve the agreement.

In this paper we present the use of many-body potentials to describe the ejection of atoms from solid surfaces due to keV ion bombardment. The potential is derived from the embedded atom method (EAM) of Daw and Baskes[9-10]. This many-body potential significantly improves the agreement between the measured and calculated peak positions and widths in the energy and angular distributions of Rh atoms ejected from Rh{111}.

Our prescription for modeling the ion bombardment process has been described in detail elsewhere for the Ar^+ ion bombardment of Rh{111}[4]. The essential change here is that a many-body EAM potential has been used to describe the Rh metal interaction. The EAM considers the metal as ion cores in an electron sea[9-11]. The energy for the i^{th} atom is given by

$$E_i = F(\rho_i) + 1/2\ \Sigma\ \phi(r_{ij})$$

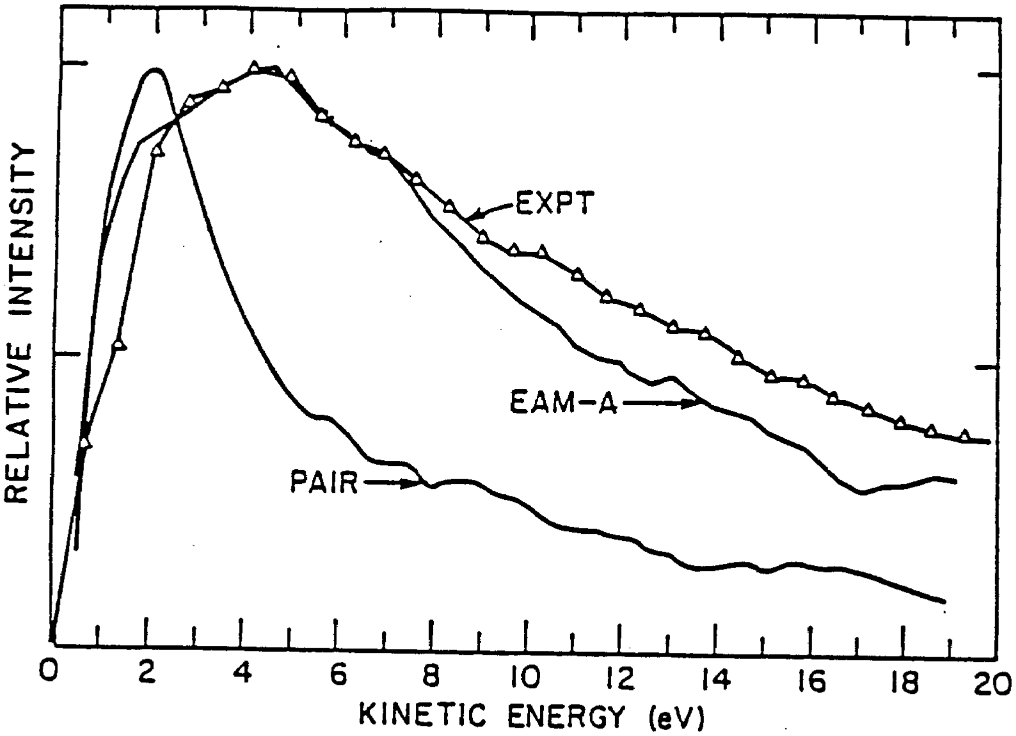
where r_{ij} is the distance between the i^{th} atom and the j^{th} atom. The first term is the embedding function, which is the energy of the interaction of the ion core with the electron sea of density ρ_i. For convenience this density is assumed to be the sum of the atomic densities of the other atoms at the position of the atom of interest. The second term is short ranged and is the ion-core repulsion at a distance of separation, r_{ij}, of the two cores. It is of interest that the EAM has been successfully applied to describing bulk properties at thermal energies and even surface reconstructions[9-11]. However, to our knowledge it has not been applied to systems where the solid actually dissociates as in the sputtering process.

The EAM potentials are empirically derived and as a consequence the $F(\rho)$ and the $\phi(r)$ must somehow be determined. Daw, Baskes and Foiles have developed a prescription for fitting the functions to equilibrium properties of the metals[11]. We have used their basic scheme for fitting the functions but have included the EARN data in the base of known experimental quantities. Our procedure for fitting the functions and our justification for picking one potential (EAM-A) as the best one so far is described in detail elsewhere[12].

The most dramatic change in the predicted distributions in going from the pair to the EAM potentials arises in the angle integrated energy distributions. As shown in the Figure the distributions from experiment and calculations using the EAM-A interaction are in excellent agreement while the calculated distribution using pair potentials is quite different. The increase in agreement in the energy distributions is fairly consistent among all the EAM potentials tested but the angular dependence does depend on the specifics of the EAM potential[12].

Is the better agreement fortuitous or is there a sound basis for it? It has been clear to us in the past that the pair potential description in the surface region is inadequate as the number of nearest-neighbor atoms in the surface region is different from that in the bulk. An examination of the EAM potentials and the pair potential in the surface region shows several differences important for the particle ejection process.

First the surface binding energy of the EAM potentials is larger (5.1-5.3 eV) than that of the pair potential (4.1 eV) even though all potentials have been fit using the bulk heat of atomization of Rh of 5.75 eV. The conventional wisdom of sputtering theory is that the peak position in the energy distribution is proportional to the energy cost to remove an atom from the surface[13-15]. However, a ~1 eV increase in the binding energy does not in itself justify a ~2 eV shift in the peak position. The EAM potentials are relatively flat in the attractive portion of the entire surface region. There is a 3-4 eV attraction for the ejecting atom even above a neighboring atom. The pair potential has only ~1 eV overall attraction. Thus particles ejecting at more grazing angles will experience a larger attraction to the surface in the EAM potential than the pair potential. This will tend to make the peak in the energy distribution shift to larger energies and will also pull the particles away from the surface normal.



Angle integrated experimental and calculated kinetic energy distributions. In all cases the curves are peak normalized.

This increase in the surface binding energy in the EAM description causes fewer particles to be ejected with low kinetic energies. Thus to a first approximation the change in the features in the energy distribution are a result of a decrease in the peak intensity, resulting in a more intense high energy tail. This increased binding energy also dramatically affects the total yield. For the pair potential calculation the calculated yield is 6.1 Rh atoms per incident Ar particle while for EAM-A the yield is 3.7 atoms per Ar, a value in better agreement with experimental yields. Note also that the ratio of the yields between the two methods of calculations is 6.1/3.7=1.6 whereas the ratio of surface binding energies is only 5.1/4.1=1.2. Transport theories of sputtering suggest that the yield is inversely proportional to the binding energy[16]. From our calculations it is clear that the nature of the potential surface also influences the expected yield.

It is apparent that a many-body potential is necessary to quantitatively describe the EARN distributions of sputtered atoms although at this time we do not believe that EAM-A is the perfect potential. Work is underway to improve the EAM fitting procedure and to find other many-body interaction potentials.

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