THE INFLUENCE OF MEGAEVENT COLLISION SEQUENCES ON SECONDARY IONIZATION

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I. Introduction

There are now several successful models which provide a reasonable description of secondary ion formation in SIMS.[1,2] For clean and adsorbate–covered surfaces where the bonding is delocalized, a tunneling model has been extensively tested.[2] In this situation there is resonant charge exchange between the desorbed particle and the surface in the 1–5 Å region above the solid where there is still significant electronic coupling. Since a high degree of electronic overlap in a metal results in the rapid dissipation of the initial electronic excitation, it is presumed that excitations within the solid do not contribute to the ionization probability, R^+. The tunneling model has accurately predicted the dependence of R^+ with ionization potential and surface work function. The appropriate velocity dependence of R^+ is still undecided with various functional forms under discussion.

Recently, Nourtier and coworkers[3] have presented a model in which the electronic structure of the solid is sufficiently distorted to shift the interaction of the ejecting particle with the substrate from the delocalized metal s–band to the more localized d–band. As a consequence of this perturbation, it is possible that excitations created in the collision cascade by a particular mechanism can survive to escape the surface. For clean metals where the value of R^+ is inherently quite low, this effect may have a major influence on the ion distributions even though the corresponding neutral particle distribution may be unaffected. Molecular dynamics calculations of the ion–bombardment process have revealed that there are certain high action impact points on a single crystal target that produce an anomolously high yield of ejected particles.[4] This type of collision event has been discussed for many years[1] and has recently been termed a "megaevent".[4] At this point, it is unclear whether there is a connection between

Nourtier's proposal for the creation of localized states, "megaevents", and any observable property associated with R^+.

During recent years, our laboratory has acquired the unique capability of being able to measure the trajectories of both ionic *and* neutral species desorbed from bombarded clean single–crystal targets. The ion distributions are obtained using a specially designed movable quadrupole mass analyzer with a 90° sector kinetic energy prefilter.[5] The neutral distributions are determined by a pulsed–laser–based methodology. Multiphoton resonance ionization is used to selectively convert the neutral species to positive ions at a specific point in space and at a known time. Energy and angular distributions are then determined using a time–of–flight imaging detector.[6] Both of these methods are sensitive enough to yield trajectory data in the low–dose or static regime. In this paper, we compare the velocity distribution of Rh^+ ions and Rh atoms desorbed from a clean Rh{111} crystal. We find significant structural features in the ion distributions. Preliminary molecular dynamics calculations suggest that "megaevents" may indeed be responsible for this structure.

II. Results and Discussion

The relevant kinetic energy distributions for Rh and Rh^+ ions desorbed from Rh{111} are shown in Fig. 1. The curves were obtained using 3 keV Ar^+ ion bombardment at normal incidence with detection at a polar angle, $\theta_d = 45°$. The total ion fluence during an experiment was approximately 5×10^{13} ions/cm^2. The most striking feature of these results is the doubly peaked structure in the ion distributions which is not evident in the neutral atom distribution. The low energy peak occurs at 7–10 eV and the higher kinetic energy peak is seen at 22–28 eV. The relative intensity of the two peaks varies with the polar and azimuthal angles of detection. The high energy structure is most intense at $\theta_d = 45°$ at an azimuthal angle ϕ of +30°. As seen in the figure, this feature is reduced by about 50% for $\phi = -30°$. We also find that it steadily diminishes in intensity with increasing polar angle for all azimuthal directions.[7] We have not seen any evidence for this structure with the neutral distributions at any detection angle. Note that in our case, the effect cannot arise from patches of impurities on the surface[8] or from direct recoils which are observed for glancing angles of incidence.[9]

Molecular dynamics calculations provide an important clue to the origin of this structure. Using this approach we have successfully computed kinetic energy distributions which accurately match the experimental ones.[10] By analyzing the computed trajectories, we find that when second–layer atoms are ejected, they are accompanied by the ejection of several first–layer atoms. In fact, the yield of these processes can be 2 to 3 times as large as the average yield. This effect has also been noted for bombarded Cu

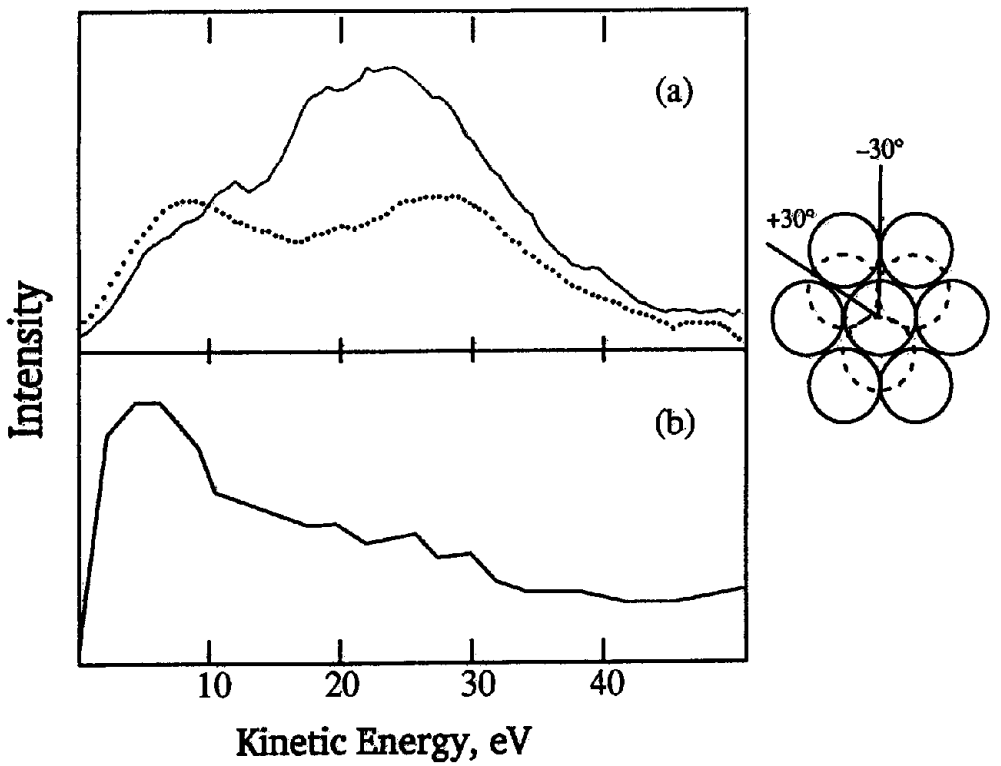

Fig. 1. Kinetic energy distributions for Rh^+ ions (a) and Rh neutral atoms[10] (b) ejected from Rh{111} during bombardment at normal incidence by 3 keV Ar^+ ions. In (a), the distributions are shown for the $\phi = +30°$ azimuth (solid line) and the $-30°$ azimuth (dotted line). In (b) the $+30°$ azimuth is shown. In all cases, the polar angle of detection is 45°.

crystals.[11] Moreover, these high yield collision sequences lead to ejection of atoms predominantly along the $\phi = +30°$ azimuth for a restricted energy range. The ratio of intensities for $\phi = +30°$ to $\phi = -30°$ is displayed in Fig. 2 for 3 different secondary particle kinetic energies. The yield minimum is defined as the threshold of ejected particles per impact event in order for that collision sequence to be included in the calculation of the azimuthal angle intensity ratio. The enhanced yield for the 20 eV secondary Rh atoms relative the the other kinetic energy ranges is clearly evident in the figure.

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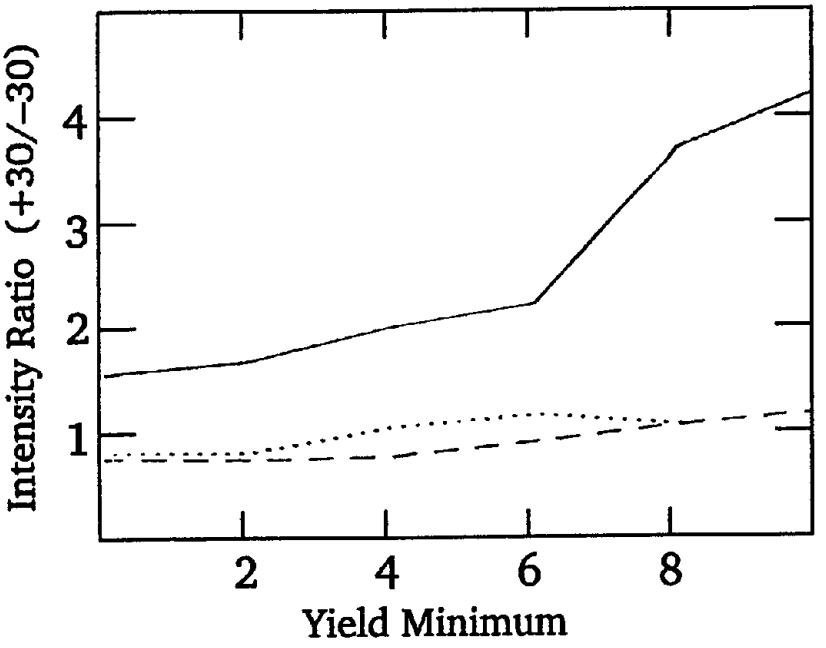

Fig. 2. Calculated yield ratio along two different azimuthal directions of Rh{111} as a function of the minimum number of ejected particles per impact event that the collision sequence must produce in order to be counted. The calculations utilized 3000 trajectories with normally incident 3 keV Ar^+ ions. The kinetic energies of the ejected Rh atoms are 8–eV (broken), 20–eV, (solid), and 30 eV(dotted).

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