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EJECTION OF ATOMS FROM RARE-GAS SOLIDS BY LOW ENERGY CASCADES

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The results of a molecular dynamics calculation are presented for collisional ejection of atoms from excited low temperature rare-gas solids. An atom in the solid is assumed to receive an amount of kinetic energy consistent with an energetic, non-radiative decay of an excited dimer and the number, energy and direction of ejected atoms are calculated as functions of depth in the solid of the initially energized atom. The results are compared to the standard binary-collision cascade model and to a thermal-spike model for ejection. The yields integrated over depth are found to depend nearly linearly on energy deposition as in the collision cascade model. They also scale with the surface number density and the surface binding energy. The calculated angular and energy distributions are relatively insensitive to the exciting energies and exhibit distinct differences from the cascade and thermal models. However, the peak position in the energy spectra is consistent with the cascade model if the surface binding energy, which is about two thirds the cohesive energy for these solids, is used. Experimental results on the electronic sputtering of solid argon are briefly considered.

1. Introduction

In this paper we examine the behavior of low energy collision cascades in weakly bound solids. This is of interest in the sputtering (desorption, ablation) of low temperature condensed gas solids produced by electronic excitation as a means of studying, e.g., the non-radiative relaxation processes occurring in such materials [1]. In fact, studies of the sputtering of rare-gas solids by fast ions (~ 1 MeV/amu) have demonstrated that the electronic relaxation

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processes are quite complex [2–4]. That is, on excitation the atoms in a Van der Waals solid become chemically active. An excited state produced by the incident ion diffuses through the crystal until it interacts with a ground state atom forming an excited dimer. As the atoms in these eximers have inter-nuclear separations much smaller than the normal lattice spacings, the relaxation process eventually involves a repulsive displacement in which the two atoms forming the dimer acquire kinetic energy. Johnson and Inokuti [2] review the non-radiative relaxation processes associated with excited dimers formed in rare-gas solids. In the lowest excited state these dimers are known to relax via emission of a photon. The transition to the ground state leaves the system with over 1 eV of repulsive energy [3–5]. In the highly excited states comparable amounts of repulsive energy may also be deposited due to crossing of the bound states of the dimers with repulsive states, as in predissociation or dissociative recombination processes which occur in the gas phase. If the relaxation occurs close to the surface the repulsive energy available may result in the ejection or sputtering of atoms from the solid. Low energy ($\sim$ keV) incident ions also sputter condensed gas solids efficiently. In this case the kinetic energy is deposited in the solid by direct collisions with the incident ion.

In order to interpret sputtering experiments as well as experiments on sub-monolayer desorption [6], it is necessary to know the yield of ejected atoms from the solid due to the kinetic energy deposited by the relaxation process. Models for describing the cascade of events produced by an atom with a given amount of kinetic energy have been used successfully when this energy is many tens of eV [1,7]. These models have not been tested experimentally or by accurate calculations when the energy input is of the order of a few electron volts or less. In solids where the binding energy is tenths of an eV or less such energies may, in principle, result in the ejection of many atoms. As the simple expressions available for atomic ejection depend very differently on the initial kinetic energy of the atoms and the surface binding energy of the material, it is important to have such a test.

In this paper we use a molecular dynamics procedure described earlier [8,9] to calculate the number of atoms ejected from solid argon and xenon and the energy and angle spectra of these ejected particles. Results are given versus kinetic energy input over a narrow range of low energies from 0.3 to 3 eV, and we discuss how the results can be scaled to other atomic solids. Such energies might be appropriately associated with the kinetic energy acquired by the atoms of an excited dimer on relaxation in the solid. For comparison we also simulate the repulsive decay of a dimer following radiative decay to the ground state. These calculations, therefore, will be useful for interpreting electronically induced surface sputtering (desorption, ablation) whether the initiating species are photons, electrons or ions. In a subsequent paper we shall consider molecular solids.

2. Calculations

2.1. Kinetic energy deposition

The first set of calculations is designed to determine the ejection yield if a quantity of energy ΔE is deposited as kinetic energy a distance z into the solid. The energized atom could have been struck by another atom in the solid or be part of the repulsive decay of an excited state. The molecular dynamics calculations used to describe the behavior of the atom in the solid are similar to those described previously [8,9], so only a discussion of the interaction potentials is presented here.

The argon atoms are arranged in a face centered cubic array of lattice constant 5.31 Å. The interaction among the atoms is assumed to be pairwise additive with a Morse function used for the pair potential. Since this potential includes an attractive interaction no effects due to surface binding need to be accounted for post facto. Published Morse parameters are used [10]: $D_e = 0.0114$ eV, $\alpha = 1.425$ Å^{-1}, and $R_e = 4.04$ Å. For numerical efficiency we have truncated the potentials at a distance of 6.38 Å. With this cut-off an atom still interacts with second nearest neighbors but the bulk cohesive energy is 0.061 eV per atom as compared to 0.08 eV if infinite range parameters are used. As the purpose of the simulation is to test models for atomic ejection use of the exact binding energy is not necessary as long as the differences are kept in mind when applying the results to ejection data for argon. The potential energy of {100} and {111} surface atoms in this description are 0.042 eV and 0.044 eV respectively. In calculating the yield we assume the initial direction of motion of the energized atom is random and, therefore, we average over the orientation of the velocity vector by performing several calculations for each value of ΔE and depth z of the atom in the solid. Approximately 20 different velocity orientations have been calculated for each combination of values of z (from 0–14 Å), ΔE (0.3, 0.7, 1.0, 2.0, and 3.0 eV) and exposed crystal face ({100} and {111}).

Similar calculations are also performed for a xenon crystal with the {100} crystal face exposed. The Morse parameters used are $D_e = 0.026$ eV, $\alpha = 1.366$ Å^{-1} and $R_e = 4.375$ Å, and the cut-off distance is 7.37 Å. In this case we chose these parameters so the truncated potentials yield the correct heat of sublimation (0.17 eV), lattice constant (6.13 Å) and compressibility (2.75×10^{-11} cm^2/dyn) of solid Xe. The removal energy of a {100} surface atom with these parameters is 0.11 eV. As in the above example, this is approximately 2/3 the bulk cohesive energy. The same reduced energies (ratio of ΔE to Morse well depth D_e) are used for the initial kinetic energy of the Xe atom as were used for the Ar atoms in order to determine how the results scale with ΔE and binding energy. The values of ΔE used for Xe are 0.68, 1.68, 2.28, 4.56 and 6.84 eV.

The calculated yields as a function of depth z are plotted in fig. 1 for the five values of ΔE and the two different argon crystal faces. The depth is given in terms of crystal layers. The magnitude of the difference in yields for the two faces is of the same order as the statistical uncertainty. Virtually all of the yield is due to atoms that were originally in the first layer. For a given value of ΔE the maximum yield occurs when the excitation energy is initially 2–3 layers deep. Except for the case where the excitation energy is initially in the 1st layer ($z = 0$) the energized atom itself does not eject. If the energized atom is in the 1st layer it ejects when the initial velocity is oriented out of the solid. For ΔE equal to 0.3 and 0.7 eV the ejection of the energized atom dominates the yield. Clusters of atoms, e.g., Ar_2 and Ar_3, are observed primarily from the calculations at $\Delta E = 1$–3 eV and with the excitation in layers 2 or 3. There are not sufficient clusters to report reliable yields. However, as in the results from sputtering calculations and experiments, more clusters are observed to eject from the {111} rather than the {100} face [11]. The Xe results are also shown in fig. 1. For the same values of reduced energy ($\Delta E/D_e$) the results for Xe are within the statistical error of the Ar results implying that scaling of these results with binding energy is appropriate even though the Xe binding energy is more than twice that of Ar with the potential parameters used here.

In order to calculate the average total yield produced by incident particles the sputtering contribution from each layer, weighted by the likelihood of an excitation event in that layer, must be summed over all layers. If the solid is
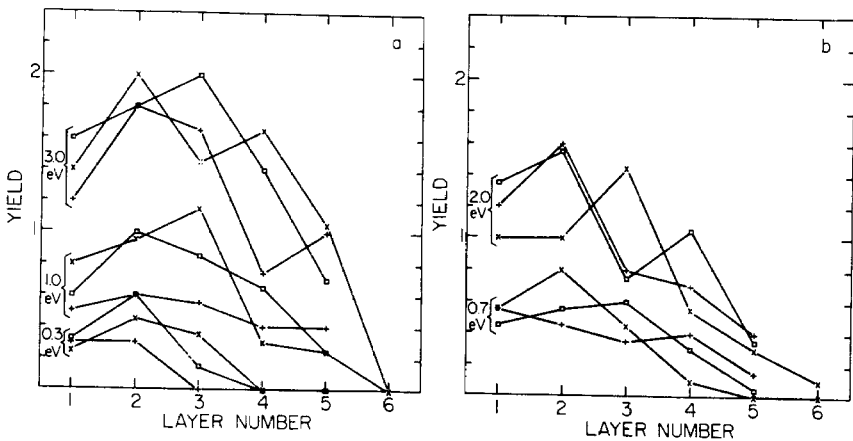

Fig. 1. Calculated yield versus layer depth for the indicated values of ΔE used for argon. The energies, ΔE, scaled for Xe are 0.68, 1.68, 2.28, 4.56 and 6.84 eV. The lines act only as a guide for the eye. (O) Ar{100}, interlayer spacing is 2.655 Å. ($\times$) Ar{111}, interlayer spacing is 3.066 Å. (+) Xe{100}, interlayer spacing is 3.066 Å. (For argon the binding energy is lower than measured values.)

amorphous then

$$Y(\Delta E) = \int_0^\infty P(z, \Delta E) \, Y(z, \Delta E) \, dz. \tag{1}$$

In eq. (1), $P(z, \Delta E)$ is the probability per unit path length of energy ΔE being deposited at a depth z in the solid. When $P(z, \Delta E)$ is independent of depth, or varies slowly over the region where $Y(z, \Delta E)$ is non-zero, then

$$Y = \Delta z_s / \lambda, \tag{2}$$

where λ is the mean-free-path for producing an event which initiates atomic motion and Δz_s is a weighted sputter depth [1],

$$\Delta z_s = \int_0^\infty Y(z, \Delta E) \, dz.$$

Although we shall use eq. (2), recent results on proton sputtering of solid argon indicate that $P(z, \Delta E)$ is not always a constant [4]. In our calculations, in which the solid is crystalline and $Y(z, \Delta E)$ in eq. (2) is determined for layers with discrete separations, Δz_s is a sum of the contributions from each layer times the spacing between layers.

We considered two models for calculating Δz_s for electronic sputtering of condensed gas solids [1,12]. The first is based on the model for collision cascades [7,12] in which

$$\Delta z_s = \frac{c}{n \bar{\sigma}_d} \left(\frac{\Delta E - U}{U} \right). \tag{3}$$

In this expression U is the surface binding energy (although heat of sublimation is often used [7]), n the number density, and $\bar{\sigma}_d$ is the momentum transfer (diffusion) cross section for collisions of atoms in the solid [12]. As the diffusion cross section is slowly varying with the energy of the atoms the bar indicates an average value. In the standard expression for sputtering by direct collision with the incident ion, a value of $\bar{\sigma}_d \approx 3.8$ Å^2 is generally used and $c \sim 3/(2\pi^2)$ [7,12]. For low energy collisions between atoms (or molecules) in an amorphous condensed-gas solid it is reasonable to write [1] $\bar{\sigma}_d \propto n^{-2/3}$. If, in addition, a hard sphere interaction is used then $c \sim 1/8$. The second model considered assumes that collisions rapidly thermalize the deposited kinetic energy, ΔE, after which this energy is transported as in a mini-thermal spike [1,12,13]. In this case

$$\Delta z_s = 0.02 \bar{\sigma}_d n^{1/3} \left(\frac{\Delta E - U}{U} \right)^{5/3}. \tag{4}$$

which has a dependence on ΔE and $\bar{\sigma}_d$ which is very different from the expression in eq. (3).

In fig. 2, Δz_s divided by the separation, l, between layers is plotted versus $(\Delta E - U)/U$ where U is the binding energy of an atom on the surface. The

values shown are calculated using the results for the yield given in fig. 1. Also shown are two curves for Δz_s each having a dependence on ΔE like that in eq. (3) or eq. (4). It is apparent that the dependence on ΔE found from our calculations is much closer to that of the collision cascade model over the energy range shown.

It is intriguing that on scaling both ΔE and Δz_s the three calculations show reasonable agreement. This implies that the expression in eq. (3) may be used for ejection by low energy atoms if $(n\bar{\sigma}_d)^{-1}$ is replaced by a quantity proportional to the layer spacing in the material. This is consistent with our observation that atoms leave primarily from the surface layer. Noting that $l = (nA)^{-1}$, where A is the net area occupied by an atom on the surface, then A essentially replaces $\bar{\sigma}_d$ in eq. (3). The straight line drawn through the data, which shows reasonable agreement, is obtained from eq. (3) if the interaction is $c \sim 0.12$ and $\bar{\sigma}_d \sim A$. For an amorphous solid this is equivalent to writing $\bar{\sigma}_d \sim n^{-2/3}$. Care should be taken when drawing conclusions from this result as our calculation of $\Delta z_s/l$ gives only the ratio $cA/\bar{\sigma}_d$ using eq. (3) and not c or $\bar{\sigma}_d$ separately. Also, if the bulk cohesive energy rather than the surface binding energy is used for U then this ratio is about 1.5 times larger.

Both the kinetic energies and the binding energies considered here are small. However, the ratio $\Delta E/U$ is large at the highest energies considered. This is one condition for the standard collision cascade model. The other criterion for this model is that only binary collisions are important [7]. Since the cross-sec-
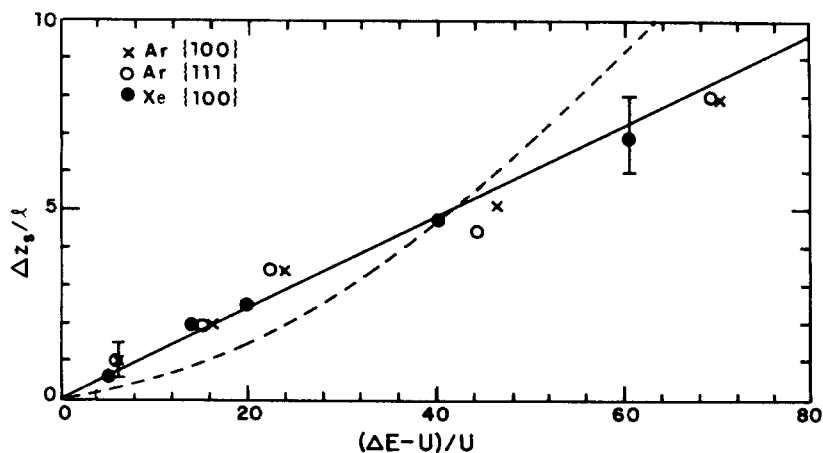

Fig. 2. Sputter depths, Δz_s, calculated by summing contributions from each layer in fig. 1 plotted versus energy input, ΔE, scaled to the sublimation energy, U. l is the layer spacing. The solid line, $\Delta z_s/l \sim 0.12\,(\Delta E - U)/U$ has a dependence like eq. (3). Dashed curve, $\Delta z_s = 0.01 l/((\Delta E - U)/U)^{5/3}$ has a dependence like eq. (4). The error bar indicates our estimate of the uncertainty in the yield.

tional radius equivalent to $\bar{\sigma}_d$ is of the order of the nearest neighbor distance this condition must break down, although apparently not so severely as to significantly change the dependence of Δz_s on ΔE. Finally, the observation of a few clusters implies that some non-linear processes are occurring.

The net energy distribution of the ejected Ar atoms is plotted in fig. 3 for a ΔE value of 1.0 eV. This is a sum of contributions from layers 1 through 4 and the two crystal orientations. The total distribution is normalized to 3.15, the average of the {100} and {111} yields. Also shown are predicted distributions from the collision cascade model,

$$Y(E) \propto E/(E+U)^{n+1},\tag{5}$$

normalized to 3.15. In eq. (5), E is the energy of the ejected atom and the expression applies in the present context for $(E+U) \leq \Delta E$. A value of n equal to 2 is appropriate for the collision cascade model of Thompson [14] and Sigmund [7]. This applies to sputtering due to direct collisional energy transfer to the atoms of the solid by an incident ion and the interaction cross section between the ejected atoms and the atoms in the solid is treated as a constant. A value of $n = 3$ has been shown to apply if the energy deposited by the ion becomes thermalized in a very narrow region along the ion track [15,16]. It also is appropriate to the cascade problem if the interaction cross section between atoms in the solid is linear in E [16]. However, the predicted distribution with $n = 3$ did not fit the data very well.

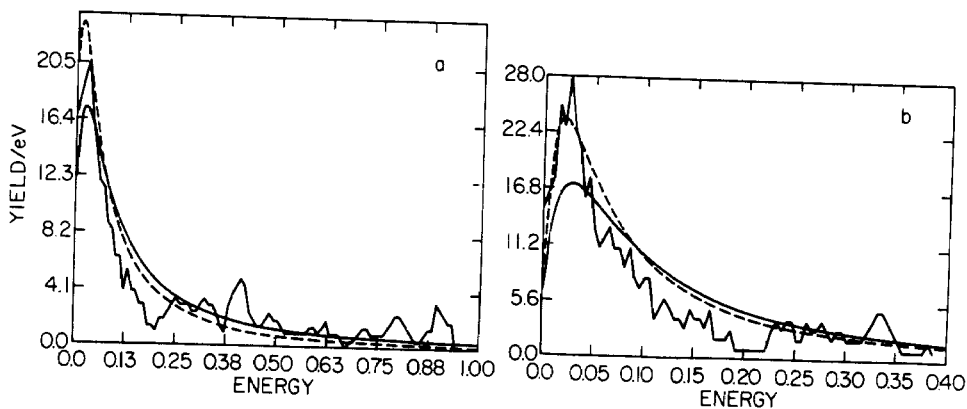The calculated distribution has a larger contribution near $E = (\Delta E - U)$

Fig. 3. Calculated energy distribution of ejected argon atoms for $\Delta E = 1.0$ eV and the potentials for argon in the text. The curves are constructed from contributions from layers 1–4 (≤ 7.5 Å.). The solid line is for $n = 2$ and $U = 0.061$ eV the bulk cohesive energy in the simulation, in eq. (5) and the dashed line is for $n = 2$ and $U = 0.043$ eV, the binding energy of a surface atom, in eq. (5). The energy distributions are normalized to 3.15 (see text). In (a) a resolution of 0.05 eV is used in producing our calculated spectrum while in (b) the resolution of 0.025 eV is used.

than do the distributions from eq. (5) as seen in fig. 3a. If there is an excitation near the surface, atoms can eject without many collisions occurring in the solid. In this case much of the original kinetic energy ΔE is removed by one ejected particle. This phenomenon is generally not observed in standard sputtering experiments, since the incident particle's initial momentum is directed into the solid, and several collisions are needed before a surface particle ejects. However for electronic sputtering (ablation, desorption) one *would* expect direct ejection from the top layers.

The calculated distribution does have a distinct low energy region, as seen in fig. 3b, corresponding to cascades which have evolved more fully. Remarkably, the shape and peak position change very slowly when ΔE is changed from 0.3 to 3.0 eV. The present calculations are similar to the assumptions involved in the derivation of the binary-collision cascade model which leads to eq. (5). However, there are two differences. We use simultaneous interactions between all atoms and not binary collisions, and we include a surface.

Although the calculated curve appears similar to those predicted by eq. (5), the peak widths are different. The calculated distribution is narrower, an observation commented on elsewhere [17]. It is difficult to determine the exact peak position due to the statistical uncertainty in the calculated distribution. In the analytic expressions for the energy distribution [14], the peak occurs at U/n and it is assumed that U should be the surface binding energy, although the heat of sublimation, a bulk quantity, is often used as a substitute value [7]. Shown in fig. 3b are the energy distributions as predicted by eq. (5) for $U = 0.043$ eV (averaged surface binding energy for $\{100\}$ and $\{111\}$ faces) and $U = 0.061$ eV (bulk cohesive energy). It is obvious that the better fit with the calculated distribution is obtained by using the value of the surface binding energy for U. This is sensible since virtually all of the ejected species are surface atoms and the yields in fig. 1 are of the order of unity. The analytic form for the energy distribution in eq. (5) predicts that at very low energies the yield is linear in E. Using a finer mesh than that in fig. 3b, the calculated results do appear to approach zero roughly linearly in E as $E \rightarrow 0$.

Although the energy spectrum presented here has been calculated for single crystals, we feel that the several features discussed are also applicable to electronic sputtering of amorphous and polycrystalline samples. In comparing the simulation with a sputtering experiment in which a number of monolayers have been eroded, the use of a surface binding energy may be questioned. That is, the cohesive energy of those ejected atoms which were, initially, not surface atoms must have been expended to remove them from the solid [7]. However, in all sputtering processes additional energy is available beyond that used in the ejection process. This energy is sufficient in these low cohesive energy systems to allow the lattice to adjust after removal of a surface atom. Therefore, as long as the yield per excitation event is low and ejections of neighboring atoms are separated by the lattice relaxation time, we suspect that

the effective binding energy per ejected surface atom will be lower than the bulk cohesive energy in these weakly bound systems even if many monolayers are removed. If the number of atoms ejected per event is large or neighboring events are close in time these results do not apply. Unfortunately, no energy spectra are available yet for electronic sputtering of Ar. However, spectra for the electronic sputtering of D_2O have been reported [18]. The enhancement expected at high energies, based on our description, is not observed, probably because of the large uncertainties in the data in the important energy region. At low energies the measured spectrum differs considerably from both our calculated spectrum and eq. (5). Whereas there was concern that this was an artifact of the breakdown in the cascade model for low energy events, it is seen from our simulations that it must be due to physical effects beyond those considered here (e.g., density of excited states) [18,16].

In fig. 4 we give the polar angle distribution of the ejected argon atoms integrated over azimuthal angles and averaged over the two crystal orientations. The distribution is sharply peaked at small angles and is very different from the $\cos \theta$ distribution of the standard collision cascade model and the thermal-spike model. This indicates that atoms are redirected on exiting due to interactions with other atoms in the same layer. Such peaked distributions have been observed before in simulations [19,20]. Dumke and co-workers [21] have measured the angular distributions of material ejected from a liquid gallium–indium eutectic alloy and have found polar distributions which fit a $(\cos \theta)^{2-3.6}$ form, but not as sharply peaked as the distribution in fig. 4. The calculated distributions are relatively insensitive to exciting energy and to
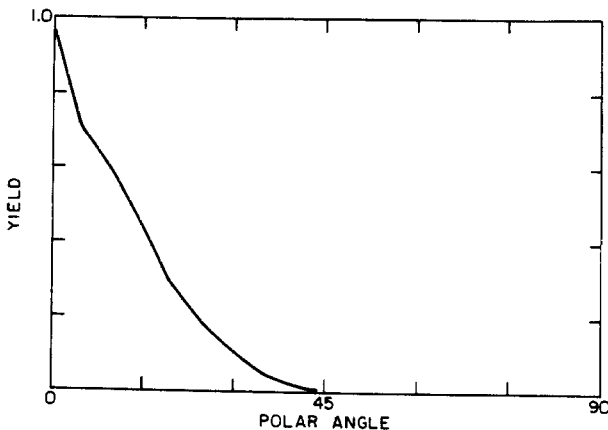

Fig. 4. Polar angle distribution of ejected argon atoms from both the {100} and {111} faces for $\Delta E = 1.0$ eV.

which crystal face is exposed. However, for the less dense surface layer {100} atoms exit with somewhat larger polar angles.

2.2. Excimer decay

In order to describe more accurately the sputtering produced following the repulsive decay of an excited dimer, we construct a dimer from two nearest neighbor atoms using all possible combinations of neighbors. These dimers are constructed ignoring the distortion of the rest of the lattice as discussed by Schwentner [5]. We assume that the excimer has already been formed, and hence the internuclear separation is less than the nearest neighbor distance in the crystal. We also assume that the dimer has decayed, perhaps radiatively [2,3,5], so that the atoms start interacting on the ground state potential curve but at a small internuclear separation. These atoms subsequently interact with other atoms in the solid via the ground state potential as in the previous calculations. (In fact at least one of the atoms may be in an excited state when highly excited eximers decay non-radiatively [2].)

To model the repulsive separation following the radiative decay of an excited Ar_2 to its ground state, two of the atoms in the crystal are placed a distance of 2.3 Å apart. This corresponds to the equilibrium separation of the $^3\Sigma_u^+$ excited state [5]. The potential energy of the pair of Ar atoms in the ground state is 1.9 eV based on the potential curves employed for the argon solid. At the time of the decay we allow the eximer to be oriented slightly off axis with the constraint that the total *additional* potential energy between the two atoms and the remainder of the crystal is < 0.2 eV. Thus the available energy is at most 2.1 eV. Because we are considering single crystals, two different configurations of nearest neighbor excimers are examined. In the first case the atoms are nearest neighbors in the same layer and in the second case they are in adjacent layers.

The calculated yields from the excimer study are given in fig. 4 as a function of depth z in the crystal. Overall trends are similar to those displayed in fig. 1. There is, however, a striking difference between the calculated yields when the atoms originate from the same layer and when they originate from adjacent layers. In the face centered cubic crystal nearest neighbor atoms in adjacent layers align in a close-packed row. This provides an extremely efficient collision mechanism (called a focuson in some of the older literature [22]) for transferring energy to the top atom in the row.

To construct Δz_s one sums these yields and multiplies by half the layer spacing. This gives $\Delta z_s \sim 6.0$ *l*, if one extrapolates to deeper layers than shown in fig. 5. This value of Δz_s is heavily influenced by aligned ejection process and, therefore, its use in an amorphous material and its relationship with our earlier results is hard to evaluate. A value of $\Delta z_s \sim 4$ *l* is not unreasonable if the focuson contribution in the deeper layers is attenuated. This range of

values of $\Delta z_s = 4\text{--}6$ *l* corresponds to an excitation of 1.7–2.5 eV of kinetic energy in one atom (cf. fig. 2), which is an amount of energy equivalent to that deposited in the excimer decay. Therefore, the yield is roughly independent of the details of depositing this energy.

The electronic sputtering of rare-gas solids is thought to be a result of the decay of excited dimers [2,3]. Yields for the electronic sputtering of solid argon [23,24] and xenon [25] have been measured for fast (~ 1–2 MeV) protons and helium ions. These yields have been found to vary somewhat non-linearly with the number of excitations per unit path length produced by the ions (λ^{-1} in eq. (2)). Ignoring this non-linearity and using eq. (2) with $l = n^{-1/3}$ for randomly oriented crystals, the results for argon would require excitation energies of the order of 3.5 to 8 eV to describe the yields if we use two thirds of the sublimation energy, 0.08 eV, for *U*. The maximum energy available after radiative decay of the first excited state is about 1.9 eV, as discussed above. Therefore, the measured yields imply (i) that additional kinetic energy depositions (e.g., repulsive electronic recombinations [2]) must be occurring [4], (ii) that the energizing events do not act separately [1], or (iii) that transport of excited state energy to the surface occurs so that λ^{-1} in eq (2) is enhanced near the surface. Reimann et al. [4] have used the results here to consider the role of
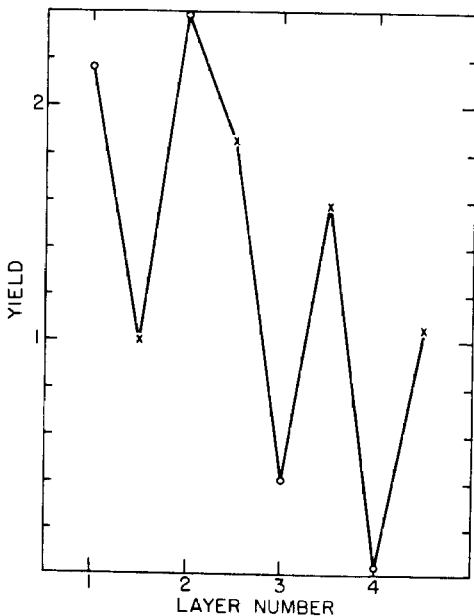

Fig. 5. Calculated yield versus depth of the center of mass of the excimer. The lines act only as a guide for the eye. The crystal is Ar{100}. (O) Both excimer atoms from the same layer. (×) Excimer atoms from adjacent layers.

free-exciton diffusion to the surface. The results available for solid xenon [25] are consistent with much small values of ΔE than for argon.

3. Conclusions

We present results here which can be used to interpret measurements of electronic sputtering of rare-gas solids. We found that the yields, integrated over depth, exhibit a nearly linear dependence on energy input, ΔE, a dependence which is similar to that for the binary-collision cascade model. These yields simply scale with the binding energy, U, of a surface atom, and they scale with the layer spacing, l, or the surface density, A^{-1} where $l = (nA)^{-1}$. The calculated yields are, not surprisingly, not at all consistent with a mini-thermal-spike model. We found that the form of the angular distribution of the ejected particles was insensitive to ΔE and the energy distribution is sensitive only at those energies close to $(\Delta E - U)$. Neither distribution is well described by the standard binary-collision cascade model. The polar angle distribution is strongly peak at small angles. The peak in the energy distribution is narrower than the binary-collision one and enhancements occur at energies close to $(\Delta E - U)$. However, the peak in the calculation does occur at an energy close to that predicted in the standard collision cascade model when we use the surface binding energy (approximately two thirds the bulk cohesive energy for these systems).

We compare the calculated yields for the repulsive decay of an excimer with calculations from a model in which individual atoms are given an equivalent amount of kinetic energy. Because the cascades from the two separating atoms in the excimer do not significantly interact we found that it is adequate to consider motion imparted to individual atoms in the solid.

We use these calculations to show that the observed yields for electronic sputtering of solid Ar cannot simply be explained from that energy released on decay of the lowest excited dimer. Measured deviation in the behavior of the yields and energy spectra from the calculations presented here are clear evidence for energizing processes other than those considered or for non-linear effects. These calculated data, therefore, can form a basis for interpretation of experimental results. Previously, the description of low energy ejection processes was sufficiently uncertain that reasonable interpretations could not be made. We expect, therefore, that these calculations will encourage sputtering measurements on these solids. Such experiments are a direct way of studying the energetic, non-radiative relaxation processes occurring in these insulating materials.

Acknowledgements

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