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Relative efficiencies of symmetrical and asymmetrical collisions in Ar^+-induced Al L_{23} Auger yields

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Computer simulation with the MARLOWE program is used to study the relative contributions of symmetrical and nonsymmetrical collisions to the Auger L_{23} emission from an aluminum solid target irradiated by low- (5 keV) and high- (50 keV) energy argon ions. The inner-shell ionization process is considered to take place when the distance of closest approach between two collision partners gets smaller than a certain threshold distance which does not have to be the same in both symmetrical (Al-Al) and asymmetrical (Ar-Al) collisions. After cross checking the emission threshold measurements, the theoretical and experimental estimates of ionization thresholds in gases, absolute Auger yield measurements, and computer simulations using the ionization threshold as a parameter, it was found that the threshold distances for the symmetrical and asymmetrical collisions are 0.36 and 0.44 Å, respectively. The relative contribution to Auger emission of both kinds of collisions was substantial, both with 5 and 50 keV incident energy.

INTRODUCTION

Auger electrons may be emitted when solid materials are irradiated by heavy ions. Most authors[1] working with low incident energies (from 1 to 10 keV) conclude that the dominating contribution originates with symmetric collisions (between two target atoms) as opposed to asymmetric ones (between incident ion and target atom). The first calculations carried out by computer simulation of collision cascades only took into account symmetrical events.[1(c),2] The following two types of experimental results led many authors to conclude that the symmetrical collisions played the dominant part:

(1) The primary energy threshold for Auger emission is independent of the nature of the incident ions.

(2) The distance of closest approach in head-on nonsymmetrical collisions (Ar-Al) at the threshold energy is larger than the sum of the radii of the orbitals involved in the electron promotion, as shown in the simplified correlation diagrams.[3]

While these arguments indicate that threshold emission is essentially due to symmetrical collisions, they do not show to what extent the relative nonsymmetrical contribution depends on the primary energy. An attempt at evaluating this quantitative contribution was made by Viaris *et al.*,[4] using Al-Cu and Al-Fe alloys with various aluminum concentration under argon irradiation. They concluded that the relative nonsymmetrical contribution is 0 at 5 keV incident energy and increases up to 18% at 15 keV. This conclusion conflicts with our simulation results shown below. In their interpretation, the aforementioned authors assume the following: that there is no surface enrichment effect due to preferential sputtering, and that the depth distribution of the sources of electrons does not depend on the Al concentration.

The first assumption is supported by surface monitoring with Auger electron spectroscopy (AES) while the second is not discussed and, consequently, the size of this contribution is not known.

In our previous study,[5] we found a parametric adjustment for our simulation model, which provides satisfactory agreement between calculated yields and absolute experimental measurements in various conditions regarding energy, incidence angle, and target surface. Although both symmetrical and nonsymmetrical collisions were taken into account in the ionization process, no particular attention was paid to their relative contributions. In this study, an additional step is made, i.e., the use of the threshold ionization distance as a parameter. Both low (5 keV) and high (50 keV) energy situations are investigated.

THE SIMULATION MODEL AND METHOD

The collision cascades are calculated with the MARLOWE (Ref. 6) program using the binary-collision approximation. We use the Molière potential[7] with Firsov screening lengths.[8] The crystal structure is generated by a cell containing the first and second neighbors that can be translated close to any collision site; this enables the selection of the next collision partner. To simulate the polycrystal a three-dimensional (3D) random rotation of the crystal structure is generated before each cascade. Suitable energy thresholds and cutoffs limit the target displacements and atomic motion. The inner-shell ionization criterion is based on a threshold distance r_c as suggested by Vrakking *et al.*[1(e)] By a method similar to that used by Andreadis *et al.*,[2] it was found that the emission and electron transport in the bulk are governed by a charac-

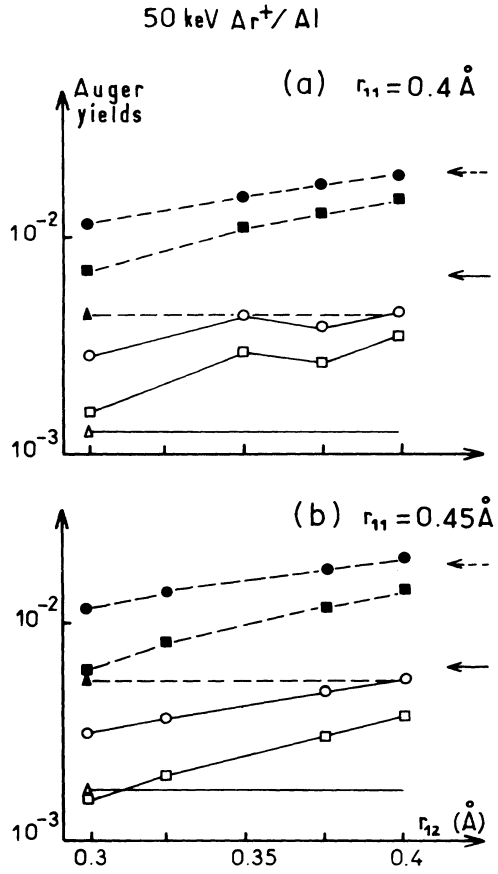

FIG. 1. Dependence of the Auger yields on the ionization-threshold distance for asymmetrical collision, r_{12}, with 50 keV incident energy and two values of the ionization-threshold distance for symmetrical collisions, r_{11}. Bulklike yield: ●, total; ←---, experiment; ▲, from symmetrical collisions; □, from asymmetrical collisions. Atomiclike yield: ○, total; ←, experiment; △, from symmetrical collisions; □, from asymmetrical collisions.

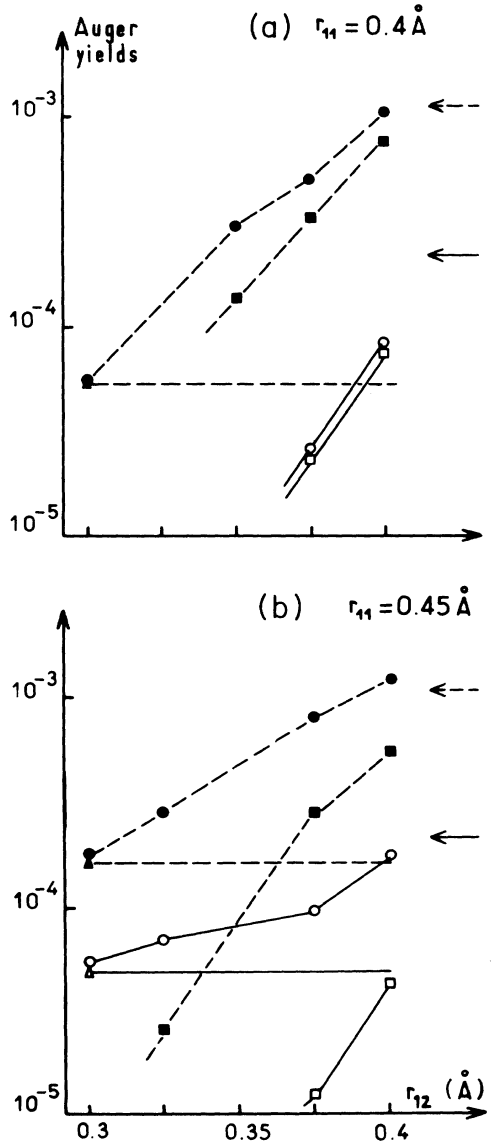

FIG. 2. Dependence of the Auger yields on the ionization-threshold distance for asymmetrical collisions r_{12}, with 5 keV incident energy. (For further details, see caption to Fig. 1.)

teristic relaxation time and an average inelastic mean free path. In addition, in order to be emitted, escaping electrons have to overcome a surface-energy barrier with planar symmetry and the relaxation of sputtered ions may be due either to an atomiclike process or a bandlike Auger transition through a Hagstrüm neutralization process.[9] The model parameters were given in earlier papers.[5]

To distinguish between contributions of symmetrical and nonsymmetrical collisions to both the bandlike and atomiclike emission efficiencies, extensive simulations are required to get satisfactory statistics, especially at low energy when the emission yields are small. It is not necessary to repeat each set of cascade calculations for each pair of ionization-threshold distances. Indeed, the MARLOWE code is designed in such a way that several distinct Monte Carlo procedures for ionization, electron transport, and escape can easily be run simultaneously in relation to the same atomic collision cascades. This way, the dependence of emission efficiencies on the threshold-ionization distances for a specific incident energy can be calculated immediately with the same statistically relevant set of atomic collision cascades. The results presented below were obtained with 380×10^3 cascades at 5 keV and 80×10^3 cascades at 50 keV.

RESULTS AND DISCUSSION

Figures 1 and 2 show the dependence of the bandlike and atomiclike Auger yields on the ionization-threshold distance r_{12} in asymmetrical Ar-Al collisions. The calculations are performed for two values of the ionization-threshold distance r_{11} in symmetrical Al-Al collisions. The total yield, symmetrical and asymmetrical contributions, are shown. The Auger yields relative to symmetrical Al collisions are, as expected, independent of r_{12} (see Figs. 1 and 2).

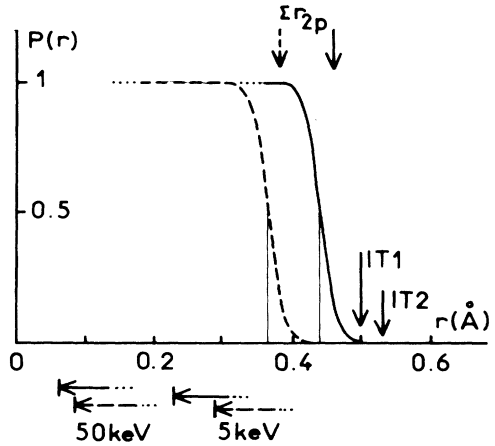

FIG. 3. Al $2p$ ionization probabilities $P(r)$ vs the distance of closest approach r: —— (Al-Al); — — — (Ar-Al). Distances of closest approach in head-on collisions for: ←, symmetrical (Al-Al) and ←– – –, asymmetrical (Ar-Al) systems. For the symmetrical system, the maximum energy is $4M_1M_2/(M_1+M_2)^2$ times the incident energy. IT1, IT2 ionization threshold values from Baragiola [Ref. 1(c)] and Vrakking [Ref. 1(e)], respectively.

In order to select a reasonable pair of values r_{11} and r_{12} for the ionization threshold distances, one has to rely on (i) energy-threshold measurements from solid [e.g., Vrakking,[1(e)] 750 eV and Baragiola,[1(c)] 900 eV]; (ii) the experimental and theoretical estimates in gases which are available for the Ar-Si system.[10] A scaling rule based on the values of the involved orbital radii,[11] allows us to deduce the threshold distances for the Ar-Al and Al-Al pairs. These two requirements could be met when the relation between distance of closest approach and the collision relative energy are governed by the Molière potential with the Firsov screening length corrected by the formula suggested by O'Connor.[12] It is assumed that the dependence of the ionization probability on the internuclear distance, r, follows Fermi's law,

$$P(r)=\left[1+\exp\left[\frac{r-r_c}{d}\right]\right]^{-1},$$

where r_c is the threshold distance and $d=0.01$ Å.[13] In addition to these requirements, the calculated Auger yields must reproduce the absolute experimental values. The highest consistency of the simulation model is found for the following ionization threshold distances:

$r_{12}=0.36$ Å (r_c for Ar-Al collisions)

$r_{11}=0.44$ Å (r_c for Al-Al collisions) .

When these results are taken into account, the ionization-probability dependence on the colliding-particle distance r can be drawn for Ar-Al and Al-Al pairs (Fig. 3). The ionization probability changes abruptly from 0 to the maximum value for a particular value of the internuclear distance; this justifies the use of a step function in the simulation model.

As it can be seen in the same figure, the distance of closest approach in head-on collisions at maximum energy is about five times smaller than the ionization-threshold distance at 50 keV incident energy, while it is even less twice at 5 keV. This is the reason why, as shown in Figs. 1 and 2, the Auger emission yield is not very dependent on the threshold distance at high incident energy while it is critically so at low energy.

With such an adjustment we found a good agreement between the absolute experimental Auger yields and the calculated values for 50-keV projectiles (for both bulklike and atomiclike Auger emission the difference is less than 10%). At 5 keV, the experimental values are higher than the calculated yields (Figs. 1 and 2). In view of the low Auger yields at this low incident energy, it is difficult to decompose the Auger spectra (in atomiclike and bulk components), this involving greater uncertainty in the experimental estimations. On the other hand, the use of a stronger screening of the interaction model potential[12] should result in a reduction of the distance of closest approach. Consequently the calculated low-energy Auger-emission yield could also be increased, the yield for high incident energies remaining almost unchanged.

The following contributions to Auger emission are calculated (in percent):

	Asymmetrical collisions	Symmetrical collisions
5 keV	40	60
50 keV	67	33

Thus it turns out that, at low energy, the contribution of nonsymmetrical collisions to the Auger emission is more significant than is often assumed. Therefore, no contribution may be neglected at low or high energy.

ACKNOWLEDGMENT

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