Study of atomic excitation in sputtering as a function of the projectile incidence angle

P. Larsen and E. Veje
Physics Laboratory II, H.C. Ørsted Institute, Universitetsparken 5, DK-2100 Copenhagen, Denmark
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Solid targets of elemental beryllium, boron, magnesium, and gold have been bombarded with 80-keV Ar^+ ions. Intensities of photons emitted from sputtered particles have been measured as functions of the angle of incidence of the projectile. Generally, the excitation intensities increase with increasing angle of incidence, but a few levels show much stronger angular variations than the majority of levels do. The results are discussed. It is concluded that whereas the common increase in excitation is caused by the general increase in the number of swift, sputtered atoms, a few levels are in addition selectively excited through molecular-orbital electron-promotion processes taking place in binary collisions.

I. INTRODUCTION

This work is a continuation of our attempts[1-4] to systematically study atomic excitation in sputtering processes. In our previous work,[1-4] we have bombarded various targets at normal incidence. Here we report and discuss results obtained by bombarding targets of solid elemental beryllium, boron, magnesium, and gold at different angles of incidence of the projectile. Hitherto, not only our work but also the vast majority of similar studies carried out in other laboratories have been performed at a fixed angle of incidence, which most frequently has been chosen to be 90°. Only in one case has the angular variation been studied.[5] That was for magnesium bombarded with 4-keV Ne^+ and Ar^+ ions, and only the excitation of one final atomic level, namely the $3p\ ^1P$ level in neutral magnesium (Mg I), was studied.[5] That work was carried out under such vacuum conditions that the target surface was covered with an oxide layer. It was found that the level-excitation probability per incoming projectile increased rapidly with increasing angle of incidence, and a similar behavior was observed for projectile excitation.[5]

In the solid target, the atoms are initially held together with bonds which are produced by the valence electrons. For Be, Mg, and Au, the bonds are metallic; thus the valence electrons are initially delocalized. Boron has a covalent bond for which the valence electrons are more localized.

During the sputtering process, valence electrons will rearrange themselves from initially being describable by some solid-state wave function, to finally being described by some atomic eigenstate wave function. Atomic excitation in sputtering events is the outcome of the rearrangement of the valence electrons from the solid to the free atom (or ion). This has been described in Refs. 3 and 4 in which references to previously proposed models are also given. Such references shall not be repeated in this paper.

In Refs. 3 and 4, as well as in previous works, only the case of normal projectile incidence to the surface was considered. When the target surface is tilted, the total sputtering yield will increase because that fraction of the projectile energy, which is deposited close to the surface, is increased.[6] Consequently, one may expect from this that the total number of excited atoms emitted per incoming projectile will increase monotonically with the angle of incidence, at least until very large angles, where a substantial fraction of the projectiles will become reflected at the surface. But, in addition, when the angle of incidence of the projectile is increased, the total number of sputtered particles emitted with relatively high velocity or kinetic energy will increase.[6] Such particles result from fairly violent collisions between projectiles and target atoms (*P-T* collisions), possibly combined with subsequent, recoil-induced collisions among target atoms (*T-T* collisions). Intuitively, one may expect that the increase in the total number of particles sputtered with relatively high velocity, caused by increasing the angle of incidence, may lead to an increase in the total number of excited atoms emitted per incoming projectile. Especially for highly excited atoms, where a relatively large amount of transfer from kinetic energy to excitation energy is involved, one may expect an increase in excitation over and above the increase in total sputtering yield.

When the target surface is tilted, one more feature may show up. The number of violent *P-T* and *T-T* collisions will essentially be independent of the tilt angle, but target atoms set into large relative motions will predominantly move in a forward direction (forward with regard to the initial projectile motion). This means that the number of atoms having experienced a violent collision immediately before escaping from the solid will increase with increasing angle of incidence. Thus possible excitation effects which can be related to violent binary collisions may be expected to manifest themselves especially at large angles of incidence. Such binary-collision-induced excitations can be expected to be very specific, since it is well known from gas collisions at fairly low kinetic energies that level excitation is very selective.

II. EXPERIMENTAL

The accelerator[7] and also the experimental equipment[2,4] have been described before. Therefore only the essentials are mentioned here.

Solid targets of elemental beryllium, boron, magnesium, and gold (high-grade purity, 99.99%) were bombarded with 80-keV Ar^+ ions. In addition, the gold target was also bombarded with 80-keV Xe^+ ions. The residual gas pressure in the target chamber was around 10^{-9} Torr$\simeq 10^{-7}$ Pa, so that once the target surface had been cleaned because of the heavy ion bombardment, it stayed clean. This was confirmed by a strict linearity between

signal and beam intensity and also by a high degree of reproducibility of the signals.

Photons emitted from sputtered particles were analyzed with a 1-m scanning monochromator, 170—1000 nm. The observation direction was perpendicular to the beam axis and also perpendicular to the target normal, see Fig. 1. In other words, the angle of incidence of the projectiles was changed by rotating the target; the axis of rotation being the same as the optical axis of the observation direction. A lens was placed so that the distance between the lens and the entrance slit of the monochromator equaled the focal length of the lens. Therefore, the observation volume was a cylinder with a radius equal to the radius of the lens (2 cm). Such a fairly large observation volume ensured that most of the excited, sputtered atoms decayed within the observation volume, independent of the lifetime of the excited state in question.

In measurements such as the present, in which sputtering is studied with optical spectroscopy, the front surface of the optical component which is placed next to the target will gradually become coated with sputtered material. Such a coating will change the transmission, making the overall detection efficiency time dependent. Therefore, it is essential to keep such a surface contamination as small as possible and to check the transmission repeatedly, to allow for possibly necessary corrections. The material deposited on the front surface of the first optical component (in our case, the window of the target vacuum chamber, cf. Fig. 1) will be as small as possible, with a detection geometry similar to the one used here, because, as described above, the observation direction was in the horizon of the target for all angles of incidence. In the arrangement used here, we checked the transmission frequently and observed essentially no change in transmission during the period of all of the measurements.

For all angles of incidence, the beam current was measured in front of the target by placing a properly designed Faraday cup in the beam path[2] so that the relative level excitation per *incoming* projectile could be determined (and thus *not* per beam-current or beam-charge unit measured on the *target*). In addition, the total current to the target was recorded during each measurement. This allowed for determination of the secondary-electron emission yield, as described in Ref. 8. The secondary-electron emission yields were highly reproducible from day to day and agreed closely with data published by others. This also indicated that the surfaces were freed of oxygen.

For all spectral lines studied here, the target was rotated clockwise as well as counterclockwise, yielding two data sets of the change of excitation as a function of the angle of incidence, as well as two corresponding data sets of the secondary-electron emission yield versus the angle of incidence. The secondary-electron emission-yield curves were very smooth and appeared to be symmetric around normal incidence, as expected (within the total uncertainty of 5% or less). This indicated that the angular scale was free of a possible error in the position of the zero point. The optical signals, obtained by turning the target clockwise and counterclockwise to the same angle of incidence, were found to deviate slightly and systematically from each other. Such small deviations can be explained from nonuniformities of the photocathode and/or the monochromator grating and mirrors, making the resultant detection efficiency slightly position dependent. The data shown here resulted from averaging the signals obtained at the two different settings of the same angle of incidence.

The results are shown in Figs. 2—7. The data points belonging to excitation of different levels are on different scales. The curves drawn in Figs. 2—7 are guides for the eye only. All the data except those for the $3p\ ^2P$ level in

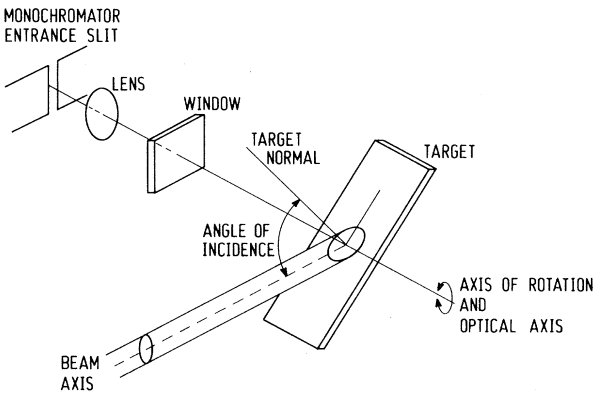

FIG. 1. Detection geometry shown schematically. Note that the axis of rotation is the same as the optical axis.

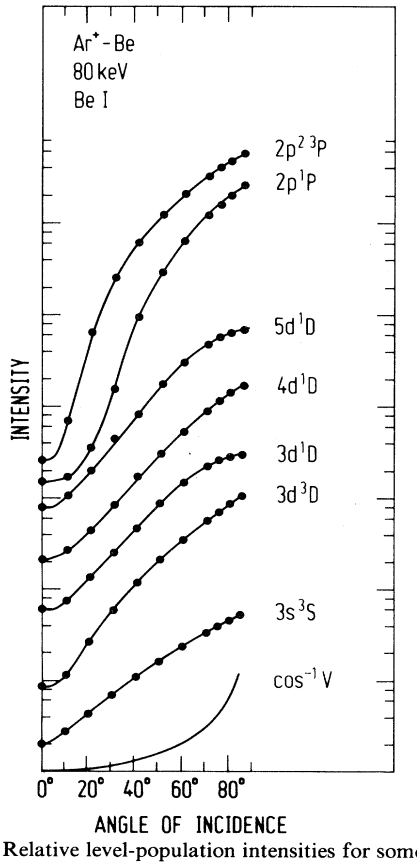

FIG. 2. Relative level-population intensities for some levels in neutral beryllium (Be I) plotted semilogarithmically vs the angle of incidence of the projectile.

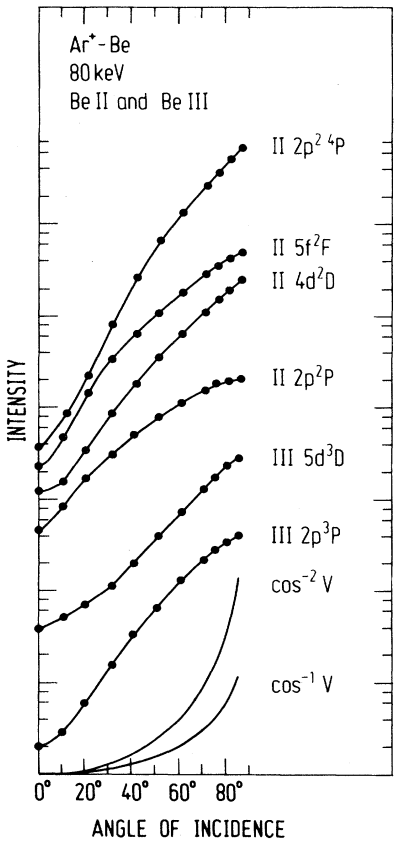

FIG. 3. Relative level-population intensities for some levels in singly and doubly ionized beryllium (Be II and Be III) plotted semilogarithmically vs the angle of incidence of the projectile.

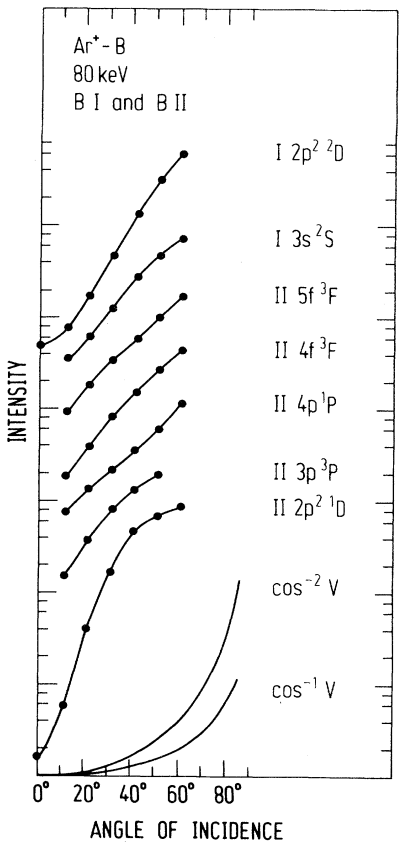

FIG. 4. Relative level-population intensities for some levels in neutral and singly ionized boron (B I and B II) plotted semilogarithmically vs the angle of incidence of the projectile.

Mg II are uncorrected for cascade repopulations. We have recently published relative level populations for Be, B, and Mg, obtained at normal incidence.[3,4] There[3,4] we observed that of all levels studied, essentially only the $3p\ ^2P$ level in Mg II was substantially repopulated from cascades (for a detailed discussion of the cascade problem, the reader is referred to Ref. 4).

Concerning cascade contaminations, it can be added that for each element and ionization state, many levels show similar increases in excitation with increasing angle of incidence. This implies that for such levels, the fraction of level repopulation does not vary with the angle. Thus the cascade uncorrected curves can on relative scales represent curves corrected for cascade repopulations.

For boron the level-excitation efficiencies are very small at normal incidence, as discussed in Ref. 3. Therefore, data obtained at normal incidence were generally so uncertain that they are not included in Fig. 4.

III. RESULTS

For all elements and levels studied here, we observe that the level excitations increase with the increasing angle of incidence, cf. Figs. 2—7, at least up to very large angles of incidence where there may be a maximum, see especially Fig. 7. However, different levels and different elements show quite different variations. Especially, the curve for the $3p\ ^2P$ level in singly ionized magnesium (Mg II) is very flat, cf. Fig. 6, in contrast to, e.g., the curve for the $3d\ ^1D$ level in Mg I (Fig. 5) as well as some of the beryllium and boron curves. On the other hand, many of the curves for gold (Fig. 7) are fairly similar to many of those for the lighter elements.

IV. DISCUSSION

A. Total sputtering yield

The total sputtering yield will increase with increasing angle of incidence, simply because the total amount of energy deposited close to the surface by the projectile will increase.[6] The change in the total sputtering yield is not well known for the combinations of projectiles and targets studied here, but there are reasons to believe[6] that the total relative sputtering yield will increase somewhere inbetween $\cos^{-1}v$ or $\cos^{-2}v$, v being the angle of incidence. Therefore, each of the figures (2—7) contains a $\cos^{-1}v$ curve at the bottom. In some of the figures $\cos^{-2}v$ is also included. It is easy to see that all of the experimental curves given in Figs. 2—7 deviate from the cosine curves. The deviations are not only qualitative, but also quantitative, the shapes of the cosine curves being very different from the experimental curves. Thus the increase in level excitation with the increasing angle of incidence is not ex-

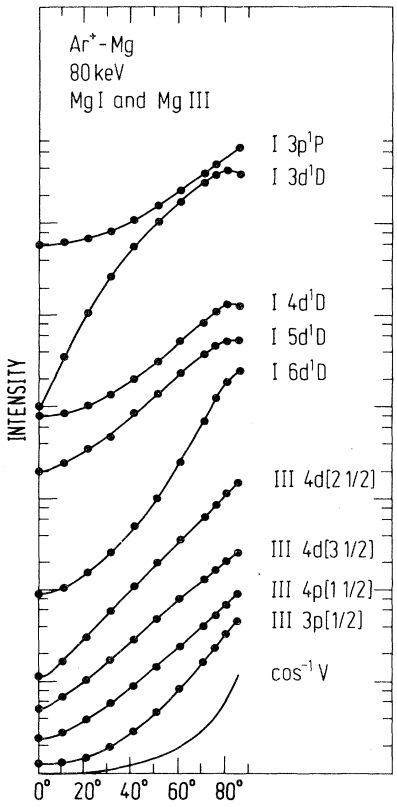

FIG. 5. Relative level-population intensities for some levels in neutral and doubly ionized magnesium (Mg I and Mg III) plotted semilogarithmically vs the angle of incidence of the projectile.

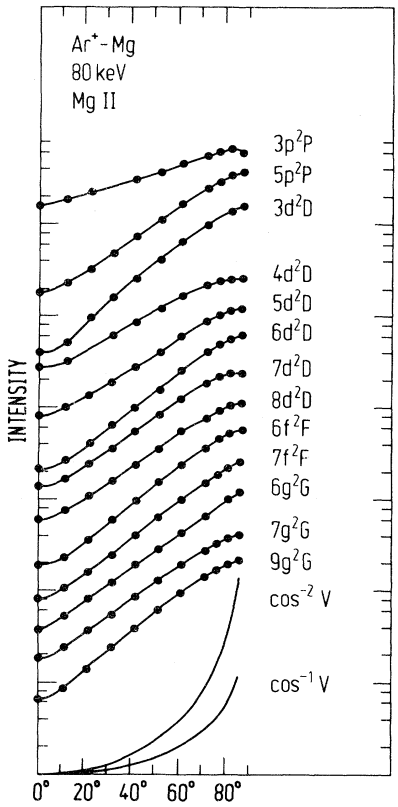

FIG. 6. Relative level-population intensities for some levels in singly ionized magnesium (Mg II) plotted semilogarithmically vs the angle of incidence of the projectile.

plained solely from the increase in total sputtering yield. Naturally, part of the increases observed may be related to the increase in the total sputtering yield, but also other phenomena appear.

B. Binary collisions

For beryllium and boron we observe (Figs. 2—4) that the excitation for final terms involving one or two $2p$ electrons increases very steeply with increasing angle. Other levels have flatter curves. This selective excitation of terms involving a $2p$ configuration can be understood in terms of recoil-induced, binary atomic collisions among target atoms (T-T collisions), as described in the following.

A natural starting point for discussing binary atomic collisions at such low kinetic energies as those of relevance here is the molecular-orbital (MO) electron-promotion model, as outlined by Barat and Lichten,[9] because of its relevance to binary atomic collisions in gases.[9,10] According to the MO model, when the two atoms come close together during the collision, their electrons will adjust their motion from one-center atomic orbitals to two-center molecular orbitals. Later in the collision, when the atoms separate, the electrons will readjust their motions back to atomic orbitals. Excitation may occur if, during the course of the collision, an empty molecular orbital comes close in energy to an electron-carrying orbital. Then an electron may be transferred to the originally empty orbital and stay there. Possible final level excitations can be estimated qualitatively from the so-called correlation diagram which is a schematic representation of how the levels in the separated atoms correlate to the levels in the united atom when the internuclear distance is reduced during the collision.

The correlation diagram of relevance for beryllium and boron T-T collisions are readily constructed from rules given by Barat and Lichten,[9] namely that (i) the number of nodes in the radial part of the wave function (equal to $n-l-1$) is conserved during a collision, and (ii) the Pauli principle is obeyed.

A correlation diagram for beryllium and boron T-T collisions is shown in Fig. 8. Molecular orbitals which carry electrons in the incoming channel are drawn with solid lines, whereas initially empty orbitals are given with dashed lines. The valence electrons have been disregarded because they are initially being described by some solid-state wave function. According to Fig. 8, one or both of the $1s$ electrons may be transferred to the $2s$ or $2p$ final atomic states in T-T collisions, but not to levels with principal quantum number $n>2$. The electron promotion from the $1s$ to the $2s$ or $2p$ configuration is combined with a $1s$ vacancy creation, but that vacancy need not be located in the same atom as that in which the promoted electron finally will be localized. Thus we learn from Fig. 8

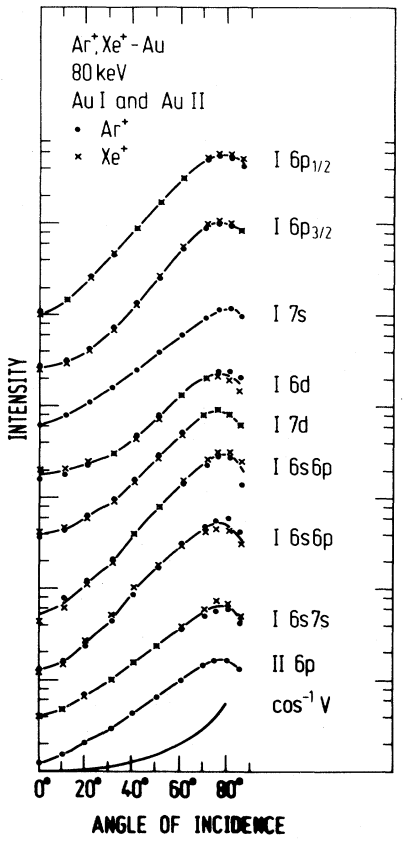

FIG. 7. Relative level-population intensities for some levels in neutral and singly ionized gold (Au I and Au II) plotted semilogarithmically vs the angle of incidence of the projectile. Data obtained with the use of Ar^+ projectiles are shown with closed circles and data with Xe^+ projectiles are given with crosses. The Xe^+-induced data have for each level studied been normalized to those obtained with Ar^+ projectiles.

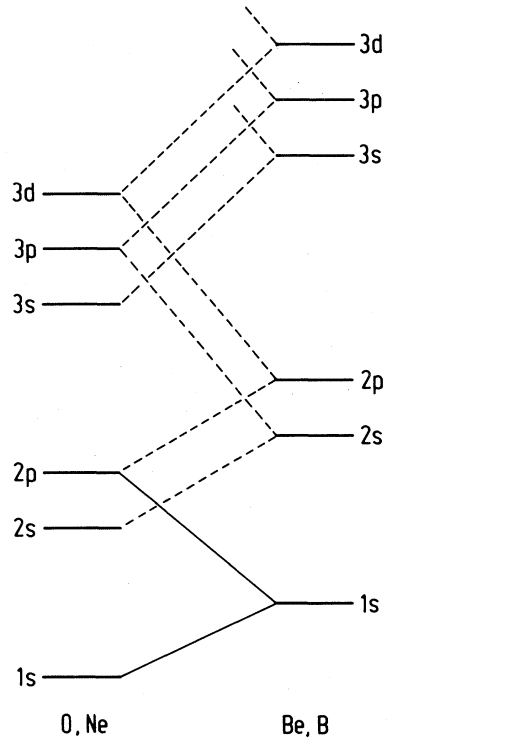

FIG. 8. Diabatic MO diagram for beryllium and boron T-T collisions shown schematically. On the right-hand side of the figure are shown some terms of the separated beryllium or boron atoms (not to scale), and the corresponding united atoms terms (oxygen and neon) are given on the left-hand side (not to scale). The terms of the separated and the united atoms have been connected by straight lines to indicate how the levels correlate during a binary collision. Correlations for terms carrying electrons in the incoming channel are shown with solid lines, whereas originally empty orbitals are given by stipulated lines. See also the text.

that for beryllium and boron, recoil-induced T-T collisions lead to selective excitation of the $2s$ or $2p$ levels, and that the selective $2s$ or $2p$ level excitation may or may not be combined with a $1s$ vacancy creation. Of the two electron-promotion possibilities, only the one that leads to the $2p$ configuration can be observed optically, because the $2s$ configuration is the ground state. Thus in the present work only $2p$ level excitations can be observed.

Now that the *excitation* process has been described, the *survival* of atomic excitation must also be considered, as follows. The total amount of $1s$-$2p$ electron promotions *created* from recoil-induced T-T collisions is presumably mostly independent of the angle of incidence of the projectile. Many of the created electron promotions are destroyed in subsequent T-T collisions in the solid, which may be soft collisions, because there are several reasons to believe that excited atomic species cannot exist inside the solid.[1–4] Only that fraction of the total amount of T-T collision-induced excitation, which initially is located at or very close to the surface, may survive and show up in sputtering. Therefore, when the target surface is tilted, the resultant amount of T-T collision-induced excitation emanating from the surface will increase. The increase will be large, especially at large angles of incidence, because target atoms recoiling at some small forward angle may then escape from the surface. Note that the survival probability discussed here has nothing to do with radiationless deexcitations. The latter will be discussed later.

Thus, to recapitulate, the very steep curves seen for the excitation of the $2p$ and $2p^2$ configurations in Be and B (see Figs. 2–4) can be understood from a selective $2p$ MO electron-promotion process induced by T-T collisions, combined with an increase in escape probability with increasing angle of incidence.

After having discussed T-T collisions, also collisions between projectiles and target atoms (P-T collisions) have to be considered. The correlation diagram of relevance for the Ar-Be and Ar-B collisions is given schematically in Fig. 9. Binding energies are not on a linear scale, but the levels are ordered correctly, based on binding energies calculated from relativistic Hartree-Fock-Slater atomic wave functions.[11] It is readily seen from Fig. 9 that P-T collisions do not lead to excitation of Be or B.

The MO correlation diagram of relevance for magnesium T-T collisions is given in Fig. 10. Following ideas similar to those outlined during the discussion of the Be

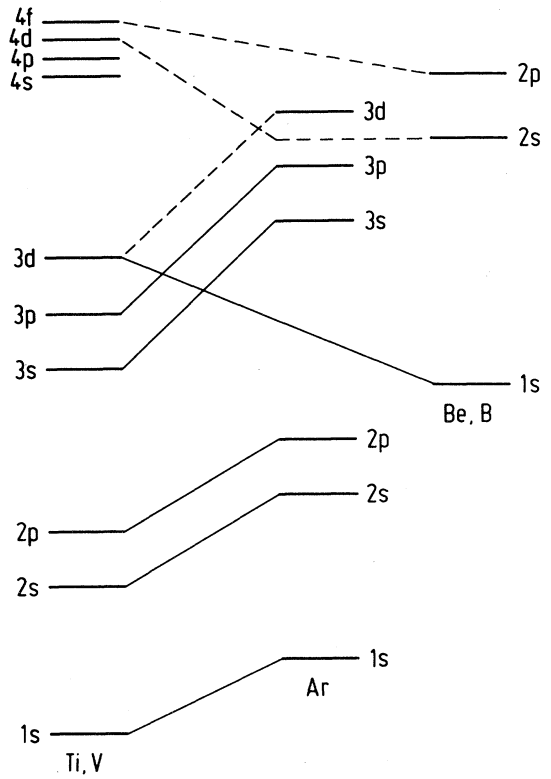

FIG. 9. Diabatic MO diagram for argon-beryllium and argon-boron P-T collisions shown schematically. See also the caption to Fig. 8 and the text.

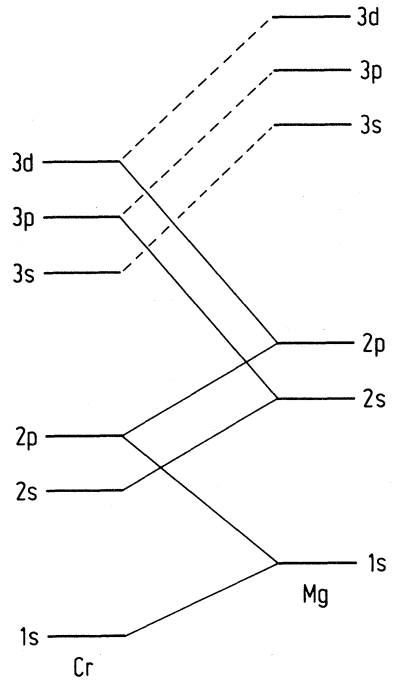

FIG. 10. Diabatic MO diagram for magnesium T-T collisions shown schematically. See also the caption to Fig. 8 and the text.

and B results, it is concluded that recoil-induced T-T collisions among the Mg atoms may result in preferential excitation of the $3s$, $3p$, or $3d$ levels in magnesium (of which the $3s$ cannot be observed optically), so that for Mg, steep angle-of-incidence curves might be expected for levels involving $3p$ or $3d$ electrons. From gas collisions[12] it is well established that among levels of the same principal quantum number, but with different values of the orbital angular momentum quantum number, the d levels will be preferentially populated, much above the p levels. Taking these facts together, T-T collision effects should be expected to be more pronounced for the d levels. Indeed, of all of the magnesium curves observed, those of the $3d$ levels are the steepest ones, see Figs. 5 and 6. Especially, the Mg I $3d\ ^1D$ curve is steep (Fig. 5), in agreement with expectations from the MO diagram. However, in addition, the curve labeled $6d\ ^1D$, Mg I, is also very steep, much steeper than the $4d$ and $5d$ curves for Mg I. This is an interesting observation. The $6d\ ^1D$ level in Mg I is known to have a high degree of configuration mixing with doubly excited valence-shell terms of configuration $3d^2$, and it is undoubtedly this $3d^2$ nature which makes the curve much steeper than the $4d$ and $5d$ curves. We have recently found[4] that the $6d\ ^1D$ level in Mg I has an anomalously strong level population in sputtering at normal incidence. Also, that was explained in terms of configuration interactions, in agreement with the above-mentioned explanation of the peculiar behavior of the $6d\ ^1D$ curve (Fig. 5).

The Mg II $3p\ ^2P$ curve is surprisingly flat (Fig. 6). As seen from the Mg-Mg MO correlation diagram (Fig. 10), also the $3p$ level can become excited in T-T collisions. However, the $3p$ level in Mg II has a binding energy larger than the work function of the solid Mg metal. This makes it possible to populate the $3p$ level in Mg II through a resonant charge-transfer process[1,4] in contrast to Mg I levels. Already at normal incidence, the population of the $3p\ ^2P$ term in Mg II is remarkably large,[1,4] as explained in terms of resonant charge transfer.[1,4] The process leading to the strong population at normal incidence will naturally also be active at other angles, and because of its efficiency, it will blur contributions from other possible mechanisms such as those induced by T-T collisions, which in themselves may have some strong angular dependence. Similarly for beryllium, the Be II $2p\ ^2P$ curve is flatter than the other $2p$ curves of beryllium. This is because the Be II $2p\ ^2P$ level can become excited through resonant electron transfer.[1,4] Thus the relatively flat curves for the $3p\ ^2P$ level in Mg II and the $2p\ ^2P$ level in Be II confirm the ideas proposed in Refs. 1—4, namely that excitation in sputtering at normal incidence largely results from electron pickup when the sputtered particle leaves the solid. The ideas[1-4] shall not be repeated here in detail. As for beryllium and boron, P-T collisions cannot explain any selective outer-shell excitation in magnesium (see Fig. 11).

There is little to say about the Mg III curves given in Fig. 5. The $2p$ core vacancy may become created in T-T collisions as well as in P-T collisions. The further the target is tilted, the larger the probability will be that an atom with a core vacancy escapes from the surface. At the surface, the escaping atom may pick up an electron from the valence band, resulting in sputtering of excited Mg^{2+}

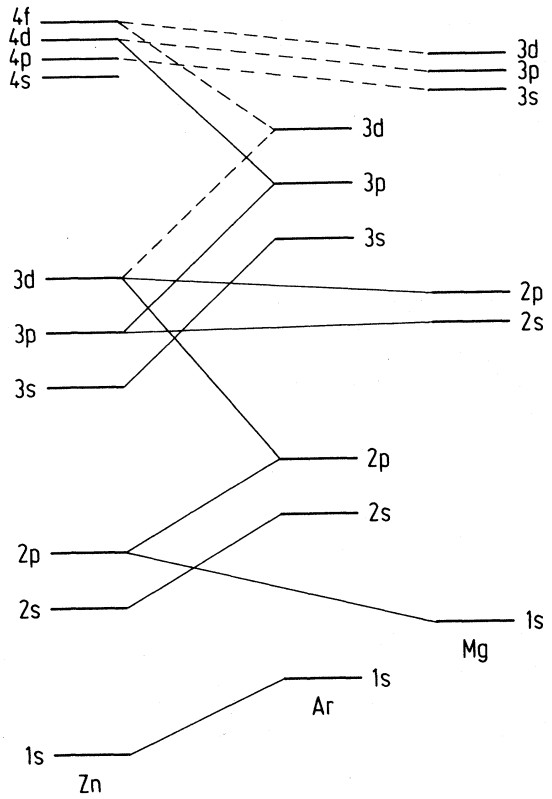

FIG. 11. Diabatic MO diagram for argon-magnesium *P-T* collisions shown schematically. See also the caption to Fig. 8 and the text.

(Mg III). However, so many possible steps are involved in the creation of Mg III that essentially no information can be deduced from the Mg III curves.

For gold the MO correlation diagrams for *P-T* and *T-T* collisions are uncertain, due to the large number of electrons. However, *T-T* collisions will presumably lead primarily to the promotion of the 5*d* electrons because these are the outermost of the inner-shell electrons. The 5*d* electrons will correlate to the 6*f* orbital if the rule of conservation of the number of nodes in the radial part of the wave function is obeyed. Thus *T-T* collisions will lead to a selective excitation of the 6*f* level. But the position and the transitions from the 6*f* level in Au I are unknown. This is very unfortunate, and leaves gold as a fairly uninteresting case here. However, it is interesting to see that several Au curves are steeper than some of the Mg curves, although Au is very heavy, and therefore, the majority of sputtered Au atoms will be very slow.[13]

C. Increase in kinetic energy

Except for the few steep curves discussed in the preceding section, the curves for each element are fairly similar. The steadily increasing common curves undoubtedly reflect that when the angle of incidence is increased, the total number of swift, sputtered atoms will increase, leading to an increase in total excitation.

The curves belonging to the *d* terms in Mg II are interesting from systematical trends (see Fig. 6). The $3d\,^2D$ curve is steep, showing the contributions from *T-T* collisions at large angles, as discussed above. The $4d\,^2D$ curve is much flatter, and then, the curve shape changes gradually, showing a steady increase in steepness, but even the 8*d* curve is flatter than that of the 3*d* level. This clearly indicates that a special mechanism, namely the *T-T*-induced MO promotion is active for the 3*d* level, as already discussed. The other *d* levels as well as all other Rydberg levels, in Mg II are excited through nonresonant electron pickup processes when the sputtered atom leaves the surface, as described in Refs. 1—4; see especially Ref. 4. All these levels have binding energies smaller than the work function of solid magnesium. Therefore, electron pickup will be nonresonant for these levels. The higher the excitation energy of a particular level is, the larger the amount of energy transferred from kinetic energy to excitation energy will be, or, in other words, the further the electron pickup process will be from resonance. When the target is tilted, the number of swift, sputtered particles is increased, and the highly excited atoms presumably are predominantly atoms sputtered with a relatively large velocity or kinetic energy. Thus it is understandable that the curves increase in steepness with increasing value of the principal quantum number *n*, for levels within the same term series, as found for the $nd\,^2D$ levels (and also other levels) of Mg II, Fig. 6.

D. Radiationless deexcitation

Radiationless deexcitation of a sputtered, excited atom in the vicinity of the solid surface has been discussed several times in the literature; see, e.g., Ref. 4 and references given therein. The necessary condition for a one-electron nonradiative deexcitation to occur is that the upper level of the excited, sputtered atom energetically coincides with an empty state in the conduction band of the solid metal. If this condition is fulfilled, the excited electron may jump back from the atom to the metal. This is a resonant ionization process. Thus radiationless deexcitation is in principle possible, e.g., for the high-lying levels of Mg II studied in this work, but not for the $3p\,^2P$ term in Mg II. The probability for radiationless deexcitation increases with the time the excited atom spends in the vicinity of the surface, and also with the geometrical size of the excited orbital. In other words, radiationless deexcitation can be expected to influence the results, i.e., acting as a drain of the level excitation, predominantly at large angles of incidence, because then the majority of the sputtered atoms will leave the surface at some large angle to the surface normal so that the excited atoms remain in the vicinity of the surface for relatively long times. Also, the drain should be more pronounced for highly excited levels because of their geometrically large sizes. However, returning to Fig. 6, there are no indications of radiationless deexcitations draining the level excitations at large angles of incidence and primarily occurring for highly excited levels. Actually, the curves for the $6,7,8d\,^2D$, the $6,7f\,^2F$, and the $6,7,9g\,^2G$ levels in Mg II are very similar, cf. Fig. 6. These findings do not necessarily exclude the existence of radiationless deexcitations, but the results indicate that any possible drain caused by radiationless deexcitation is

counteracted by the increase in excitation probability caused by the general increase in the number of swift, sputtered atoms. Thus we conclude that we have failed to observe any effects due to radiationless deexcitations, in agreement with conclusions of previous works.[3,4,14]

V. CONCLUSIONS

We have studied atomic excitation in sputtering as a function of the projectile incidence angle. Generally, the excitation yields increase with increasing angle of incidence. This is explained from a general increase in the number of swift, sputtered atoms when the target is tilted. In addition to this general increase, binary collisions may at large angles of incidence lead to strong excitation of selected levels. Such processes can be described in terms of MO electron-promotion processes. For a level such as the $3p\,^2P$ term in Mg II, the angular variation is not pronounced. This is because the level is already strongly excited at normal incidence, due to resonant electron pickup. Possible influences from radiationless deexcitation processes are discussed, and it is concluded that we have failed to observe any.

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