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COMMENT

Comments on the production of autoionizing states in the scattering of argon ions from a magnesium surface

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Abstract. This paper comments on the results of recent studies of Ar^+ scattering on Mg, as well as Al and Si, surfaces by Blum *et al* and Nixon *et al* . The strong excitation of Ar^{**} $3p^4 4s^2$ states is reported in these works, especially for low (1 keV) collision energies. This result contradicts our earlier work and the work of other authors. We re-investigated Ar^+ scattering but did not find any evidence of Ar^{**} state production. However, Ar^{**} state production is observed in Ar^{++} collisions and we conclude that a contamination of the Ar^+ beam with Ar^{++} in the work of the above authors is responsible for their observations. We also comment on the identification of structures due to excited sputtered Mg.

In a recent series of experiments we reported upon a study of excited state production in He, Ne and Ar neutral, singly and, in some cases, doubly charged ion scattering on Na, Mg, Al and Si surfaces [1–6]. We investigated ion scattering and performed measurements of emitted-electron spectra. We were thus able to show that in He and Ne ion scattering production of autoionizing states such as He^{**} $2s^2$ and Ne^{**} $2p^4 3s^2$ is observed. On the other hand we were not able to observe the production of autoionizing states of Ar: Ar^{**} $3p^4 4s^2$. This general trend in excited state production is similar to the one observed in gas-phase collisions involving these species (see e.g. [7] for Na–Ne and [8, 9] for Ne, Ar–Mg) and allowed us to conclude that the primary excitation mechanism is the same. Specifically, the origin of excited state production can be accounted for in terms of the Fano–Lichten–Barat [10] electron promotion model. The final state population is, however, quite different because of electron capture and loss processes near the surface in the outgoing trajectory. This and other aspects, such as core rearrangement processes, are discussed by us in some detail elsewhere [6, 11].

More recently Blum *et al* [12] and Nixon *et al* [13] reported the production of the autoionizing states of Ar in the scattering of Ar^+ on Mg, Al and Si, which we and, previously, other authors [14] did not observe. The object of this note is to attempt to explain this rather strange discrepancy, which, as we shall see below, we assign to spurious Ar^{++} ions in the work of Blum *et al* and Nixon *et al*. We also comment on the assignment of sputtered Mg excited states proposed by Blum *et al*.

As a cross check of our earlier work we repeated measurements of electron spectra for the low keV scattering of Ne^+, Ar^+ and Ar^{++} ions on Mg, and these results are reported in the following. The energy range coincides with that in the work of Blum *et al*. The experimental set-up is described elsewhere, but we note briefly that ions are produced in a simple discharge source, velocity selected in a Wien filter and then deflected through 90° in

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order to avoid stray neutrals and photons from reaching the detector. With a 120 V filament to anode discharge voltage the source produced a 4 keV Ar^{++} beam whose intensity is about 30% that of a 2 keV Ar^{+} beam. The electron spectra are recorded using a tandem 45° parallel plate analyser, which can rotate in the horizontal plane, in the 0 to 135° (ϕ) range with respect to the incidence-beam direction, and thus allows an accurate study of kinematic effects due to electron emission from a moving source [2]. The pass energy was 10 eV, corresponding to a 100 meV resolution. Measurements were made for grazing incidence angles (α) in the 1.5 to 15° range. The sample was polycrystalline Mg, which was hand polished to 1 μm. *In situ* cleaning consisted of annealing and grazing incidence (3 to 6°) sputtering with Ne^{+}.

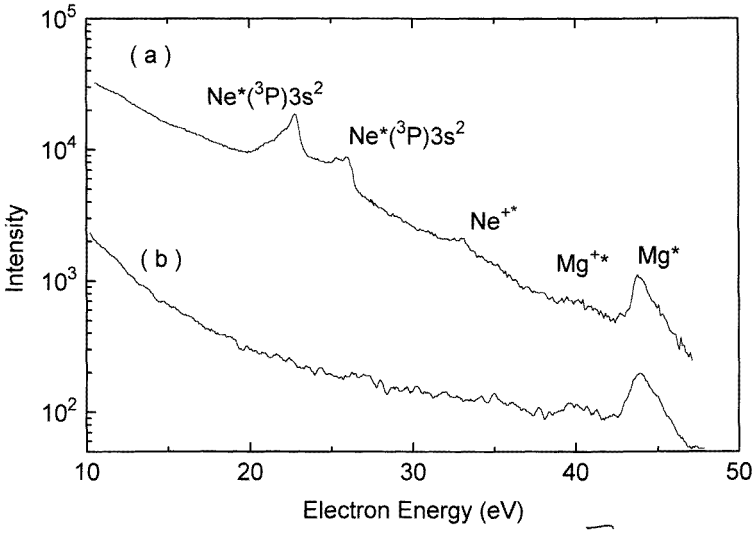
Figure 1 shows electron spectra measured with 3 keV Ne^{+} and Ar^{+} ions incident at 6° on the Mg surface. As reported previously, in the case of Ne we observe peaks due to autoionizing states of Ne and Ne^{+*} [2] and at energies of about 40 eV, a series of peaks due to Mg^{+*} and Mg^{*} states. In the case of Ar^{+}, on the other hand, the spectrum only shows structures due to the Mg^{+*} and Mg^{*} states. No clearly identifiable structure at energies of the order of 13 eV is observed. The full spectrum is given in both cases so that the relative heights of the Ne^{**} and Mg^{*} peaks could serve as a reference for the transmission function of the analyser, which is otherwise not known precisely.

Figure 1(c) shows spectra obtained using 2 keV and 4 keV Ar^{+} beams and a 4 keV Ar^{++} beam. Again, in agreement with our earlier work [2–6], we do not observe any structure corresponding to Ar^{**} in the case of the Ar^{+} beam. However, as reported by us previously [5], in the case of Ar^{++} a clear structure due to Ar^{**} $3p^4(^3P)3s^2$ may be seen at an energy of 13.4 eV. A smaller structure is seen at an energy of 14.9 eV and corresponds to Ar^{**} $3p^4(^1D)3s^2$. The existence of the two structures is related to a small singlet core state component in the Ar^{++} beam. Figure 1(c) also shows a spectrum acquired for a 3° incidence. For smaller incidence the Ar^{**} structure is strongly enhanced. This is more clearly illustrated in figure 2. The numbers beside each curve indicate the integral intensity of the Ar^{**} peak after a smooth background subtraction and a normalization of the incident beam intensity. Note that this is an estimate since the beam intensity measured on the sample includes a component due to part of the beam incident on the side of the sample.

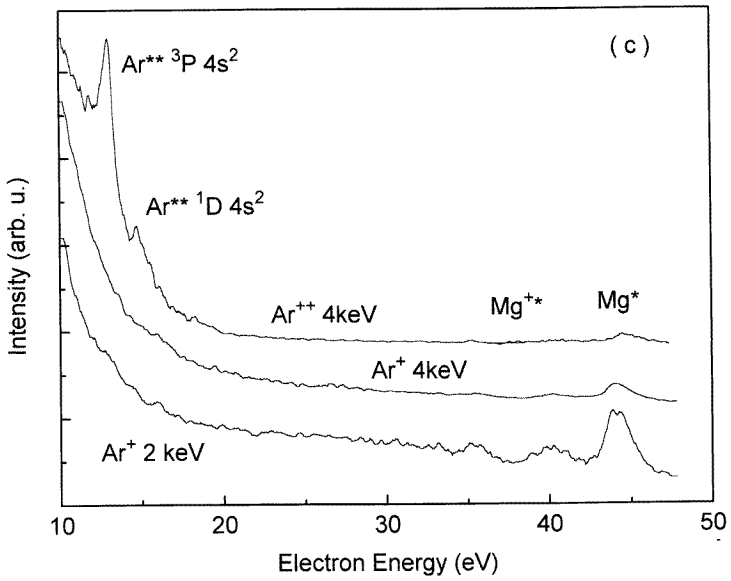
These repeated measurements clearly show that no Ar^{**} state production is observed in Ar^{+} collisions. This is in agreement with our earlier study, the work of other authors [14] and the results of charge fraction measurements by Rabalais *et al* [15], which show that Ar^{+} fractions are very small at 1 keV and increase very slowly as the energy increases to 10 keV. The non-excitation of Ar concords with gas phase measurements and with the prediction of the Fano–Lichten–Barat model. Actually, an extremely small excitation of Ar^{**} or Ar^{-*} has been reported [9] in the gas phase for 5 keV Mg^{+} collisions with Ar. However, we were unable to find any statistically significant evidence for collisions with an Mg surface.

The results of a study of projectile autoionizing state production in Ne^{++} scattering have been very recently discussed by us elsewhere, along with a brief discussion of Ar^{++} scattering. The Ar^{**} states are produced in these collisions in the *incoming* trajectory in an electron-capture-and-loss sequence, before the ions hit the surface. This follows from an analysis of the Doppler shift of the peaks in the spectrum reported in [5]. The larger peak of Ar^{**} for smaller incidence angles corresponds to the smaller perpendicular velocity of incident ions, resulting in a larger capture probability. This peak is also found to be larger at smaller incident energies. At a 10 keV energy for 3° incident angle the peak was no longer visible.

In our opinion the observation of the Ar^{**} states in the work of Blum *et al* and also



(*a*)



(*b*)

Figure 1. The energy spectra of electrons produced in 3 keV (a) Ne^+ and (b) Ar^+ ions incident at $\alpha = 6^\circ$ ($\phi = 30^\circ$). (c) Results for 2 and 4 keV Ar^+ ions and 4 keV Ar^{++} ions ($\alpha = 3^\circ$, $\phi = 35^\circ$). Note that the curves are arbitrarily shifted with respect to each other.

of Nixon *et al* for scattering on Al and Si, corresponds to the presence of a fraction of Ar^{++} in their beam, which they themselves acknowledge as a possibility. On the basis of numerous experimental measurements on electron impact ionization and simply considering the ratio of two to one electron ionization cross sections, one would expect that for the 150 eV electron energy they employ, the beam should contain an Ar^{++} fraction of roughly 10%. The high-energy dependence of Ar^{**} production they report corresponds exactly to

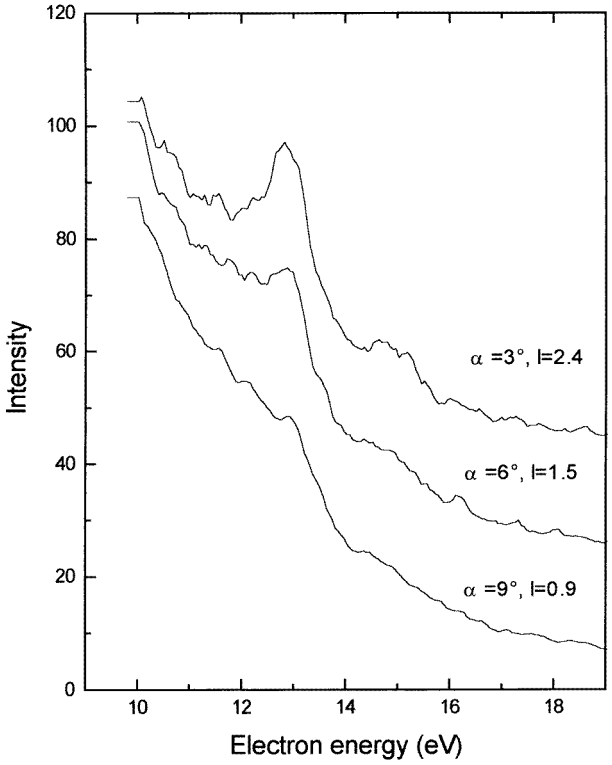


Figure 2. The incidence angle dependence of Ar^{**} state production in Ar^{++} collisions for an energy of 4 keV.

the trend mentioned above, i.e. that capture is favoured at lower energies. However, the very sharp fall for energies below 700 eV appears rather curious. Nixon *et al* also note that excited states were observed for grazing incidence conditions and their results were highly sensitive to the incidence angle. Furthermore, an inspection of the data of Blum *et al* for a 2 keV incident beam, which would correspond to 4 keV Ar^{++}, shows that the position of the peak is the same as in our spectrum. Its position is determined by the *incoming* ion kinematics and by Ar^{**} and Ar^{+} level shifts near the surface [5]. The level shifts result in an emission with a peak position which at 4 keV lies 0.8 eV higher than the expected position. These features are discussed by us elsewhere along with computer simulations of the capture processes [5]. A somewhat puzzling feature in the measurements of Blum *et al* is the fact that they observe a strong signal for a 15° incidence, whereas our measurements show that at such an angle the signal should be weak. A possible explanation is that when using an Ar^{++} beam along with an intense Ar^{+} beam, sputtering of the sample leads to a local modification of incidence angles. Thus the actual incidence angle could be much smaller. Additionally, Blum *et al* used a repulsive (−10 V) potential to accelerate electrons. This procedure could, under favourable geometrical conditions result in an enhanced, solid-angle related, collision efficiency.

Finally, a comment about the production of excited Mg states is necessary. Blum *et al* seem to contest the fact that the three main structures are not of the same origin. Originally, Baragiola [16] suggested the following main assignments for these peaks: 44 eV peak—

Mg^* $2p^5 3s^2 3p$; 40.1 eV peak—$2p^5 3s3p$ Mg^{+*}; 39.5 eV peak—$2p^5 3s^2$ Mg^{+*} state. The assignment of the 44 eV peak to Mg^* was based on the fact that this peak was strongly attenuated on an oxidized Mg surface. We have recently reported a study [4] of excited state production during the initial stages of oxidation of Mg and concluded that the peaks could be assigned as in the work of Baragiola, but our results also suggested that the 44 eV peak has a contribution from the $2p^5 3s4s$ Mg^{+*} state and the 40.1 eV peak has a contribution from the $2p^5 3s^2 3p$ Mg^* state. This was based on the variation of the relative intensities of these peaks as a function of oxygen exposure. These results do not support the suggestion of Blum *et al* that these Mg peaks arise from the same type of charged species.

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