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REPLY TO COMMENT

Reply to comments on the production of autoionizing states in the scattering of argon ions from a magnesium surface

T E Gallon
Department of Physics, University of York, Heslington, York YO1 5DD, UK

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Abstract. We reply to the preceding comment by Guillemot and co-workers.

We have recently published a series of papers [1, 2, 3] describing the measurement and analysis of the electron spectra produced when beams of Ne, Ar and Kr ions are scattered from the surfaces of Mg, Al and Si. As we indicated in the description of our experimental method the ion gun used was a commercial gun (Kratos Mini-Beam) which has a minimum setting for the electron ionizing energy of 50 eV. This means that beams of Ar and Kr ions will be likely to have some doubly ionized species mixed with the singly ionized species since the ionizing energy cannot be reduced below the second ionization energy for these gases. This was indicated clearly in our papers. We made little comment on the excitation mechanism since we were principally concerned with reporting new and detailed structure and with identifying and indexing the spectra. We showed that the considerable fine structure in the spectra could be indexed in terms of excitation during the scattering process into autoionizing states of the *neutral* gas atoms with subsequent decay via electron emission to the ground state. Such Auger emission occurs *after* the collision process, i.e. after the excited atom has left the surface and is moving towards the spectrometer, as evidenced by the Doppler shift in energy which we were able to measure systematically over a range of incident ion energies.

Guillemot *et al* [4] suggest that the data we obtain for Ar and Kr arises solely because of the presence of doubly ionized species in our beam. The data which they present for velocity-selected 4 keV Ar^{++} ions shows clearly that the cross-sections with doubly charged ions are very much higher than for the singly charged species. However, their figure 1 is a little puzzling since the Mg peaks shown seem very small. Our measurements were made at slightly higher angles, but at 4 keV the ratio of heights of the Mg:Ar principal peaks is $\sim$10:1 in our data. If this ratio applies to the data of Guillemot *et al* then the expected Ar signal would be of the order of the noise of their spectra. This suggests that their data may not be sufficiently noise free to eliminate completely a contribution from Ar^{+}. Guillemot *et al* state that they and other workers have tried and failed to observe the excitation of autoionizing states in Ar^{+} scattering from solids. Unfortunately, they appear to have overlooked the work of Brenten *et al* [5], referred to in [2] and [3]. Brenten *et al* also used a velocity-selected ion beam, set to select only Ar^{+}, and scattered from a tungsten target with sub-monolayers of K absorbed onto the surface. The data which they have published for Argon resembles ours very closely, showing similar structure at the same energies.

The difference between the work described in [2], [3] and [5], which found the production of autoionizing states, and that outlined in [4], which did not, is that the former

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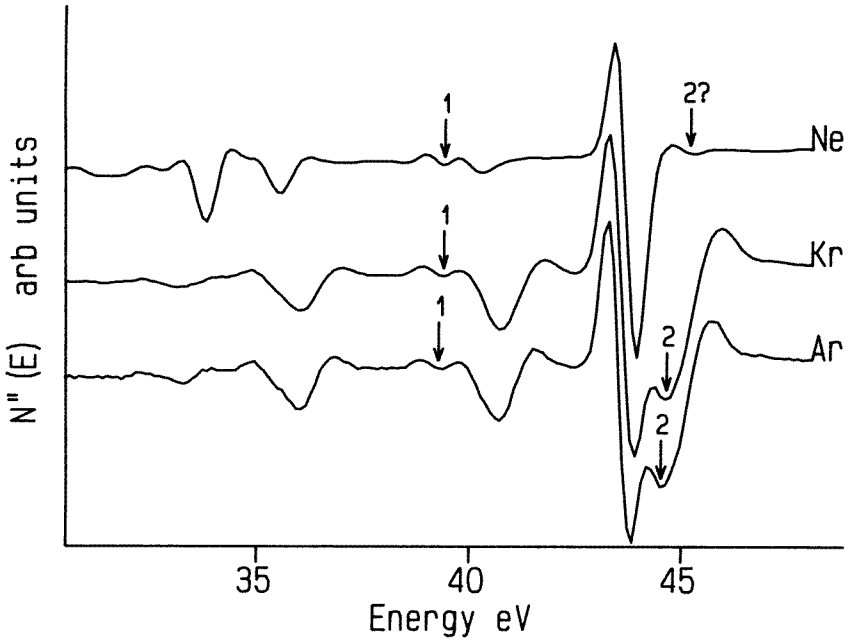

Figure 1. Second derivative spectrum of magnesium excited by Ar ions accelerated through 1200 V, Kr ions accelerated through 2000 V and Ne ions accelerated through 4000 V.

measurements were made at low energies. We showed that the excitation cross-section for the production of Ar spectra decreased rapidly above a 750 V accelerating potential and was considerably reduced at 4–5 kV. We also found that the cross-section appeared to fall below 750 V as well, a result which we were unable to explain. Guillemot *et al* also draw attention to this unexpected behaviour at low energies. One possible explanation is that the decrease at low energies arises because of the defocusing of the ion gun at energies below the design range. If the beam area is larger than the 'field of view' of the input lens then only part of the beam will contribute to the recorded spectrum, but all of the beam will be measured as current to the specimen. In addition, the signal is very small at low energies and it is difficult to align the beam with the optic axis of the spectrometer. Misalignment of the beam will also cause the beam current to be overestimated; indeed in extreme cases current striking the side of the specimen holder could be measured as part of the beam current. These effects will only be important at low energies and will lead to an apparent reduction in the excitation cross-section at low energies. To overcome these problems it is necessary to develop a self-consistent measurement of beam current. This is in hand at present and first results indicate that the intensity of the Argon peak increases with reduction of ion energy down to accelerating voltages below 200 V and possibly lower. We hope to present this work for publication in the near future. This suggests that examination of relative contributions from Ar^{+} (if any) and Ar^{++} to the production of autoionizing states in Ar^{0} is best made at lower energies than those used by Guillemot *et al*.

The purpose of publishing the Mg data [3] was to show that, with a favourable signal to noise ratio, the autoionizing spectrum is not a three-peak structure but contains at least five peaks with energies at approximately 44 eV, 40 eV, 36 eV, 33 eV and 31 eV. We attempted to index this as a Rydberg-like series and found that we could indeed label

the peaks as arising from an initial configuration in Mg^0, decaying to a series of lower energy configurations in Mg^+ with emission of an electron. Because of the difficulties in the calibration of electron spectra, the natural breadth of peaks and uncertainties in the calculations, no definite labelling of the Mg peaks can be made solely on energetic grounds and all of the proposed labels [6, 7] remain possibilities. The suggestion of Guillemot *et al* that the 44 eV and 40 eV peaks are composite is interesting. There is little suggestion of structure in these peaks in the data taken in the $N(E)$ form but in searching for small structures it is often useful to examine the data in differential form. Figure 1 shows second-derivative data for the Mg peaks with Ne, Ar and Kr excitation. The differentiation was performed numerically in such a way as to model the applied modulation method normally used to obtain the derivatives of Auger peaks from solids [8]. The equivalent modulation would be ∼0.6 V p–p, well inside the measured width of the peaks.

In the case of the peak around 40 eV the situation is quite clear. The peak shows reproducible structure on the low-energy side—peak 1 in figure 1. Measurement of the minimum in the second derivative gives the main peak energy as 40.5 eV, compared to 40.3 eV from the $N(E)$ data, with the new structure having an energy of 39.4 eV. Whether this new structure represents one or other of the suggested processes or whether it is due to coupling in the final state is impossible to state although it is close to the energy estimated from optical data [1] for the transition $1s^22s^22p^53s^23p \rightarrow 1s^22s^22p^63p + e$ [3]. The data for the 31 eV and 33 eV peaks are too noisy to make any statement on additional structure; the peaks visible with Ne excitation arise from overlap with Ne structure [3]. No extra structure can be identified for the 36 eV peak even though the signal to noise ratio for this peak is similar to that for the 40 eV peak.

The situation with the 44 eV peak is complex. The data recorded with Ar and Kr ions appear very similar. At lower energies a well-defined minimum, labelled 2 in figure 1, is present in the second derivative with energy ∼0.6 eV above the main peak. However, as the incident ion energy increases this peak broadens and moves to higher energies merging into the derivative of the 44 eV peak at about a 4 keV ion accelerating voltage when the energy difference is ∼1 eV. Structure is also seen above the 44 eV peak with Ne excitation but this does not vary with incidence energy and remains constant at 1.3 eV above the main peak. The effect of the growth and decay of the small structure in the Ar data can be seen in the apparent width of the peak measured in the p–p width of the first derivative curve. At a 1 kV ion accelerating voltage the measured separation is 1.6 eV and this falls to 1.2 eV at 4 kV. This contrasts with the case of Ne excitation where the p–p separation in $N'(E)$ remains at 0.8 eV with ion energies between 1 keV and 4 keV. The size of the shift seen with Kr and Ar excitation is of the order of the Doppler shift seen in Kr and Ar peaks and it is tempting to identify this peak as arising from the narrow jet of atoms emitted at high energies which Pepper and Aron [9] suggest is superimposed on a broad low-energy distribution. However, the evidence for such a jet is indirect and in any case too much emphasis should not be placed on changes in second-derivative spectra since these are difficult to interpret.

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