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Production and Decay of Double *L* Vacancies in Argon and Phosphorus[*]

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Measurements have been made at two laboratories which indicate that the structure reported at 450–550eV in the electron spectrum from Ar^+-Ar collisions by Ogurtsov, Flaks, and Avakyan is spurious. It is argued that the double *L* vacancies which they invoke to explain the structure are more likely to decay by the two-step Auger process $L^2 \to LM^2 \to M^4$ than by the one-step process $L^2 \to M^3$ suggested by these authors. Evidence supporting this is found in our electron spectra from P^+-Ar collisions, where it is known from energy-loss and charge-state measurements that double *L* vacancies are produced in phosphorus.

I. INTRODUCTION

Fano and Lichten[1] have interpreted energy-loss measurements[2,3] in Ar^+-Ar collisions in terms of the now well-known molecular-orbital promotion model. The three energy-loss peaks observed are presumed to be due to the promotion of zero, one, and two L electrons from the pair of colliding atoms. These and more recent energy-loss measurements[4,5] for Ar^+-Ar collisions do not have sufficient resolution to unambiguously distinguish the promotion of two electrons in one atom from the promotion of one electron in each of the two atoms. However, the spectrum of Auger electrons ejected after such promotions would be different in the two cases and can be used to determine which process actually takes place.

Ogurtsov, Flaks, and Avakyan[6] (hereafter referred to as OFA) have presented spectra of electrons detected at 54.5° in 15-keV Ar^+-Ar collisions in which there appears to be structure in the energy region 450–550 eV. Without identifying lines, they assign this group to transitions of the type $L^2_{2,3} \to M^3 + e^-$. If correct, this would indicate not only that is it probable that double vacancy states are produced in argon atoms in such low-energy collisions, but also that these states decay by a one-step process involving the simultaneous filling of two vacancies with all of the excess energy being given to a single M-shell electron, which is then ejected.

Though this result might be of considerable significance, it has never been confirmed. We have attempted to obtain such spectra under similar conditions by experiments on four different sets of apparatus at two laboratories, but have found that such structure, if it exists at all, must have a cross section three to four orders of magnitude smaller than that given by OFA. We also present evidence from the Auger spectrum of phosphorus, which indicates that when such double L-shell vacancies are created, they decay by a two-step process involving the emission of two Auger electrons, rather than the one-step process suggested by OFA.

II. EXPERIMENTAL APPARATUS

Data are presented here from experiments using four different sets of apparatus. Certain common features of all four will be described first, followed by brief separate descriptions of each.

In all cases the ion beam was momentum analyzed and finely collimated before entering the target region and caught in a shielded and biased Faraday cup after passing through the target gas. Most runs were at a gas pressure of 1–2 mtorr. Electrons leaving from a short length of the beam at the scattering center passed through an electrostatic energy analyzer and were detected by an electron multiplier. Individual electron pulses from the detector were amplified and shaped by logic circuits for counting. Magnetic fields at the scattering center and analyzer were reduced to below 10 mG by three pairs of Helmholtz coils. At this level, the field would have little or no effect on electrons above 100 eV, which is the range of interest of this experiment. The collision chamber, the electrostatic analyzer plates, and the pipes connecting them were coated with colloidal graphite (Aquadag) to minimize reflection and secondary emission of electrons from the surfaces. High-purity argon (maximum 0.005% impurities) from commercial sources was used, and the gas lines to the chamber were evacuated and flushed at least twice before measurements were made.

Care was taken to prevent secondary electrons from surfaces struck by the ion beam from reaching the detector. This is extremely important since the production of secondaries from solid surfaces is much more copious than from the target gas. The estimated uncertainty in the absolute values of the cross sections presented here is 50% up to about 300 eV. This comes mainly from uncertainties in the detection efficiency and the pressure measurement.

The four sets of apparatus will be labeled A–D, and are as follows.

Apparatus A. This apparatus at the Institute of Physics at Aarhus was built by Dahl. The design is shown in Fig. 1. The 60° cylindrical analyzer and detector can be rotated continuously to give angles relative to the ion beam from 20° to 160°. The ion beam is completely enclosed by a pipe both before and after passing through the gas cell to confine any secondaries which may be produced by the beam. Pumping ports in these pipes are covered by screens at a negative bias. The analyzer and detector are likewise completely enclosed and pumping holes have biased screens on them. The circular aperture S_3 is imaged by the analyzer at the slit S_5. The sizes of S_3, S_4, and S_5 are such that no electrons passed by them can strike the plates of the analyzer. The energy resolution is 0.9%. The cone of the Mullard Channel Electron Multiplier B 419 B1 could be biased either positively or negatively, and negative voltages of as much as −100 V were used in this work. This tended to reduce the background due to spurious low-energy electrons, although due to the semiconducting nature of the cone surface, the entire cone was not at the same potential.

Apparatus B. This apparatus at the Institute of Physics at Aarhus is designed for electron-ion coincidence studies.[7] A 90°, 35-mm-radius, spherical electrostatic analyzer provides a large solid angle of acceptance for the electrons. This feature makes the electron-ion coincidence apparatus well suited also for electron studies in which the production cross section is exceedingly small, as is the case for Auger electrons originating from an $L^2 \rightarrow M^3$ process. The analyzer accepts electrons which are emitted 96°± 3° from the beam direction. The observed target length is 1 mm. The energy resolution is 1.5%. The electron detection system is a Mullard Channel Electron Multiplier B 419 B1. To suppress contributions from low-energy background electrons, a small negative potential is applied to the detector-entrance cone. The incident ion beam is defined by two circular 0.8- and 0.5-mm-diameter diaphragms, 300 mm apart. The beam current passing through the gas is monitored by a shielded Faraday cup.

Apparatus C. This is a chamber originally built by Rudd and Jorgensen[8] and slightly modified by Cacak and Jorgensen,[9] who took the Ar^+-Ar data quoted here at a resolution of 4.2%. Their published data went only to 350 eV, as the statistics were relatively poor beyond that energy for some parameters. However, if one is willing to accept a relatively large statistical error, these data are of use out to 700 eV. The chamber has fixed ports at eight angles from 10° to 160° and uses a 127° electrostatic analyzer and a Dumont Cu-Be box-and-dynode-type electron multiplier detector.

Apparatus D. Originally used by Rudd, Sautter, and Bailey,[10] this apparatus has nine fixed ports from 10° to 160° and is similar to C, except that a parallel-plate analyzer is used in place of the cylindrical one. The energy resolution is 4.6%.

In this apparatus a small count rate was present which was proportional to the beam current and gas pressure, but which was independent of any biases or magnetic fields and was approximately constant from an analyzer setting of 300–1000 V. These counts were attributed to high-energy photons from the collision center which reflected from the back plate directly into the detector. It is also possible that neutral argon atoms or high-energy ions could contribute to this residual count rate. This problem was noted before and partially corrected by coating the back plate with a colloidal

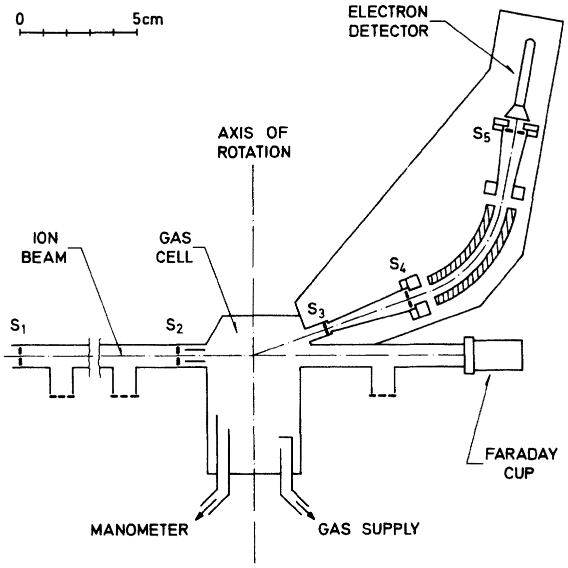

FIG. 1. Diagram of apparatus A. Analyzer and detector rotate through 360°, thus allowing electron to be detected from 20° to 160°. Ion beam is collimated by S_1 and S_2 followed by an electron suppressor and shield. Circular apertures S_3 and S_4 (both grounded) collimate electron beam before analyzer.

solution of graphite. This reduced the effect, but did not eliminate it. For the earlier work at low energies this residual background represented a small correction, but in the present work where very small cross sections were being measured, steps had to be taken to reduce it still further. A slot was cut in the back plate large enough to allow all photons coming through the electron collimation system to pass through. A wire mesh grid was put over the slot to eliminate field distortion, but it was later removed without an appreciable change in the behavior of the analyzer.

The efficiency of the detector decreases sharply for electrons below about 100 eV. Therefore, in earlier work we kept the first dynode at a constant positive potential of 200–300 V. We have found, however, that when detecting electrons at high energies where the cross sections are extremely small, the background due to slow secondary electrons is markedly decreased by operating the first dynode at a negative bias. Too large a negative bias, however, causes defocusing and loss of electrons. The following procedure was used. For detecting electrons up to 400 eV the detector bias was varied along with the analyzer potential so that electrons were accelerated or decelerated to 200 eV at the first dynode. Above 400 eV the negative bias was chosen to be numerically equal to half the electron energy. The multiplier voltage was adjusted in all cases to keep the total voltage across the dynode string constant at 3400 V.

With these precautions the count rate in the 300–900-eV region was reduced to approximately one count in 10 sec, compared to about 400/sec at 200 eV. This was not much above the background count rate, which was about 3/(100 sec) without the target gas.

To evaluate the contribution to the count rate due to the photons mentioned above, a bar magnet was placed in such a way as to provide a magnetic field of 25–50 G at a point along the electron path just before the analyzer. This prevented all electrons originating in the collision chamber from reaching the detector. The remaining count rate was subtracted from the total count rate to get the electron count. Absolute cross sections were calculated from the measured pressure and geometry.[11]

III. EXPERIMENTAL RESULTS

Figure 2 shows several measurements of the Ar^+-Ar cross sections in this work compared to the results of OFA. The agreement in absolute value is good at the 200-eV peak, but poor elsewhere. In particular, the upper limit of our cross sections in the 450–550-eV region is three to four orders of magnitude smaller than OFA. These authors have recently indicated[12] that the structure they reported was probably due to instrumental effects.

Because the count rates were very small above 300 eV, most of the cross sections measured in that region had a statistical uncertainty of about 100%. Therefore, the results of this experiment are consistent with the hypothesis that the cross

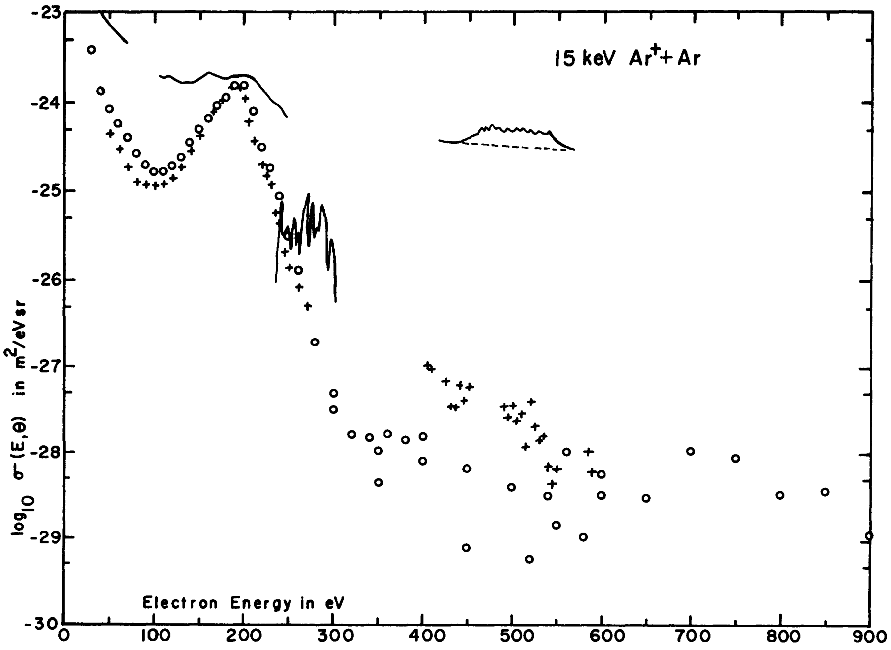

FIG. 2. Cross sections for electron ejection differential in energy and angle for 15-keV Ar^+-Ar collisions. Solid lines: data of Ogurtsov, Flaks, and Avakyan (Ref. 6) taken at 54.5°. Crosses: present data taken at 54.5° on apparatus A. Circles: present data taken at 50° on apparatus D.

section actually falls off exponentially above 300 eV at the same rate as it does between 200 and 300 eV.

In the 230–300-eV range our resolution was not great enough to detect the fine structure which OFA reported, but it appears that the general trend of their cross sections should be decreasing more than they indicate. A slight bulge in our curves at 240 eV is the only indication of the L_1-M^2 Auger transitions suggested to explain the structure in that range.

It might be expected that a higher primary energy than 15 keV would be more likely to produce the double vacancy states and thus the structure around 500 eV. Figure 3 shows three runs at 50 keV and at a larger emission angle. Again, no structure is found at that energy and the cross sections are very small. Runs were also made at 100 keV with similar results. A portion of Cacak and Jorgensen's data[9] at 300 keV is also shown, which indicates no structure at 500 eV.

There are several lines of evidence which indicate that in P$^+$-Ar collisions, double L vacancies are produced in the phosphorus atom with high probability, whereas in Ar$^+$-Ar collisions, double L vacancies in a single atom are less probable. In the inelastic energy-loss measurements of Fastrup, Hermann, and Smith,[5] the mean energy loss in such collisions shows the usual triple peak. The first one, labeled Q_{I}, is for an energy loss involving only outer-shell electrons, while the next two, Q_{II} and Q_{III}, are due to the removal of one and two L-shell electrons, respectively, in addition to the Q_{I} excitation. From Table I of Fastrup *et al.*, one sees that for P$^+$-Ar collisions, $Q_{\mathrm{II}}-Q_{\mathrm{I}}=B_L(\mathrm{P})\neq B_L(\mathrm{Ar})$ and $Q_{\mathrm{III}}-Q_{\mathrm{I}}=B_{L^2}(\mathrm{P})$ but $Q_{\mathrm{III}}-Q_{\mathrm{I}}\neq B_{L^2}(\mathrm{Ar})$ and $Q_{\mathrm{III}}-Q_{\mathrm{I}}\neq B_L(\mathrm{P})+B_L(\mathrm{Ar})$. For Ar$^+$-Ar collisions, $Q_{\mathrm{III}}-Q_{\mathrm{II}}=Q_{\mathrm{II}}-Q_{\mathrm{I}}=B_L(\mathrm{Ar})$. Here B_L and B_{L^2} are the binding energies of one and two $2p$ electrons, respectively. This indicates that the energy loss involving two L-shell electrons corresponds closely to the energy needed for the removal of both electrons from the phosphorus atom in P$^+$-Ar collisions; but in the case of Ar$^+$-Ar collisions, the energy loss is more nearly equal to that required for the removal of

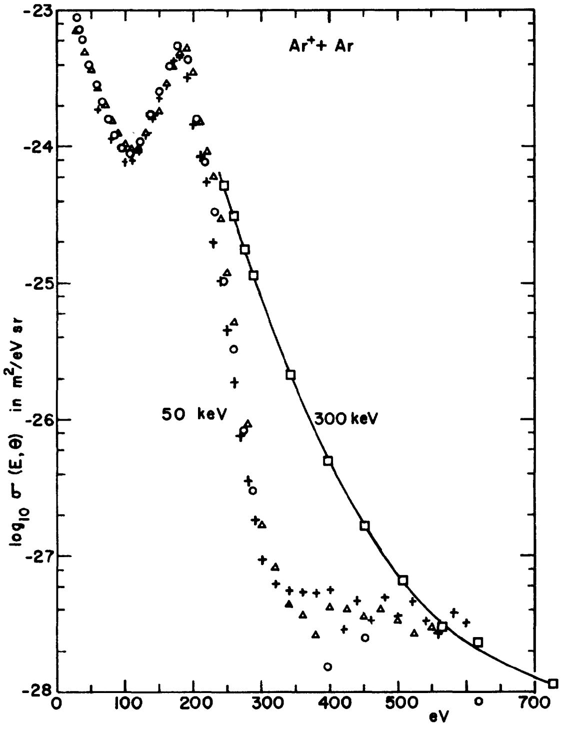

FIG. 3. Cross sections for electron ejection differential in energy and angle for Ar$^+$-Ar collisions. Crosses: present data at 50 keV and 96° on apparatus B. Triangles: present data at 50 keV and 90° on apparatus D. Circles: data taken at 50 keV and 90° on apparatus C by Cacak and Jorgensen (Ref. 9). Line and squares: data at 300 keV and 90° by Cacak and Jorgensen (Ref. 9).

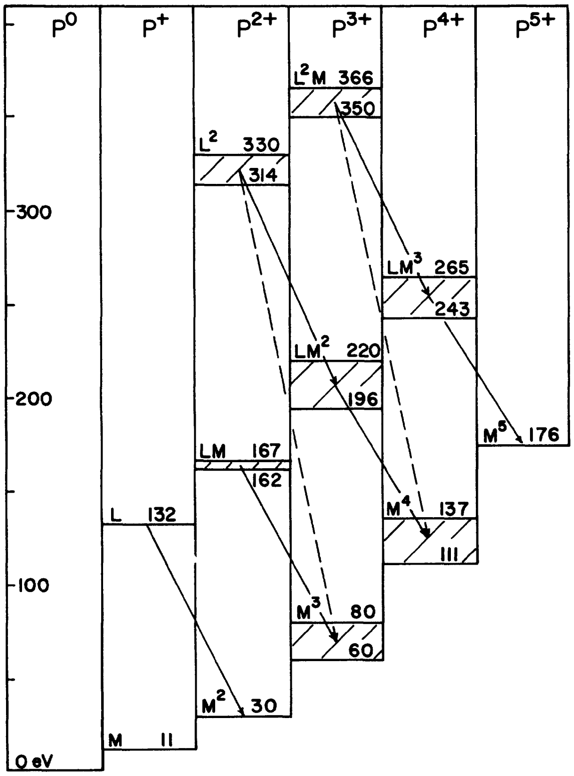

FIG. 4. Simplified energy-level diagram for phosphorus. Only $L_{2,3}$ states are considered. Dotted arrows show hypothetical one-step transitions from double L vacancy states. Solid arrows show other Auger transitions.

one L-shell electron from each of the atoms of the colliding pair. The data, however, are not able to rule out the possibility that in Ar^+-Ar collisions, a small fraction of the $Q_{\rm III}$ energy loss results in the production of double L vacancies in one of the argon atoms.

The evidence that double L vacancies are produced in phosphorus is supported by charge-state analysis of the collision products. It has been shown by Fastrup and co-workers[5] that in P^+-Ar collisions neither a $Q_{\rm II}$ nor a $Q_{\rm III}$ energy loss changes the mean-charge state of the argon recoil from that for a $Q_{\rm I}$ energy loss.

Evidence that the probability of producing double L vacancies in one argon atom in Ar^+-Ar collisions is small is also found in the inelastic energy-loss measurements. It has been shown[5] that for all asymmetric collisions with argon targets, $Q_{\rm III} - Q_{\rm II} > Q_{\rm II} - Q_{\rm I}$, where $Q_{\rm III}$ corresponds to the production of a double L vacancy in the lower-Z collision partner. This is consistent with the fact that the binding energy B_{L^2} of two $2p$ electrons is more than twice the binding energy of a single $2p$ electron. For Ar^+-Ar collisions, however, it was found that $Q_{\rm III} - Q_{\rm II} = Q_{\rm II} - Q_{\rm I}$, which is evidence that in a $Q_{\rm III}$ energy loss, two L vacancies are produced, one in each of the target atoms.

The ion-ion coincidence studies of Ar^+-Ar collisions by Kessel and co-workers[13] showed that for a $Q_{\rm III}$ energy loss, there were no appreciable correlations between the charge states of the collision products. This was in contrast to a $Q_{\rm II}$ energy loss, where a definite correlation was observed, corresponding to an L vacancy being produced in one or the other argon atom. If in a $Q_{\rm III}$ energy loss, double L vacancies were produced in one of the argon atoms, then a definite charge-state correlation should exist.

Additional evidence that double L vacancies are formed in phosphorus atoms in P^+-Ar collisions comes from electron-ion coincidence data. From the energy-level diagram of Fig. 4 it is seen that no electrons ejected from a single L vacancy state can have an energy greater than 89 eV if this results in a final charge state of P^{5+}. However, Dahl and Lorents[7] presented electron spectra taken in coincidence with P^{5+} projectiles which show structure up to energies well over 100 eV. Such energies are possible from the transition $L^2_{2,3}M_{2,3} \rightarrow L_{2,3}M^3$, the latter decaying to the ground state of P^{5+} by a second Auger process as shown in Fig. 4.

In summing up, it is much more likely that double L vacancies are produced in phosphorus in P^+-Ar collisions than in either argon atom in Ar^+-Ar collisions. This suggests that when looking for Auger electrons from the $L^2 - M^3$ process suggested by OFA, one would have a better chance of observing them in P^+-Ar than in Ar^+-Ar collisions. Such a process in phosphorus would yield electrons with energies between 213 and 270 eV. In the spectrum of electrons at 20° from 50-keV P^+-Ar collisions this would cause structure in the energy region of about 240–300 eV, owing to the Doppler shift.[14] We have studied this spectrum carefully in this region with apparatus A (see Fig. 5) and find no evidence of structure. We can put an upper limit on the cross section for such structure at about 4×10^{-25} m^2/sr at 20°. This is only 0.2% of the size of the cross

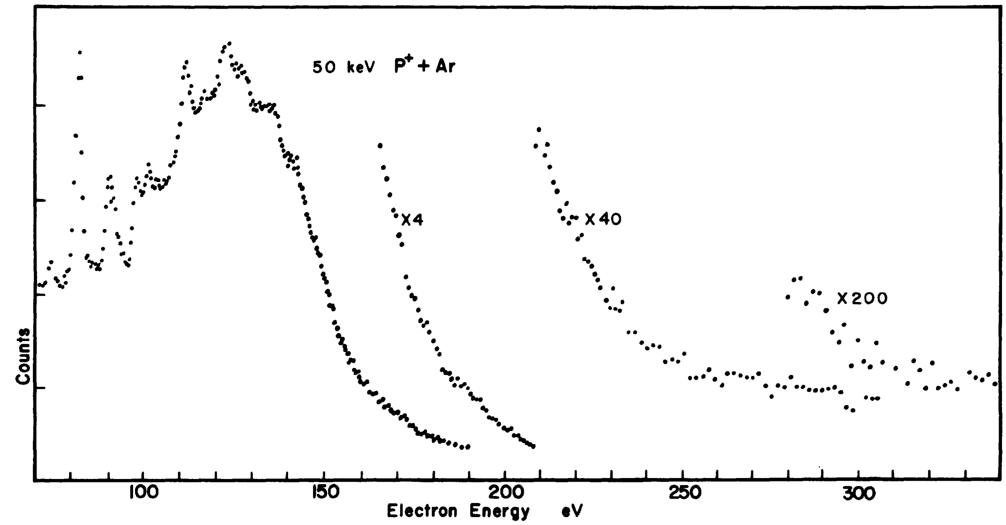

FIG. 5. Electron energy spectrum at 20° from P^+-Ar collisions at 50 keV. Data, taken on apparatus A, represent counts on a linear scale uncorrected for background and analyzer dispersion.

section for the Auger contribution in the 60–150-eV region.

IV. SUMMARY

Argon electron spectra from which the structure at 450–550 eV reported by OFA is absent have been presented. This indicates either that no double *L* vacancy states are produced or that if they are, they do not decay by a single Auger transition. Also examined were Auger spectra from phosphorus where it is much more likely that double *L* vacancy states are produced. However, even here no structure in the Auger spectrum was found in the region expected if such double *L* vacancies decayed by a one-step Auger process. It is concluded that double *L* vacancies are much more likely to decay by a two-step than a one-step process.

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