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[15]Note added in proof: Recently, a related inadequacy of a direct ionization model in O_2 has been reported [J. W. McGowan, E. M. Clarke, H. P. Hanson, and R. F. Stebbings, Phys. Rev. Letters 13, 620 (1964)].

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CHARGE-STATE CORRELATIONS IN Ar^+-Ar COLLISIONS

Edgar Everhart and Quentin C. Kessel
Physics Department, The University of Connecticut, Storrs, Connecticut
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Single collisions of Ar^+ on Ar, at 25 to 150 keV, have been studied using a coincidence scattering apparatus. For the reaction

$$Ar^+ + Ar \rightarrow Ar^{m+} + Ar^{n+} + (m+n-1)e, \quad (1)$$

the charge states m and n of both particles from the same large-angle collision are determined. The experiment finds the probabilities of seeing m and n in coincidence. In addition, simultaneous measurements of the two scattering angles θ and φ determine the inelastic energy loss Q_{mn} for the (m,n) reaction.

The first such coincidence measurements were reported recently by Afrosimov, Gordeev, Panov, and Fedorenko,[1] who studied the above reaction at 12.5 and 50 keV. Our apparatus and procedure, while different from theirs, are functionally similar, and our 50-keV data for Q_{mn} agree with theirs. However, our correlation measurements suggest a new interpretation for the phenomena.

We find no correlation between m and n. Figure 1 shows a typical case, using 50-keV, $\theta = 15°$ data. Each curve is for a specified charge state m of the scattered incident particle and shows the percentage distributions P_n for the recoil particle. Within the scatter of the data, these distributions are the same. A similar diagram results when the roles of m and n are interchanged, again showing that the distribution among the charge states of one particle is independent of the charge state of the other particle. Other data sets at 25, 100, and 150 keV confirm this lack of correlation. This result, at first sight, appears to disagree with the previous work,[1] which plots the relative

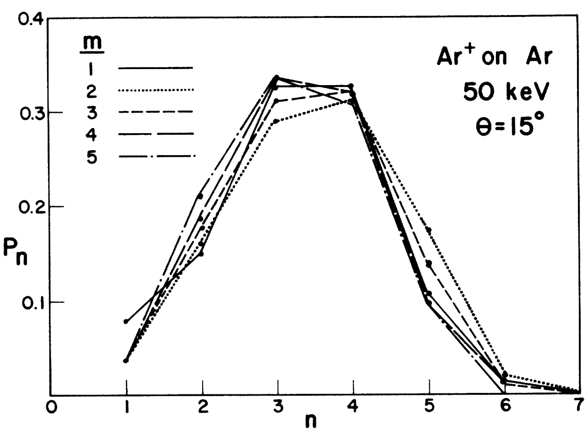

FIG. 1. Charge-state correlation diagram for Ar^+-on-Ar collisions at 50 keV, $\theta = 15°$.

number of particles for constant $m+n-1$ vs $m-n$ and obtains curves of universal shape, suggesting a form of correlation. However, detailed examination would show that their result comes about because the distributions P_m and P_n, although uncorrelated, are the same for either particle.

This absence of correlation is consistent with the Russek[2] statistical theory of multiple ionization. He assumes that the inelastic energy transferred to each atom is statistically distributed among the outer electrons in an autoionizing transition. In his model each atom gets half the inelastic energy, and the transitions occur after the atoms have separated, hence no correlation.

Inelastic energy-loss data will illustrate both the previous[1] and the present interpretation. In Fig. 2, Q_{mn} is plotted versus $\sum U_i(m,n)$, the net change in spectroscopic ionization energies before and after the interaction. Data of Afrosimov et al.[1,3] are shown to compare with our 50-keV, 15° data set, and these two agree fairly well. In that work[1] their data, and that for other data sets as well, were fitted to the same straight line of unit slope,

$$Q_{mn} = \sum_i U_i(m,n) + R_{\mathrm{III}}^*, \qquad (2)$$

where the intercept R_{III}^* was considered to be one of three characteristic excess-energy losses. However, for two reasons we suggest that the R^* concept may not be consistent with all the information on hand: (1) The slopes in Fig. 2 appear less than unity, and the intercept moves upward continuously at energies above 50 keV, not having a fixed value.[4] (2) The concept of a characteristic excess-energy loss requires an (m,n) correlation so that the final charge-state information be determined by the system as Q_{mn} is transferred. Absence of correlation negates the R^* concept.

An example, using 50-keV, 15° data, introduces our new interpretation. We assume a fairly wide distribution w in the Q values,[5]

$$w = \exp[-(Q-\bar{Q}^2)/a^2], \qquad (3)$$

with $a = 250$ eV and $\bar{Q} = 750$ eV as in Fig. 3(a). The width a is adjusted for best over-all fit, but the center value, $\bar{Q}$, is measured here. For each value of Q on this curve there is a distribution P_m as shown in Fig. 3(b). Multiplying these by w results in the weighted distributions wP_m shown in Fig. 3(c), and the values wP_n for the other particle are the same. Since the occurrence of state m in one particle and of state n in the other are independent uncorrelated events, the probability that states m and n are seen simultaneously is the product $w^2P_mP_n$, as shown[6] in Fig. 3(d) for several cases. The various Q_{mn} from the peaks of these m,n curves are plotted as the computed points on Fig. 2 where they fit the data approximately.[7] Similar calculations, with $a = 225$

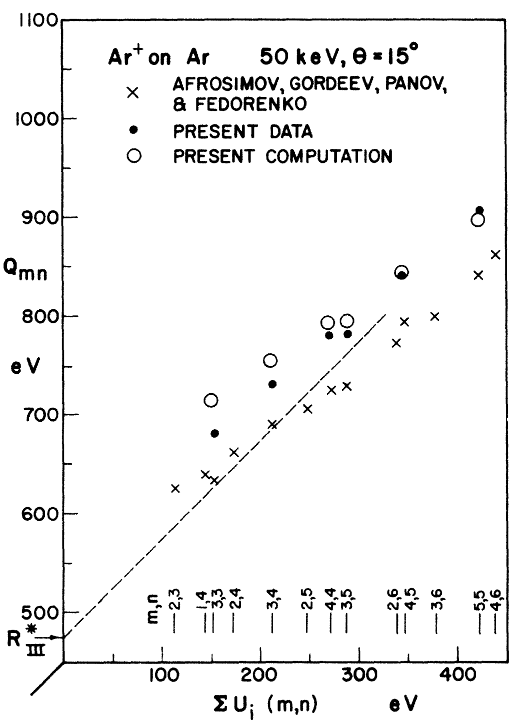

FIG. 2. Inelastic energy loss is plotted versus the net ionization energies for Ar$^+$-on-Ar collisions wherein the charge states m,n of both particles after the collision are specified. The dashed line is fitted to the data of Afrosimov et al.[1] using Eq. (2). The open circles correspond to the present data and are computed herein.

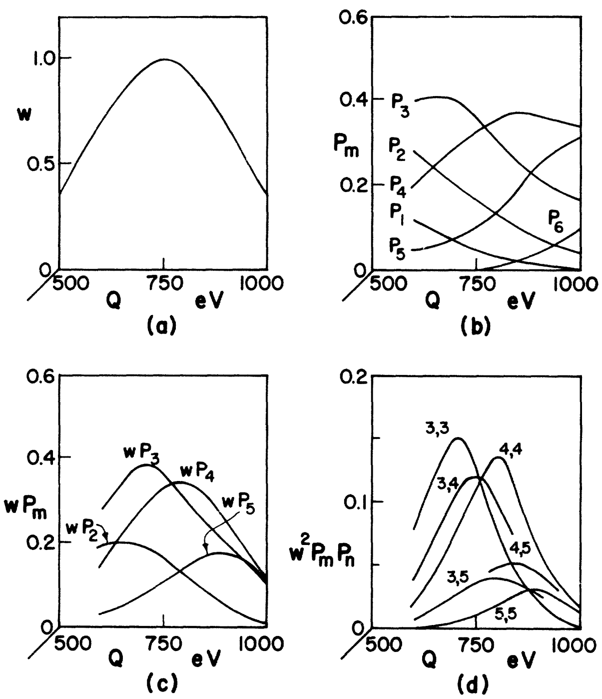

FIG. 3. These curves interpret Ar$^+$-on-Ar scattering at 50 keV, $\theta = 15°$. (a) An assumed Gaussian distribution in the inelastic energy Q. (b) Experimental charge-state distributions for either particle versus Q. (c) Charge-state distributions weighted by the assumed Gaussian distribution. (d) Squares and cross products of the weighted charge-state distributions giving the probability versus Q that charge state m is seen simultaneously with charge state n.

and $\overline{Q} = 875$ eV, fit 100-keV, 7° data.

Besides depending on a, the fit to the Q_{mn} data depends on the experimental curves of Fig. 3(b). These curves combine present $\overline{Q}$ vs θ measurements with early P_n vs θ measures by Fuls et al.,[8] and assume that P_n vs $\overline{Q}$ is substantially the same as P_n vs Q. With improved apparatus these measurements are being repeated and a revision of Fig. 3(b) may be expected, making possible a more exacting check on the present interpretation.

Under some conditions a triple structure is observed in the Q values for Ar^+ on Ar collisions. This was first seen as a fine structure at 12 keV near $\theta = 38°$ (and interpreted incorrectly) by Morgan and Everhart,[9] who did not make coincidence measurements. The three peaks were found at 50 keV for angles near 7° by Afrosimov, Gordeev, Panov, and Fedorenko,[1] who investigated these peaks extensively giving R^* values for each. In our interpretation there is instead, at those particular energies and angles, a triply peaked distribution in the Q values. The reason for this triple structure is yet unknown.

The experimental distributions used here are very similar to those computed by Russek,[2] and the present development is consistent with his model. We have benefitted from many valuable discussions with Professor Arnold Russek.

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[3]Afrosimov et al., reference 1, p. 1630. Use 0.183Å data, which correspond to 50 keV, 15°, and add $\sum U_i$ to the R_{III}^* column to find Q_{mn}.

[4]In our data the intercept R_{III}^* increases from the value shown in Fig. 2, reaching about 1000 eV for 150-keV, 13° collisions.

[5]The Gaussian form of distribution is chosen arbitrarily. Another distribution shape may ultimately prove to be preferable.

[6]The curves of Fig. 3(d) suggest a natural linewidth of about 200 eV at half-height. Allowing for instrumental resolution and thermal target motion, one would predict further broadening in Q_{mn} measurements. Our measured linewidths appear consistent, within 25 %, with our estimates. This does not agree with reference 1, which assigns a much more narrow natural width to the Q_{mn} values.

[7]The computed points do not lie on a smooth curve because $\sum U_i$ is not an appropriate abscissa for our interpretation, having been chosen to illustrate Eq. (2).

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