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Electron impact autoionization in heavy alkali metals*

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Autoionizing levels in cesium, rubidium, and potassium have been studied by electron impact in a crossed-beam apparatus. Comparisons are made with the binary-encounter calculations of Roy and Rai. The effects of autoionization and inner-shell ionization have been overestimated in the theory.

The purpose of this communication is to comment on a recent theoretical paper by Roy and Rai[1] and to add experimental data that support their calculations in part.

An excellent review of theoretical calculations of electron-impact ionization cross sections in the alkali metals has already been made by McDowell.[2] The review will not be repeated here, but for completeness newer works will be included and compared with previous calculations.

The major contribution of Roy and Rai was for the first time to take autoionization processes into account, resulting in new structures for the heavy alkali metals Cs, Rb, and K. Independently, we previously made measurements on Rb and have made new measurements on K, and found agreement with the energies at which the autoionization peaks occur, but not with the absolute magnitudes predicted by theory.

While quantum-mechanical calculations have been quite successful in predicting cross sections in Li and Na, they are not readily applicable to more complicated systems like K, Rb, and Cs. The alternative approach is then to use classical and binary-encounter theories to estimate cross sections for the heavier alkali metals. The basis of Roy and Rai's calculations is the symmetrical collision model developed by Vriens[3] along with quantal velocity distribution for the bound electron and including excitation of autoionizing levels.

According to Moore's tables[4] the lowest autoionizing states for the heavy alkali metals are those compiled in Table I. In a previous work[5] we have resolved about 20 autoionizing levels in Cs between 12 and 20 eV, but only the lowest levels are included in Table I. It is important to point out here that Roy and Rai assumed that the autoionizing contribution to the total cross section would be significant only for energies within 1 eV of the respective threshold energies. This "resonance-type" behavior appears to be supported by our experimental data. In general, binary-encounter theory is not accurate in predicting excitation cross sections for autoionizing levels, and the general agreement with experiment is only claimed to be within a factor of 2.

Our experimental potassium data were obtained with a crossed-beam apparatus that has been described elsewhere.[6,7] (The results in cesium were first obtained in a vapor-cell apparatus[8] and were later reproduced to within ±3% in the crossed-beam apparatus.[6]) Typically, the electron beam current was of the order of 10^{-8}–10^{-7} A, with an energy spread of 0.1 eV (FWHM). The density in the atomic beam was from 10^{9} to 5×10^{10} cm^{-3}. By using a linear array of capillaries, the region of overlap between the atomic and electron beams had a total length of 25 mm, which is much more than the few millimeters used in other experiments. This is certainly one reason why we have observed structures not resolved by other experimenters.

In order to facilitate comparison between theoretical and experimental results, illustrations on identical scales[9] have been juxtaposed, as shown in Fig. 1.

For the sake of comparison, only absolute[10] experimental results are included in this study, and we shall start the discussion with results for cesium [Fig. 1(a)]. The curves of Heil and

TABLE I. Autoionizing states in Cs, Rb, and K.

	Ground state	Lowest autoionization state	Energy (eV)
Cs	$5p^6 6s$	$5p^5 6s^2$ $^2P_{3/2}$ $^2P_{1/2}$	12.30 13.52
Rb	$4p^6 5s$	$4p^5 5s^2$ $^2P_{3/2}$ $^2P_{1/2}$	15.31 16.16
K	$3p^6 4s$	$3p^5 4s^2$ $^2P_{3/2}$ $^2P_{1/2}$	18.72 18.98

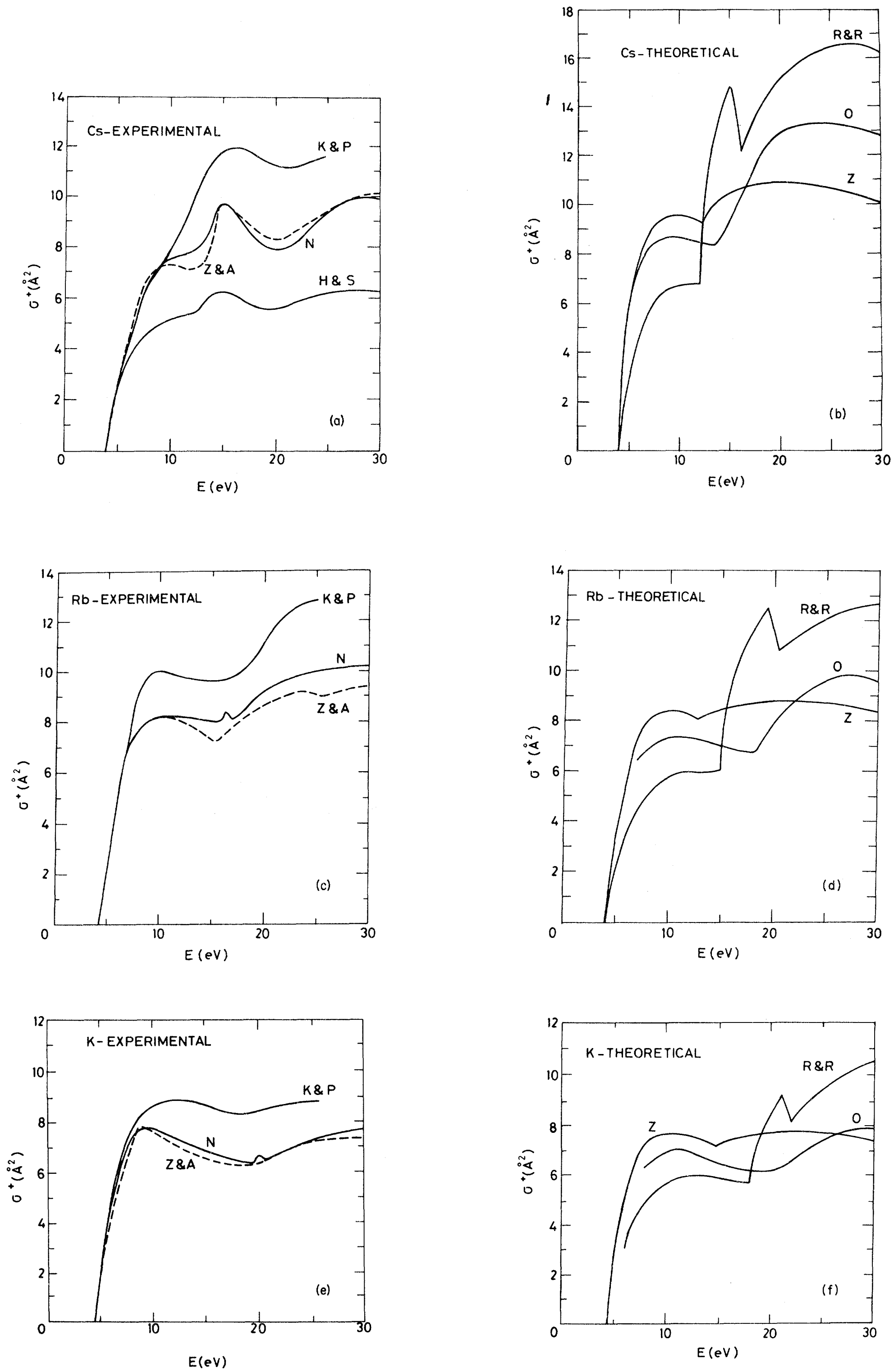

FIG. 1. Experimental and theoretical electron impact ionization cross sections in (a) and (b) Cs, (c) and (d) Rb, and (e) and (f) K. The curve labels designate the following references: Z & A, Zapesochnyi and Aleksakhin (Ref. 13); N, Nygaard (and Hahn) (Refs. 6, 8, and the new work); H & S, Heil and Scott (Ref. 11); K & P, Korchevoi and Przonski (Ref. 12); R & R, Roy and Rai (Ref. 1); Z, Zgorzelski (Ref. 15); O, Ochkur (Ref. 16).

Scott[11] and Korchevoi and Przonski[12] were obtained by passing an electron beam through a cesium vapor region. Due to uncertainties in vapor pressure and effective length of the collision region they disagree with the results of Nygaard[8] and of Zapesochnyi and Aleksakhin.[13] Both of the latter experiments employed crossed atom-electron beams. The electron gun in Nygaard's experiment was of the retarding potential difference (RPD[14]) type with an energy resolution of about 0.1 eV, while Zapesochnyi and Aleksakhin were using a 127° electrostatic monochromator with about the same resolution. There is no normalization between the curves marked N and Z & A, and the agreement in general shape is good. The maximum observed by Z & A at about 10 eV corresponds to the maximum cross section for ionization of the 6s valence electrons. This maximum was not observed by Nygaard. The reason for this is not known, since Nygaard (and Hahn) have observed structures in Rb and K that were not resolved in other experiments. The narrow maximum at 15 eV is due to autoionization and has been discussed in detail by Hahn and Nygaard.[5] The further increase in total cross section for energies above 20 eV is accounted for by the production of excited ions, some of which are excited to metastable states.[7]

The theoretical results in cesium are shown in Fig. 1(b). In addition to the calculations of Roy and Rai (R & R) we have also included those of Zgorzelski[15] and Ochkur.[16] Zgorzelski's computation was based on an improved version of Gryzinski's binary-encounter theory.[17] Ochkur's calculations[16] are also based on the binary-encounter model, but do not average the cross section over the velocity distribution of the atomic electrons. In all of the theoretical curves we note that the position of the first maximum (or "shoulder") at 10 eV is in good agreement with the experiments, and that the magnitude of this feature corresponds to the observations of Z & A and N.

The theoretical papers used different energies for ionization of 6p electrons, which explains the different sharp "onsets" in the energy range 12–14 eV. It is worth noting that Zgorzelski's calculation was based on a model with a xenon core plus a 6s-valence electron. The inclusion of autoionization by Roy and Rai results in a peak of larger magnitude at 15 eV, and is clearly overestimated as compared to the experimental curves. At higher energies Zgorzelski's results are in good agreement with the preferred experimental data, while Roy and Rai and also Ochkur give high values. We note that the position of the observed maximum at about 28 eV is in close agreement with the calculated value of Roy and Rai.

Similar results are obtained in rubidium and potassium and many of the same comments apply. In rubidium Nygaard and Hahn[6] have observed a maximum at 16.5 eV which has not been reported by other workers. This maximum is due to excitation of the autoionizing levels at 15.31 and 16.16 eV listed in Table I. Whereas the autoionization structure is hardly discernible in our experiment and has not been observed before, its contribution to Roy and Rai's total cross section has been overestimated.

In potassium we have observed a maximum at about 19 eV (reported here for the first time) which we ascribe to excitation of the $3p^5 4s^2(^2P_{3/2})$ and $3p^5 4s^2(^2P_{1/2})$ levels at 18.72 and 18.98 eV. Again, Roy and Rai have overestimated the importance of autoionization. However, their results show clearly that autoionization becomes less important for the lighter alkali metals.

The over-all accuracy for our own experimental results is ±7% for Cs, ±12% for Rb, and ±20% for K. The major contribution to the experimental error is in the determination of the atom number density, for which we used a surface ionization detector.[18]

In conclusion, we find it very valuable that Roy and Rai have made an attempt to include the effect of autoionization in the calculation of electron-impact ionization cross sections for the heavy alkali metals. However, the contributions due to autoionization and inner-shell ionization in general are too large compared to experimental data, and a more accurate theory is therefore needed.

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