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INDIRECT PROCESSES IN ELECTRON-IMPACT IONIZATION OF MULTIPLY-CHARGED IONS*

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ABSTRACT

Recent research efforts have assembled a steadily increasing array of measurements and calculations of electron impact ionization of multiply-charged ions. Significant disagreement is often found, however, between experimental results and theory due to the influence of a number of indirect processes observed in the experiments but not included in the calculations. Some of the most important of these indirect ionization processes are described and examples are discussed. New data for electron impact single-ionization of Kr^{2+} are presented in the energy range from below threshold (36 eV) to 1500 eV, and compared to related measurements and to theory.

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I. INTRODUCTION

Electron impact ionization of ions is an important area of research with applications ranging from laboratory and fusion plasmas to astrophysical studies of interstellar reaction dynamics (1). The need for more accurate ionization cross section information becomes desirable in order to understand fusion reactor processes, especially for diagnostic and impurity radiation studies, and has resulted in increased activity in electron-impact experiment and theory in recent years (2). Systematic studies have generally followed either isoelectronic (3) or isonuclear (4) series, assuming that relatively simple scaling should be possible for direct ionization either with increasing charge at fixed z or with increasing z with a fixed number of bound electrons.

As early as 1968, however, it was shown experimentally (5) that indirect ionization can dominate over the direct ionization process for singly-charged ions. It was not until 1979 that the first systematic study of indirect ionization of multicharged ions was published (3), and work has continued since then over a wide range of atomic species and for initial charge states up to 6+ in attempts to characterize indirect as well as direct ionization. This paper will briefly describe some of the indirect ionization processes under study, and present examples of each. New data for electron impact ionization of Kr^{2+} is presented and compared to theory and previous related results for ionization of Kr^{3+}.

II. INDIRECT IONIZATION

The three principal indirect ionization processes (6) which have been observed and are known to make significant contributions to total ionization may be classified as excitation-autoionization, resonant-recombination double autoionization, and ionization-autoionization. In each of these three processes, the incident electron excites or ionizes an inner-shell electron, leaving the target ion in a multiply-excited state. When the highly excited ion relaxes, the excitation energy is released through the ejection of one or more electrons, thus producing a further-ionized target ion. Of course, the highly excited ion may radiatively dispose of its excess energy, and the branching for this alternative must be considered, but lower charge-state ions will tend to autoionize whenever it is energetically possible.

A. Excitation-autoionization

If an inner-shell electron is excited during an electron-ion collision, and the resulting excited ion releases its excess energy through ejection of an electron, the process is called excitation-autoionization. This process is by far the most common indirect ionization form studied, and has the most distinctive signature. Excitation-autoionization has been found to dominate over direct ionization by a factor of 10 or more in some cases (7), and is especially important in heavy ions where more inner-shell electrons are available to contribute (8).

In cases where a single strong excitation transition to an autoionizing level is isolated in energy from other transitions, a distinctive step will be observed in the cross section. This is due to the threshold beha-

vior of the excitation cross section for an ion, which is finite and often largest at threshold and thus has a sharp onset with energy. The magnitude of the step is a direct measurement of the excitation cross section. Considering the difficulty in making absolute excitation cross section measurements on multicharged ions (9), this signature provides an excellent opportunity to test excitation calculations. In the case of Al^{2+}, for instance, the ionization measurement (10) (supported by other information) revealed that calculations (11) have greatly overestimated the importance of the 2p-3p transition, which has been under consideration for high-energy laser development. Recent theoretical work has shown that this transition is fairly sensitive to the wavefunction used and to state mixing, and that it may indeed be smaller than previously predicted (12,13). Unfortunately, isolated transitions with strong oscillator strengths are less common for ions with complex electronic structure, and experiments are more likely to reveal a continuous enhancement consisting of tens or hundreds of autoionizing transitions grouped within an energy range of 10 eV or more. Other indirect effects may also mask the sharp steps distinctive of excitation-autoionization. Detailed theoretical calculations are, however, capable of modeling the energy dependence of excitation-autoionization effects in many cases.

As an example of a typical experiment studying electron-impact ionization, including indirect effects, the ionization of Kr^{2+} teaches a number of important lessons. The experimental apparatus is shown in Figure 1, and will be only briefly described here (10,14). Ions from the ORNL-PIG ion source are extracted, analyzed, focussed, and transported to the ultra-high vacuum main interaction chamber. An electrostatic charge purifier just upstream from the collision volume removes any ions which have changed

charge in the transport system. Another electrostatic analyzer immediately downstream from the collision volume separates the main ion beam from the further-ionized signal ions.

The electron beam, which crosses the ion beam at 90°, is magnetically confined and is produced in essentially the same gun as described by Taylor et al. (15). The ion and electron currents, signal ion count rate, and beam profiles and overlap at the collision volume are all carefully measured in order to extract the absolute ionization cross section. Almost all electron-impact ionization data to date have been collected on apparatus similar to the ORNL crossed-beams apparatus (2). Details vary — for instance, inclined beams may be used instead of crossed beams, and final charge analysis may utilize magnetic instead of electrostatic fields — but despite many refinements, the principles of measurement have remained unchanged for the last 15 years (16).

The experimental results for ionization of Kr^{2+} are tabulated in Table 1 and plotted in Figure 2. Relative uncertainties are essentially the size of the plotted symbols, reflecting counting statistics at the 1 standard deviation level and relative uncertainty in the beam overlap measurement. Additional absolute uncertainties (14), such as systematic beam current and velocity measurements, detector efficiency, etc., result in an absolute uncertainty of ±8% of the peak cross section (shown in Figure 2 at 65 eV). Previously published results for Kr^{2+} are limited to a single relative measurement by Kupriyanov and Latypov (17), which was later converted to an absolute measurement (18), and a recent set of crossed beams data by Matsumoto et al. (19). The latter results agree well in shape with the ORNL measurements, but are 25% lower in absolute value.

Their assumption of unit efficiency for the microchannel plate detector could account for the discrepancy.

The ORNL data are compared in Figure 2 to calculations from the semi-empirical Lotz formula (dashed curve - ref. 20) and distorted wave (DW) calculations from Pindzola et al. (solid curve - ref. 21). Since both calculations account for direct ionization only, it may be inferred that experimental deviations from the shapes of these theories reflect the influence of indirect ionization. The distorted wave calculations in particular are expected to be a good estimate of direct ionization, and it should be noted that theory and experiment agree within the relative experimental uncertainty at energies above 120 eV. From the ionization threshold to 120 eV, however, the experimental results first rise, then fall faster than theory. This energy dependence (enhanced rise, followed by a rapid fall-off) is characteristic of non-dipole-allowed excitation followed by autoionization (22). If the excitation-autoionization involved dipole-allowed transitions, the enhancement would persist over the complete higher energy range; we observe here an enhancement with a gradual onset which appears to be reduced to insignificance by 120 eV.

The effects of indirect ionization may change dramatically along an isonuclear or isoelectronic sequence, sometimes in only a single step along the series. In recent studies of the Xe isonuclear sequence (4), it was found that in the lower stages of ionization indirect contributions were dominated by non-dipole-allowed transitions to autoionizing levels, resulting in a low-energy "hump" on top of the direct ionization cross section. This characteristic shape changes by Xe^{6+} to a strong enhancement over the entire energy range due to dipole-allowed excitations followed by autoionization. In the Kr isonuclear sequence, a rather startling change

is observed between Kr^{2+} and Kr^{3+} (Figure 3 - ref. 14). In the case of Kr^{3+}, the data are compared to the Lotz direct ionization estimate, which is in good agreement with experiment at energies of 170 eV and higher. At lower energies, however, increasing enhancement due to indirect ionization is observed from threshold to 80 eV, followed by a series of dips and rises in the cross section, and finally a rapid decline from 110 to 170 eV. The pattern of indirect ionization is quite similar to that observed in Kr^{2+}, but the effect is much more pronounced. Extrapolation of these results to higher charge states in the isonuclear sequence would lead to predictions of enormous effects. In fact, competing processes such as radiative stabilization and multiple ionization must be considered in such an extrapolation.

B. Resonant-recombination double autoionization

Due to acceleration by the Coulomb field, an incident electron may excite a target ion even when it has several eV less than the required excitation energy. The incident electron will as a result be resonantly captured by the ion into a Rydberg level, and the ion is then left in a multiply excited state. The ion may relax by ejecting the outer electron (resulting in no net change in charge), by radiative stabilization (resulting in dielectronic recombination), or, if an inner-shell electron was excited, by ejection of two or more electrons (resulting in a net single or multiple ionization event). The last alternative is a form of indirect ionization. In experiments with the energy resolution available today, the sharp onset of an isolated excitation-autoionization feature is often observed to "soften," with an onset at energies up to several eV lower than expected and more gradual increases in cross section than is

usually characteristic for an excitation feature. One clear example of this process is seen in the near-threshold cross section for ionization of Xe^{3+} (Figure 4 - ref. 4). The dominant excitation-autoionization contributions are due to 4d-nℓ transitions (where nℓ = 4f, 5p, 5d, and 5f), and direct ionization plus the distorted wave calculations for these transitions are observed to account for the experimental cross section (14). Considerable indirect ionization is observed, however, at energies below the predicted thresholds (40 to 55 eV and 70 to 73 eV) for these transitions, and this "smearing" of the onsets is attributed to resonance-recombination double autoionization. The effects of this indirect process are not dramatic, but may be important since the resulting blending or smearing of isolated features may make data analysis and interpretation more difficult.

C. Ionization-autoionization

Direct ejection of an inner-shell electron leaves the target ion excited. If two or more electrons are left in the outer orbitals, and if the excess energy is sufficient, autoionization will take place and the initial ejection of a single electron will result in a net multiple ionization event. As an example (refs. 23, 24), removal of a 2p electron from Ar^{2+}, Ar^{3+}, or Ar^{4+} leads to double ionization after the autoionization of one of the outer 3p electrons. The significance of indirect multiple ionization is twofold: first, some single ejection events do not result in only single ionization. The interpretation or planning of experiments or diagnostic measurements may be greatly influenced by unexpected jumps in ion charge state. Second, multiple ionization of multiply-charged ions is much more likely than has been predicted by direct calculations (25); it is

in fact as likely as the single ionization which triggers the autoionization chain, and becomes relatively more important as the ionic charge increases.

III. CONCLUSIONS

Indirect effects in electron impact ionization range from negligible to dominant. Careful and thoughtful contemplation of the detailed energy levels for a particular ion, however, should enable us to make at least a rough prediction of the likely importance of indirect effects for that ion. Isoelectronic and isonuclear trends are not yet usually predictable, especially for higher charge states where we have no experiments for guidance. Detailed calculations, either distorted wave or close-coupling, are often accurate but are too complex and expensive for use on all charge states of all elements. In the future, continued close cooperation between experiment and theory will be essential to the extension of studies to higher charge states, a vital step in understanding the systematics of indirect ionization. In addition, future trends may include the measurement of differential (in angle and/or energy) cross sections for electron-impact ionization, a stringent test both of theory and of experimental ability.

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Table 1. Experimental electron-impact ionization cross sections for Kr^{2+}. Uncertainties are 1 standard deviation (relative only). Absolute uncertainty near the peak cross section is ±8% at good confidence level.

Energy (eV)	Cross Section (10^{-18} cm^2)	Energy (eV)	Cross Section (10^{-18} cm^2)
25.6	0.24 ± 0.37	94.8	73.6 ± 0.6
30.6	0.09 ± 0.12	100.6	72.3 ± 0.8
33.4	0.38 ± 0.24	110	73.6 ± 0.8
35.6	1.02 ± 0.16	115	72.2 ± 0.8
36.6	4.30 ± 0.27	120	71.6 ± 0.8
37.7	13.89 ± 0.45	129	68.0 ± 0.8
38.4	19.54 ± 0.37	139	65.2 ± 0.8
39.6	25.09 ± 0.40	149	64.7 ± 0.8
40.5	29.07 ± 0.29	168	62.9 ± 0.7
43.4	42.68 ± 0.53	193	59.5 ± 1.3
45.4	50.37 ± 0.29	222	55.1 ± 0.7
47.4	56.16 ± 0.61	252	54.0 ± 0.6
49.5	59.70 ± 0.69	291	48.5 ± 0.5
50.5	61.47 ± 0.68	340	43.0 ± 0.5
53.2	64.74 ± 0.84	390	37.8 ± 0.4
55.3	66.97 ± 0.34	439	34.7 ± 0.4
60.6	71.5 ± 0.7	489	32.0 ± 0.2
65.3	73.9 ± 0.6	588	29.0 ± 0.3
71.1	75.8 ± 1.0	687	27.6 ± 0.2
74.9	77.1 ± 0.9	835	23.1 ± 0.3
81.1	76.6 ± 0.8	984	19.1 ± 0.2
84.8	76.1 ± 0.8	1232	16.1 ± 0.3
90.9	73.6 ± 0.8		

FIGURE CAPTIONS

Figure 1: Schematic of the ORNL crossed-beams apparatus.

Figure 2: Cross section for electron impact single-ionization of Kr^{2+}; the absolute uncertainty is plotted at 65 eV. The curves are direct ionization predictions from the Lotz formula (dashed curve - ref. 20) and distorted wave calculations from Pindzola (solid curve - ref. 21).

Figure 3: Electron-impact ionization of Kr^{3+}. The data are from Gregory, Dittner, and Crandall (14); the absolute uncer ainty is shown at 105 eV. Dashed curve is from the Lotz formula (20).

Figure 4: Electron-impact ionization cross section for Xe^{3+} near threshold. Data is from Gregory, Dittner, and Crandall (14); the absolute uncertainty is shown near 75 eV. The curves are distorted wave calculations (4) for direct ionization (dashed curve), excitation-autoionization added to the direct prediction (solid curve) and a convolution of the total ionization theory with the estimated experimental energy resolution (short-dashed curve). Indirect ionization calculations include excitation of 4d-electrons to the indicated autoionizing levels, with onsets in the indicated energy ranges.

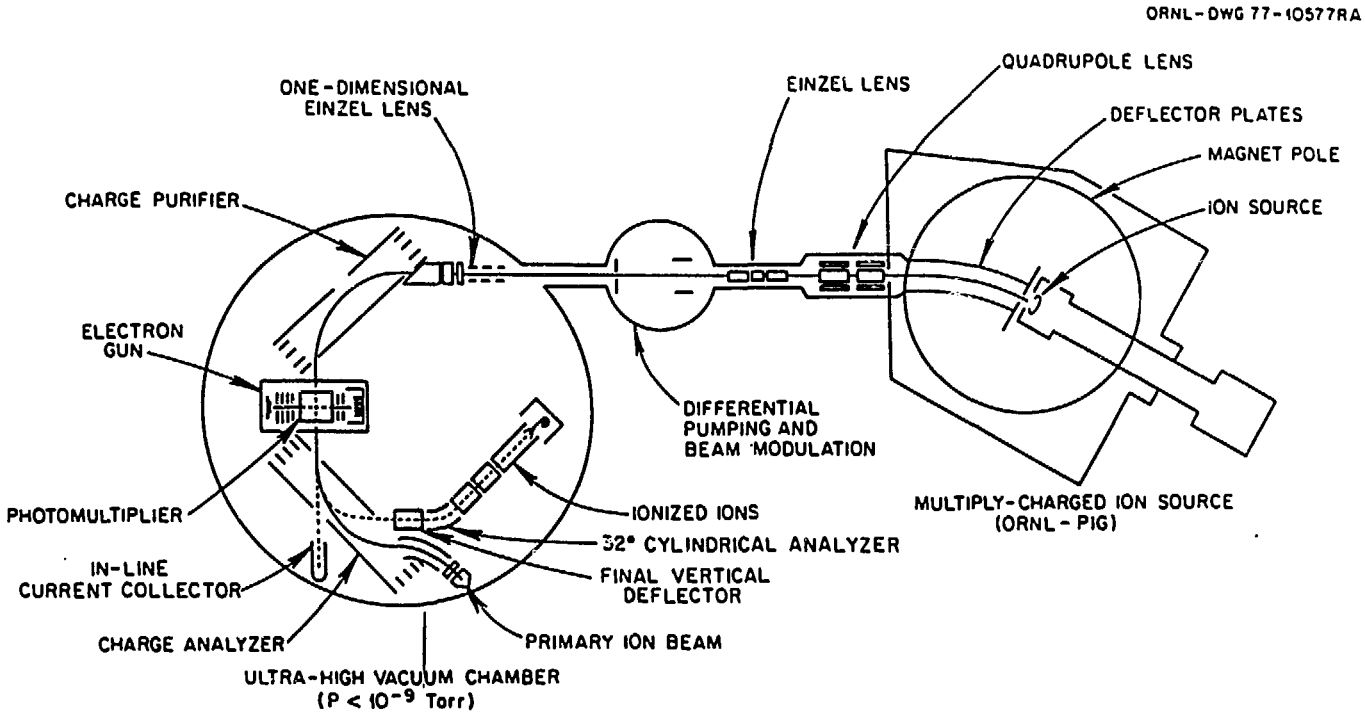
ORNL-DWG 77-10577RA
ONE-DIMENSIONAL EINZEL LENS
EINZEL LENS
QUADRUPOLE LENS
DEFLECTOR PLATES
MAGNET POLE
ION SOURCE
CHARGE PURIFIER
ELECTRON GUN
DIFFERENTIAL PUMPING AND BEAM MODULATION
MULTIPLY-CHARGED ION SOURCE (ORNL-PIG)
PHOTOMULTIPLIER
IONIZED IONS
32° CYLINDRICAL ANALYZER
IN-LINE CURRENT COLLECTOR
FINAL VERTICAL DEFLECTOR
CHARGE ANALYZER
PRIMARY ION BEAM
ULTRA-HIGH VACUUM CHAMBER
(P < 10^-9 Torr)

Fig. 1

ORNL-DWG 84-14629

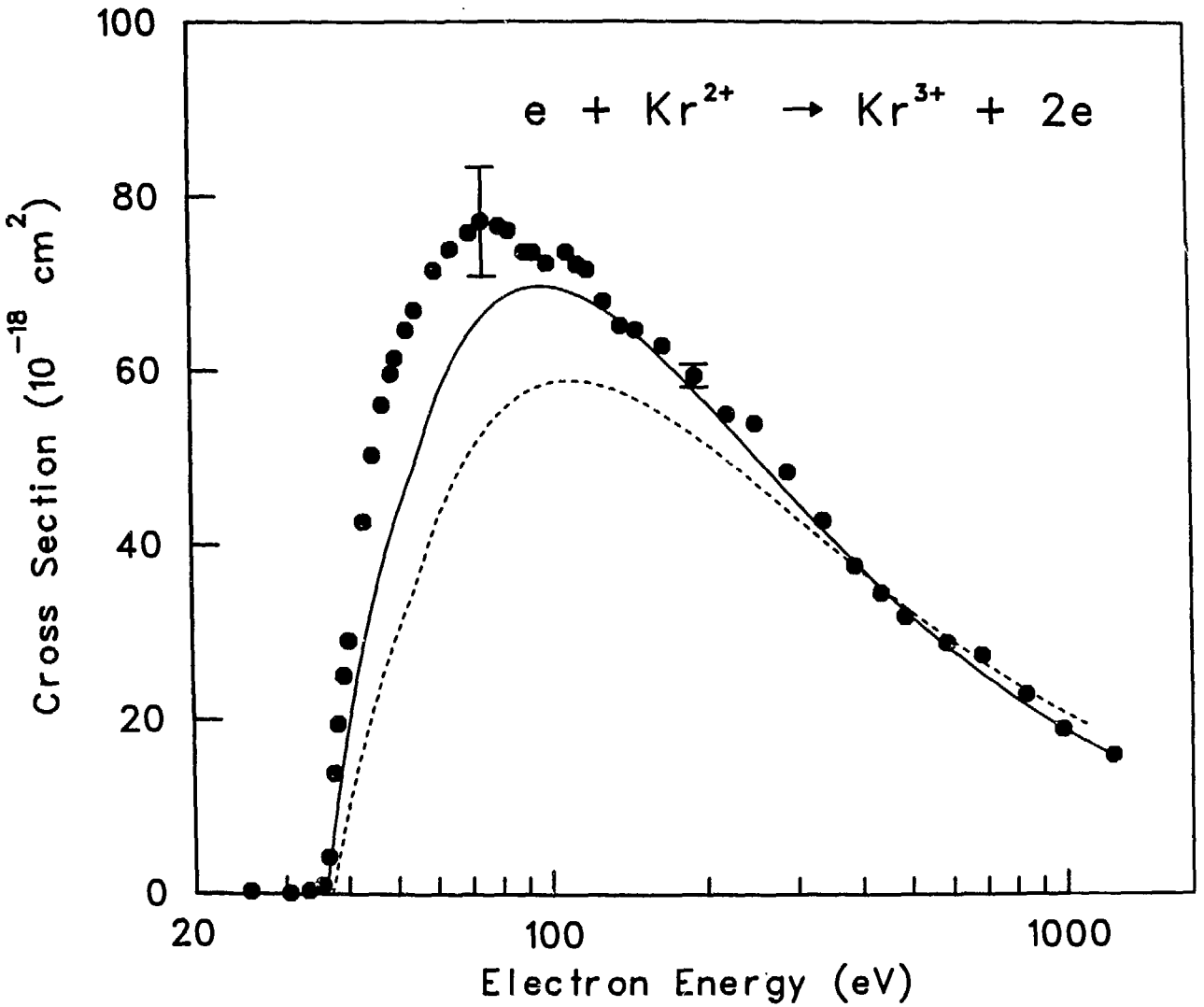

Fig. 2

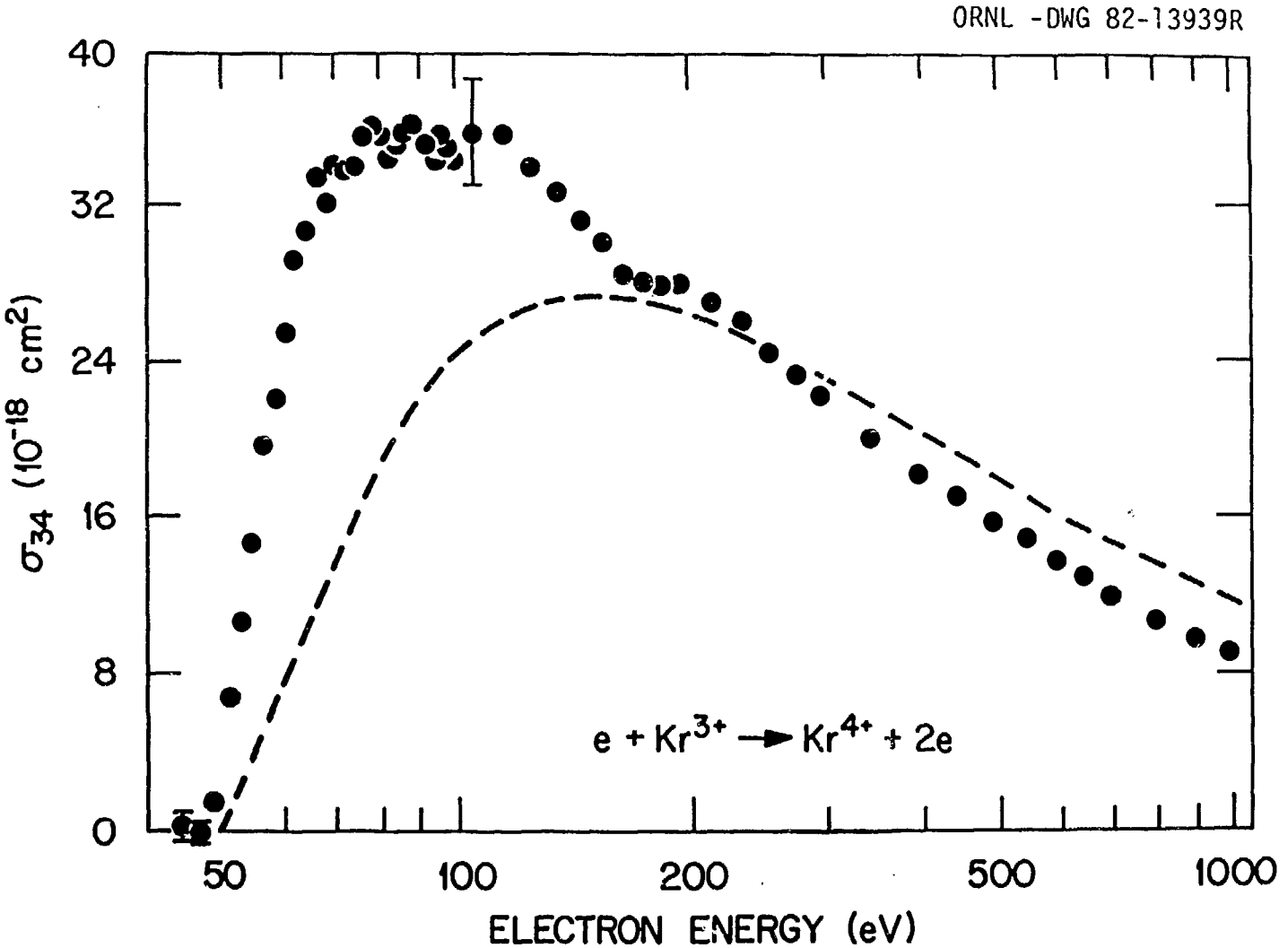
ORNL -DWG 82-13939R
40
32
24
16
8
0
σ_{34} (10^{-18} cm^2)
50
100
200
500
1000
ELECTRON ENERGY (eV)
$e + Kr^{3+} \rightarrow Kr^{4+} + 2e$

Fig. 3

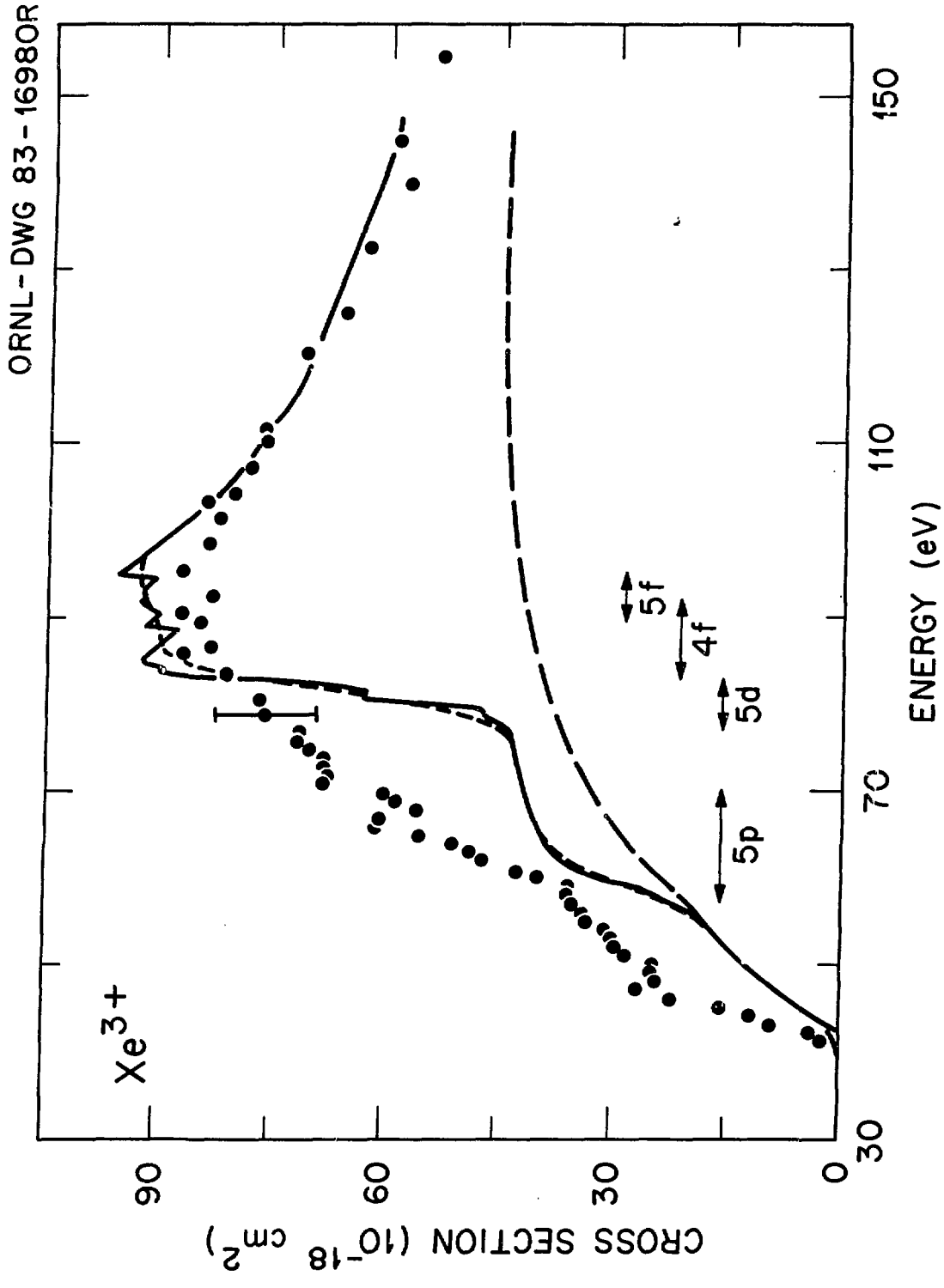
ORNL-DWG 83-16980R
Xe3+
5p
5d
4f
5f
CROSS SECTION (10-18 cm2)
ENERGY (eV)
0
30
60
90
30
70
110
150

Fig. 4