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THE ROLE OF INDIRECT PROCESSES IN ELECTRON-IMPACT IONIZATION OF MULTICHARGED IONS*

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The Role of Indirect Processes in Electron-Impact Ionization of Multicharged Ions*

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Recent studies of electron impact ionization in this laboratory are discussed with emphasis on the role of indirect processes. Examples are used to illustrate the importance of excitation-autoionization (Ti^{2+} and Al^{2+}), autoionizing metastable ions in the Na isoelectronic sequence, multiple ionization (triple ionization of Xe^{6+}), the relative importance of indirect ionization with increasing charge in the Mg isoelectronic sequence, and the extension of ionization measurements to higher-charged target ions (Fe^{9+}).

1. INTRODUCTION

Electron impact ionization is of primary importantance to the understanding of laboratory plasmas through power balance calculations, analysis of diagnostics, edge plasma studies, and other interpretative research (1). In addition, there is basic interest in the varied physical processes which result in ionization. One of the principal aims of our laboratory has been the measurement and interpretation of cross sections for electron impact ionization of multicharged ions. A large number of cross sections have been measured to test existing theories (or to provide a base for comparison with planned calculations) . Our measurements have concentrated on systematic studies of isoelectronic (same number of electrons with varying elements in appropriate charge states), isonuclear (same element with varying charge states), and isoionic (ions with similar outer electron configurations) target ion groups (2).

The crossed beams apparatus currently in use for electron impact ionization measurements ot ORNL is shown in Figure 1. In the main interaction chamber, the incident ion beam is focussed and then bent by 90° by the charge purifier. It then passes through the interaction region, where it is intersected at right angles by a magnetically-confined electron beam (3). The ion beam is then separated into charge-state components in the post-collision analyzing magnet. Incident ion beam current is measured by collection in one of two Faraday cups while signal (further ionized) ions are counted in a channeltron electron multiplier.

The new analyzing magnet extends our potential range of measurements of single ionization to ions with incident charge states up to +14. This extended range was intended to match the capabilities of the new ORNL-ECR ion source which has recently been commissioned for full-time use on atomic physics experiments (4); the source has in fact already surpassed its expected ultimate performance criteria for beam intensity and charge state output. Details of the previous ion source and other experimental details are available in previous publications (5,6).

A number of indirect processes must be considered to fully understand observed ionization cross sections (7). In addition to direct ionization of an outer shell electron, e.g.,

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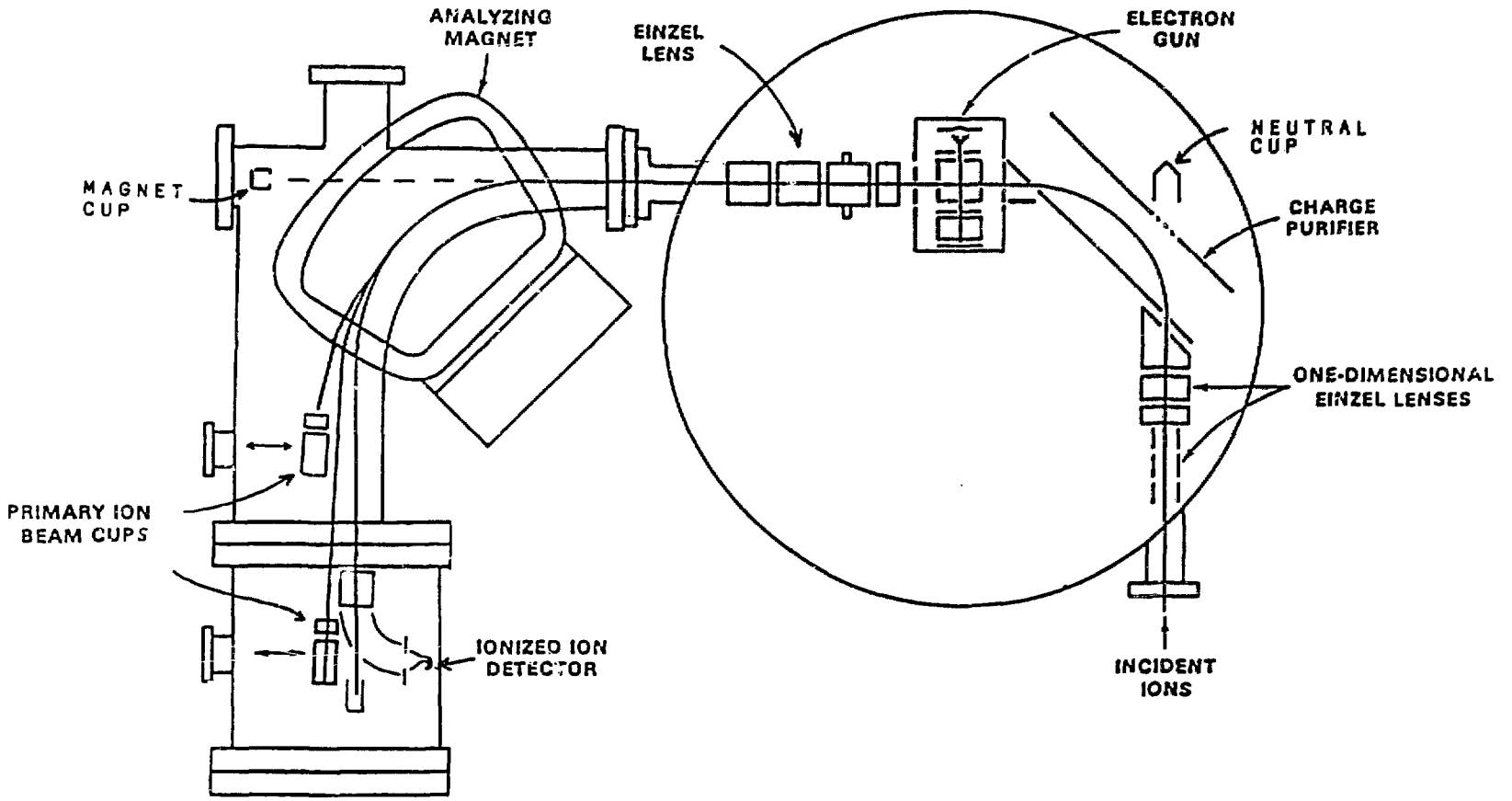

FIGURE 1
ORNL Crossed Electron-Ion Beams Apparatus.

$$e + Xe^{6+}(4d^{10}\,5s^2) \rightarrow Xe^{7+}(4d^{10}\,5s) + 2e, \tag{1}$$

removal of an electron from the target ion may occur through excitation of an inner-subshell electron followed by autoionization, as in

$$e + Xe^{6+}(4d^{10}\,5s^2) \rightarrow Xe^{6+}(4d^{9}\,5s^2\,n\ell) + e$$
$$\hookrightarrow Xe^{7+}(4d^{10}\,5s) + e. \tag{2}$$

Detailed knowledge of the energy level scheme for the target ion is generally necessary in order to make an accurate evaluation of which excited states will autoionize and which are trapped in the incident ion charge state, but rough guesses can often be made based on available inner-subshell ionization energies (8). Net multiple ionization may also result from excitation or ionization of an inner-shell electron:

$$e + Xe^{6+}(3d^{10}\,4s^2\,4p^6\,4d^{10}\,5s^2) \rightarrow Xe^{7+}(3d^{9}\,4s^2\,4p^6 4d^{10}\,5s^2) + 2e$$
$$\hookrightarrow Xe^{9+}(4d^{9}) + 2e. \tag{3}$$

In this example, direct ionization of a 3d electron leads eventually to triple ionization. Careful accounting of possible branching paths for ions with inner-shell holes must be used to optimize the usefulness of any theory, whether it is a detailed distorted-wave or close-coupling calculation or the simple Lotz formula.

Two examples of indirect ionization due to excitation-autoionization will be discussed followed by an example of multiple ionization. Recent preliminary cross section measurements on a highly-charged ion will be presented along with a discussion of our future plans.

2. EXCITATION-AUTOIONIZATION

The effects of excitation-autoionization may range from negligible to dominant, and we are still learning how to make intelligent guesses of its importance to total ionization in a given case. For this reason, a large number of comparisons between experiment and theory seems the only way to develop an understanding of the "rules" that govern which transitions and processes are important. In cases where the transitions leading to autoionizing states are well-separated in energy, the experimental data may provide an indirect measure of the excitation cross sections to compare with detailed calculations.

One of the measurements which drew our attention to the importance of excitation-autoionization was the cross section for single ionization of Ti^{3+} (9). Indirect ionization dominates over the direct process by a factor of up to ten over the entire energy range from threshold to high energies. A subsequent study (10) of the cross section for single ionization of Ti^{2+} is shown in Figure 2 along with direct ionization calculations from Pindzola et. al. (11) and the three-parameter Lotz formula (12). It is obvious that total ionization is almost uniformly twice the direct cross section, so that the direct and indirect contributions must be approximately equal. In such cases where widely used formulas (like the Lotz semiempirical formula) underestimate total ionization by a large factor for several successive charge states of an important element, any plasma modeling or diagnostic analysis based on these formulas will be very misleading and may lead to conclusions which are seriously in error.

Ionization of Al^{2+}, shown in Figure 3, has been the subject of controversy for several years (13). Detailed calculations (14) of the expected contributions of excitation-autoionization (upper solid curve) appear to overestimate

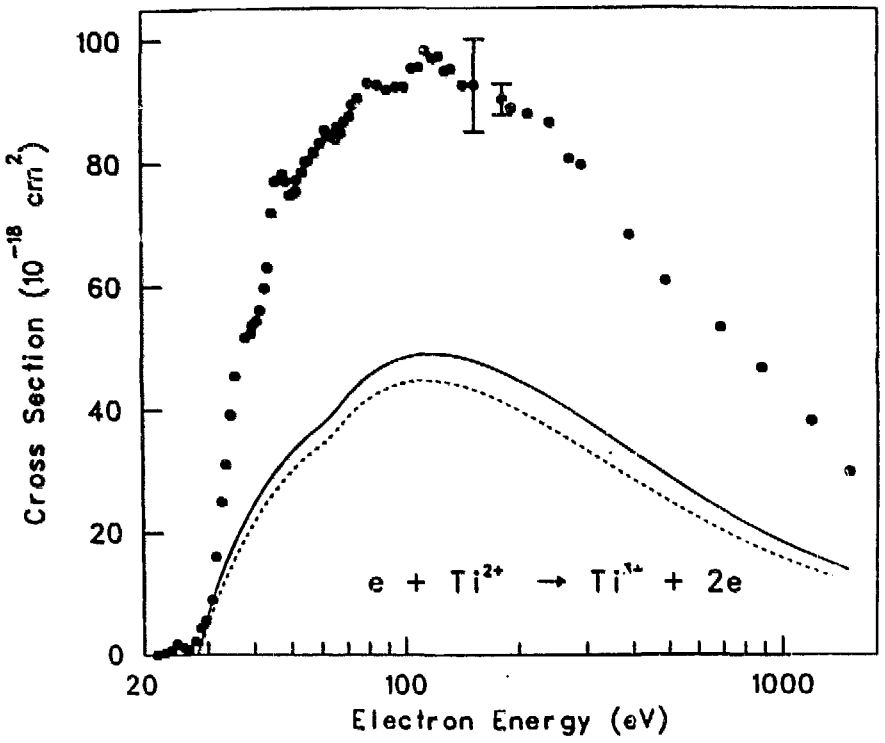

FIGURE 2
Electron impact ionization of Ti^{2+} (from Ref. 10). The solid and dashed curves are thought to be reasonable estimates of direct ionization; direct and indirect ionization contribute approximately equally to the total cross section.

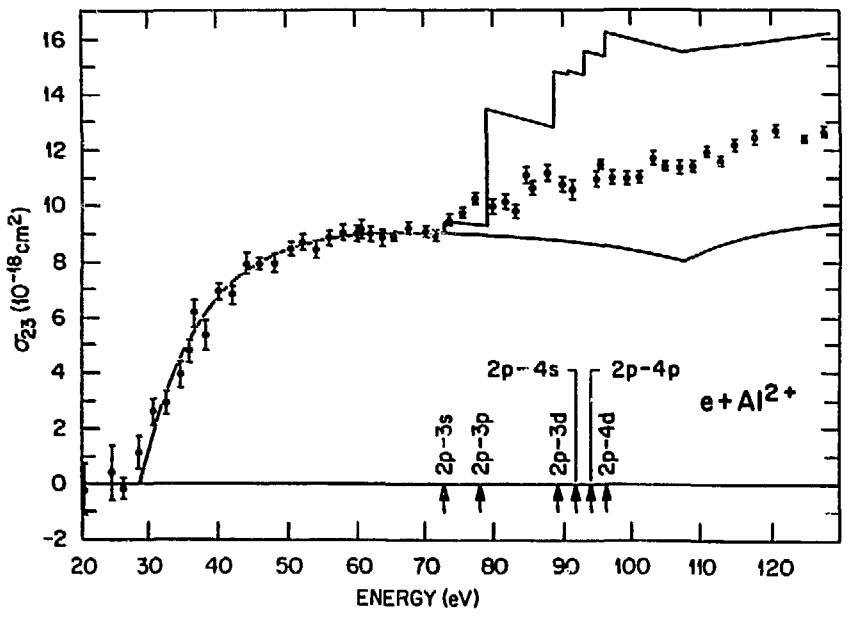

FIGURE 3
Electron impact ionization of Al^{2+} in the threshold energy region (from Ref. 13). It is now thought that the predicted 2p-3p excitation-autoionization feature does not contribute to the measured cross section because the excited ion is trapped in a metastable autoionizing state.

the indirect contribution by almost a factor of two, and although the overall disagreement is not large, this is an excellent test case for comparison between theory and experiment. Al^{2+} is also a member of the Na isoelectronic sequence which is critical to plasma diagnostics and X-ray laser development. The transitions expected to lead to single ionization are listed on the figure along with arrows indicating the excitation onset energies. A number of effects may lead to a "washing-out" of the sharp onsets indicated in the theory curve, but some fairly sharp rise should be observed at least in the case of the strongest transition (2p-3p). It should also be noted that elimination of this one transition would bring experiment and theory into good agreement.

Recent measurements and calculations of ionization in the neighboring Mg isoelectronic sequence (15,16) have led to reasoning which explains the results of both sequences; without this explanation neither sequence can currently be explained. If the excitation of a 2p electron from the ground state of Al^{2+} leaves the resulting excited Al^{2+} ($2p^5$ 3s3p) ion in one of the metastable states of this configuration, the subsequent autoionization event might not occur for several microseconds; this delay is long enough to eliminate that ion from our signal channel and probably long enough in a plasma to allow other collisions which will alter the ion configuration once again. Thus the "missing" transition may only be trapped in an unexpected metastable state. In the Mg sequence, ionization from metastable target ions leave a similar metastable ion; this ionization event is detected in our signal channel even though it should autoionize and result in net double ionization. These cases provide an excellent example not only of cooperation between experiment and theory, but also of seemingly unrelated discrepancies leading to a single unified explanation.

The relative importance of indirect ionization with increasing charge along an isoelectronic sequence is illustrated in Table 1 where the peak direct and estimated peak indirect cross sections for some members of the Mg isoelectronic sequence are compared. While the peak direct cross section decreases rapidly with increasing charge, the peak indirect cross section remains almost constant over the range studied. This only means that the energy required and probability of exciting (or ionizing, for that matter) an inner-shell electron is not very sensitive to the exact number of outer-shell electrons. As the charge state increases, however, competing processes (such as radiative stabilization in the case of excitation-autoionization) will eventually change the relative importance of the various indirect processes. We may discover new processes or new significance for processes thought to be negligible as ionization measurements are extended to significantly higher charge states.

TABLE 1

Direct and indirect ionization peak cross sections in Mg-like ions (Ref. 15).

	Direct Peak (10^{-18} cm^2)	Indirect Peak (10^{-18} cm^2)	Indirect/Direct Ratio
Al^{+}	75	3.0	0.04
S^{4+}	5.0	2.5	0.5
Cl^{5+}	2.5	2.5	1.0
Ar^{6+}	2.0	2.2	1.1

Attempts to extend measurements in the Na isoelectronic sequence to higher charge states, however, encountered unexpected difficulties. Figure 4 shows the background measured for a number of Na-like ions. This background is generally dominated by ions in the target beam which have lost one electron in a collision with the residual gas in the main chamber. In order to minimize this effect, the ion beam is charge-analyzed just before the interaction volume; the normal background rate is then 1-10 counts/sec/particle nA. The dramatic increase in background seen in Figure 4 with increasing charge state has been determined to be due to target ions which leave the ion source in one or more autoionizing metastable state(s) of the $2p^5\ 3s3p$ configuration. A crude measurement in our apparatus indicates a mean lifetime in the 1-10 microsec. range for S^{5+}; a lifetime very much longer would result in few autoionizing events in our interaction region, while states with much shorter lifetimes would have almost all decayed before reaching our charge-state purifier. Since the lifetimes of these states are predicted to become shorter with increasing charge, it is hoped that ionization of even higher-charged Na-like ions will again be manageable. Similar spontaneous autoionizing ion beams have been observed in a similar apparatus in Belgium (17) and they confirm that their background count rate is lower for Ar^{7+}. Our laboratory is also considering experiments which could take advantage of these unusual autoionizing ion beams.

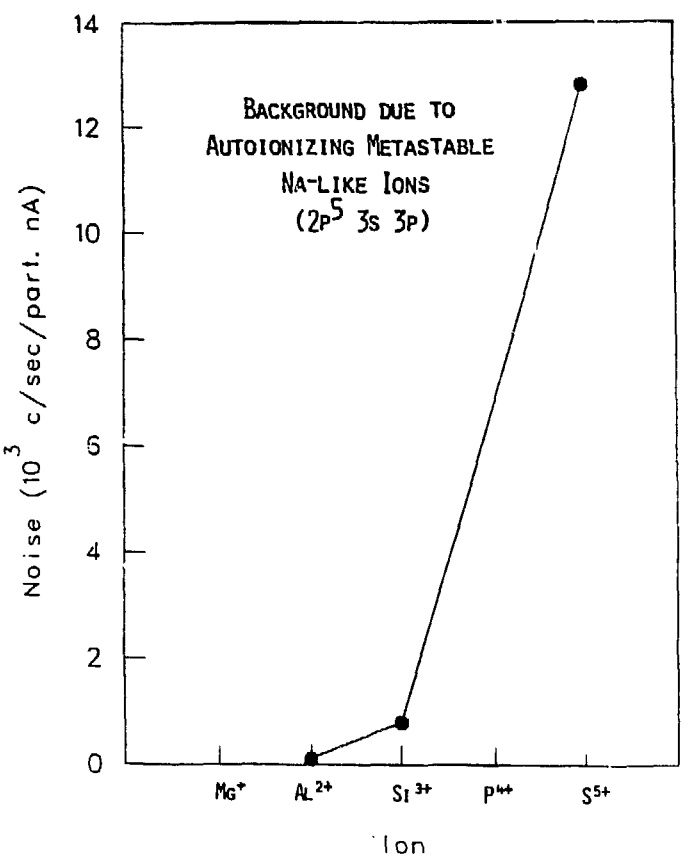

FIGURE 4

Background in ionization measurements on Na-like ions. Autoionizing metastable ions in the target ion beam are responsible for the high background at higher charge states.

4. MULTIPLE IONIZATION

The number of physical events that can result in multiple ionization is much greater than for single ionization. Excitation or removal of a single inner electron will often lead to multiple ionization as the resulting highly excited ion relaxes. In addition, multiple electron processes from a single collision can also lead to further autoionization or result in an ion trapped in its original charge state. The large number of possibilities means that considerable care must be given to studying energy levels and decay paths in order to predict which processes will be important for a given multiple ionization. The relative lack of experimental data and the difficulties involved in calculating multielectron processes has left us with considerably less data (both theoretical and experimental) to compare. The only readily available theory (18) which can be applied by the casual user is based on binary encounter approximation (BEA), and it is not immediately clear how to apply even this theory if the user wishes to take subshells into account.

As an example of recent measurements of multiple ionization, the cross section for triple ionization of Xe^{6+} is shown in Figure 5. This measurement (19) presents a rare opportunity to observe a two-electron process. In almost every previous multiple ionization study, the complete cross section curve could be explained in terms of single electron processes which result in final multiple ionization. Since such first-order processes are expected to strongly dominate over multiple-electron contributions, it is difficult to compare or isolate the two effects. Triple ionization of Xe^{6+}, however, can only occur

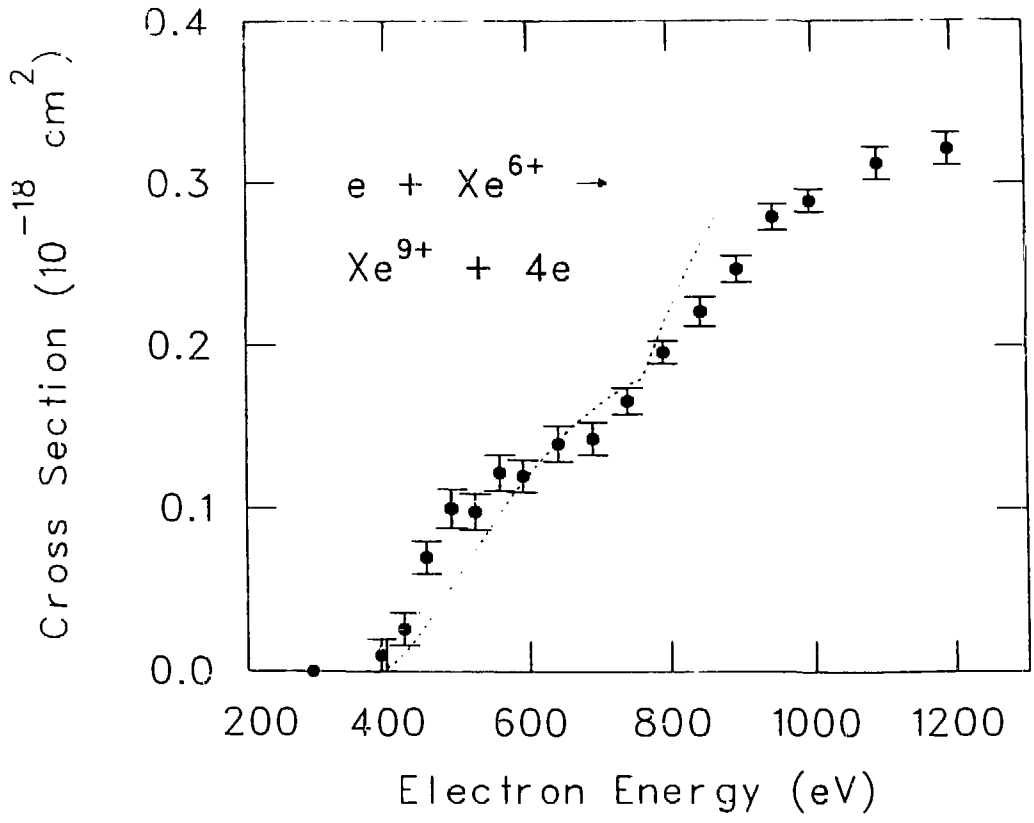

FIGURE 5

Triple ionization of Xe^{6+} (from Ref. 19). Ionization in the energy range from 374 to 670 eV can only be due to multiple-electron excitation or ionization in a single collision. The dashed curve is the sum of contributions from direct double ionization (Ref. 18) and direct ionization of a single, inner-shell electron (Ref. 12) followed by autoionization.

due to a one-electron process above 670 eV, while removal of three electrons only requires 374 eV. The dominant contribution to triple ionization is expected to come from direct ionization of a single 3d inner-shell electron which onsets at 762 eV. As can be seen from Figure 5, the cross section onsets near 374 eV (the lowest allowed energy), and a break in the slope of the data can be seen where the expected direct inner-shell ionization onsets. If the trend of the low-energy data is extended, it can be seen that the peak contributions from the multiple-electron and single-electron processes are approximately equal. This is a surprising result since single ionization is expected to be dominant. The dashed curve is the summed result of double direct ionization calculations based on Gryzinski's BEA model (18) and Lotz (12) calculations for direct ionization of a single 3d electron. Direct double ionization (followed by autoionization) is expected to be the dominant contribution in the multiple-electron region, but the good agreement between BEA calculations and experiment are considered fortuitous since a number of approximations were necessary to adapt the BEA theory to this situation.

Although only a few multiple ionization systems have been studied in our laboratory, the new ion source and experimental apparatus will make such measurements much easier and systematic studies possible in the future. Multiple ionization should be considered an important part of total ionization as well as an interesting physical process, and we intend to carry out additional studies in the future.

5. FUTURE PLANS

In recent years we have measured numerous cross section curves for ions of initial charge +2 through +6, almost all as part of systematic studies. We plan to extend measurements to higher charge states using the new capabilities of the ORNL-ECR ion source and the improved experimental apparatus. As an example of the possibilities presented by this combination, Figure 6 shows preliminary measurements of single ionization of Fe^{9+} which were collected in a

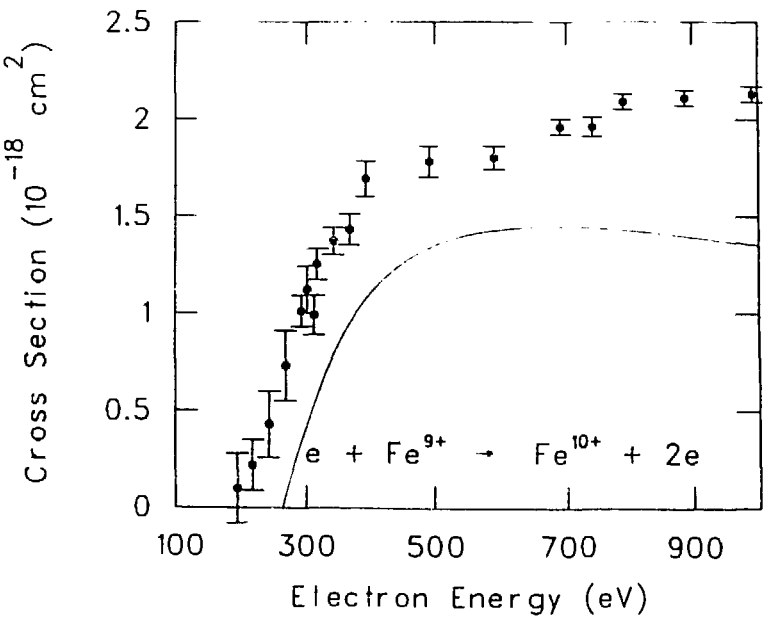

FIGURE 6
Preliminary measurements of electron impact ionization of Fe^{9+} (Ref. 20). The solid curve is distorted-wave theory from Younger (Ref. 21). Most of the target ion beam is apparently in a metastable state about 100 eV above the ground state.

few hours recently (20). Although the data are not final and complete diagnostics were not performed as is customary, it appears that the incident ion beam is mainly trapped in a metastable level approximately 100 eV above the ground state. This measurement typifies the new capabilities of such experiments, since no crossed beams data has been published for ions of initial charge state greater than +6. The ECR ion source produced a 500 nA beam of Fe^{15+} in a recent test, so that it appears that we are limited in charge state only by the current experiemntal apparatus, which was designed for incident ion beams up to charge +14.

In order to answer the needs of the fusion community, our measurements will concentrate in the near future on metallic ions, particularly on Fe and other likely plasma contaminants. Some multiple ionization studies will be used to complement our understanding of single and total ionization. Long range plans could involve analysis of the direction and/or energy of scattered (or ejected) electrons. Experiments on state-selected incident ion beams (or state-analysis of the signal ionized ions) are also exciting long-range possibilities. We intend to continue our coordination and cooperation with other laboratories involved in similar research, and we will continue to address the needs of the physics community in general and of the fusion community in particular.

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