CONF-8609169-1

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CONF-8609169--1

DE87 000352

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Manuscript for presentation to the
Joint Workshop of the US-Japan Fusion Collaborative Program

"Roles of Atomic and Molecular Processes in Plasma Modelling and Diagnostics — Resonance Effects in Electron-Ion Collisions"

Institute of Plasma Physics
Nagoya University
Japan

September 1-2, 1986

Research Sponsored by the Office of Fusion Energy
of the U.S. Department of Energy
under Contract No. DE-AC05-84OR21400
with Martin Marietta Energy Systems, Inc.

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The important role of indirect-ionization mechanisms in electron-impact ionization of multiply charged ions has been emphasized by some recent experiments conducted with the ORNL-ECR multicharged ion source. Illustrative examples of investigations of the Mg-isoelectronic and Fe-isonuclear sequences are presented and compared with the results of detailed theoretical calculations. New experimental data is also presented concerning the role of resonance effects in the ionization of Li-like O^{5+} and Na-like Fe^{15+} ions.

I. INTRODUCTION

Ionization by electron impact is a complex process, since a large number of mechanisms can cause the ejection of electrons from bound states of atomic systems. As an atom becomes more highly ionized, fewer electrons remain to screen the nucleus, and its binding energy increases. Since the direct or "knock-out" ionization cross section scales roughly as the inverse square of the binding energy, the direct ionization cross section becomes progressively smaller as the charge state of a given ionized atom increases. Indirect pathways to ionization begin to compete with direct ionization and even to dominate the ionization of many highly charged ions. Mechanisms which involve resonances and which may appear at first consideration to be exotic pathways to ionization have been predicted to make measurable contributions to the ionization of some highly charged ions.

II. IONIZATION MECHANISMS

The most important indirect ionization mechanism is the so-called excitation-autoionization process. The incoming electron excites an inner-shell electron, leaving the ion in a core-excited state which can subsequently decay by autoionization, resulting in a net ionization process. Depending on the excitation energy of the core-excited state and the branching ratio for its decay, the ejection of more than one electron may be likely, leading to a net multiple ionization process. The cross section for excitation of an ion has the distinguishing characteristic of being finite and often largest at its threshold energy. Thus a particular inner-shell excitation can produce an abrupt jump in the ionization cross section at the threshold for the excitation process. Careful measurements of the energy dependence of the ionization cross section can thus provide quantitative information about excitation processes as well.

A core-excited state may also be produced by the direct ejection of an inner-shell electron from an ion. Subsequent release of another electron by autoionization then leads to a net double ionization event. If the collisionally-ejected core electron is from a more deeply-lying shell, the decay of the vacancy state may result in the ejection of several electrons via the Auger process. This ionization-autoionization process is the most important multiple-ionization mechanism for most ions. It is generally not considered to be an indirect ionization mechanism, since the collisional process just involves the direct ejection of an inner-shell electron.

A somewhat more exotic indirect-ionization pathway has as its first step the same resonant process that leads to dielectronic recombination. In this case, an incident electron approaches the ion with just slightly less than the minimum energy required to excite an inner-shell electron, but due to acceleration in the ionic Coulomb field, gains enough energy to excite the transition. In so doing, it becomes captured into a doubly excited state of the ion which can decay by a number of pathways, including single Auger emission, double Auger emission, or radiative decay. The first leads to a scattering resonance, the second to ionization and the third to dielectronic recombination. This is a highly resonant process which can occur only for certain values of the electron energy, since there must exist a doubly-excited state of the ion into which the incoming electron can be captured. The process leading to a net ionization event has been called resonant-excitation-double-autoionization (REDA), and was first postulated by LaGattuta and Hahn[1] in 1981 as playing an important role in the electron-impact ionization of Na-like Fe^{15+}.

The mechanisms of direct single ionization, excitation-autoionization, ionization-autoionization and resonant-excitation-double-autoionization are all generally considered to be first-order collisional processes. In each case, the incident electron interacts through the two-body Coulomb interaction to promote a single target electron during the collision.

III. EXPERIMENTAL METHOD

Cross sections for electron-impact ionization of multiply charged ions have been measured using the intersecting-beams technique by a number of groups[2-5], and a schematic of the apparatus currently in use at Oak Ridge[2] for such experiments is shown in Figure 1. A mass/charge analyzed beam of multiply charged ions is directed into an ultra-high vacuum chamber, deflected through 90 degrees to remove ions which may have changed charge in flight from the ion source, and intersected by an electron beam of variable energy. The emerging ion beam then enters a double-focusing magnetic spectrometer which separates the ionized ions from the parent beam. The ionized ion "signal" is further deflected and counted by a particle multiplier, while the parent-ion and electron beams are collected in

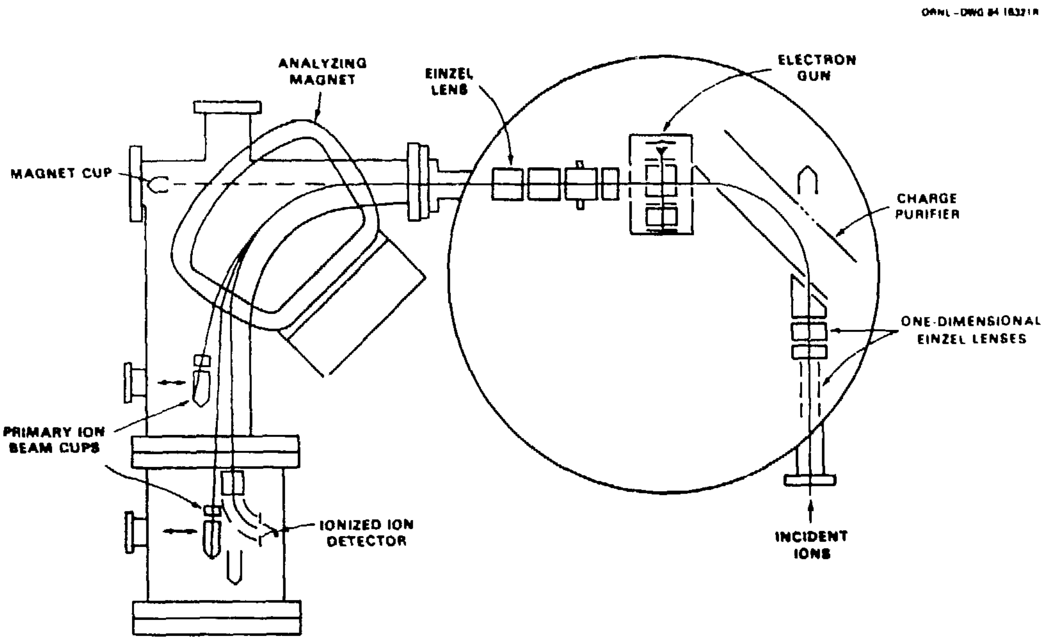

Figure 1. Schematic of electron-ion crossed-beams apparatus used to study electron-impact ionization of multiply charged ions at ORNL.

Faraday cups. The recently implemented magnetic charge analyzer system has permitted the resolution of initial/final charge ratios ranging from 4/5 to 15/16.

IV. EXCITATION-AUTOIONIZATION OF MAGNESIUM-LIKE IONS

A recent series of ORNL-JILA collaborative experiments[6] and a parallel theoretical investigation[7] has focused on electron-impact single ionization of Mg-like ions. In Figure 2, the experimental cross-section data for ionization of S^{4+}, Cl^{5+} and Ar^{6+} are compared to theoretical and semiempirical predictions for direct 3s outer-shell ionization. The cross-section measurements show abrupt changes in slope near 150, 200 and 250 eV for S^{4+}, Cl^{5+} and Ar^{6+}, respectively. These are signatures of the onset of 2p-nl inner-shell excitation-autoionization, which may be seen to increase in importance relative to the direct 3s outer-shell ionization as the ionic charge increases along the sequence.

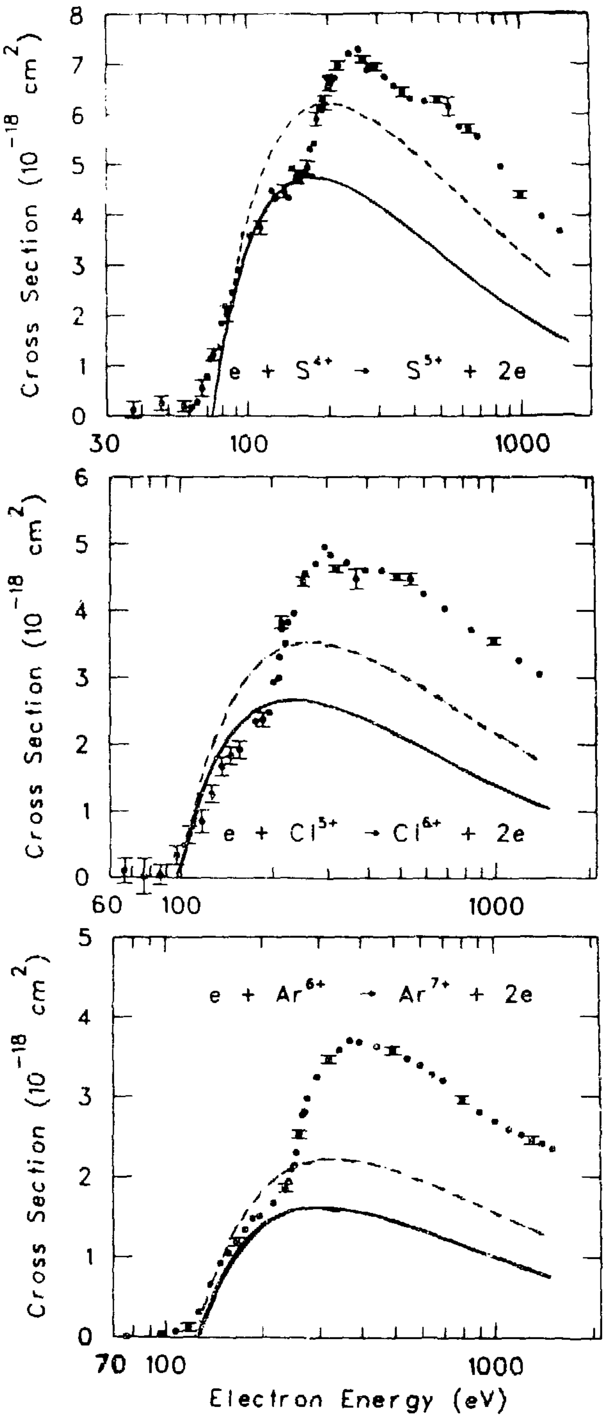

Figure 2. Comparison of experimental cross sections of Howald et al.[6] for electron-impact ionization of Mg-like ions with semiempirical Lotz formula (dashed curves) and scaled theoretical distorted-wave calculations (solid curves) for direct 3s outer-shell ionization. The abrupt changes in slope signify the onset of 2p inner-shell excitation-autoionization.

A closer inspection of the data for this sequence reveals that the cross-section measurements onset at energies which are about 10 eV below the threshold energies for excitation from the $2p^63s^2$ ground state, suggesting the presence of $2p^63s3p$ metastable ions in the experimental reactant beams. The measured cross sections also do not fall off as rapidly as expected after the peak in the cross section, suggesting that another process besides excitation-autoionization is contributing to the ionization cross section at higher energies.

The situation may be more easily understood by referring to Figure 3, which shows the relevant energy levels and transitions for S^{4+}. The only additional process which could be contributing at energies near 200 eV is direct 2p inner-shell ionization, but this should lead via subsequent Auger decay to a net double ionization, and not contribute to the single-ionization measurement. The answer to this puzzle is that while 2p inner-shell ionization from the $2p^63s^2$ ground state will result in autoionization to yield S^{6+}, the same process from the metastable $2p^63s3p$ state of S^{4+} will yield Na-like $2p^53s3p$, many levels of which are known to be metastable against autoionization with microsecond and longer lifetimes[8] — long enough to survive the experiment. These same levels have been identified as having potential importance in X-ray laser development.

Figure 4 shows a comparison between experiment and detailed distorted-wave theoretical calculations by Pindzola and coworkers[7] for ionization of metastable $2p^63s3p$ S^{4+} ions. These calculations include contributions from direct 3s and 3p ionization, 2p-nl excitation-autoionization, and direct 2p ionization. The level of agreement suggests that the metastable levels are playing a significant role in the ionization of Mg-like ions. On the basis of statistical weights alone, one would have expected a significant fraction of Mg-like ions in the experimental reactant beam to be in metastable

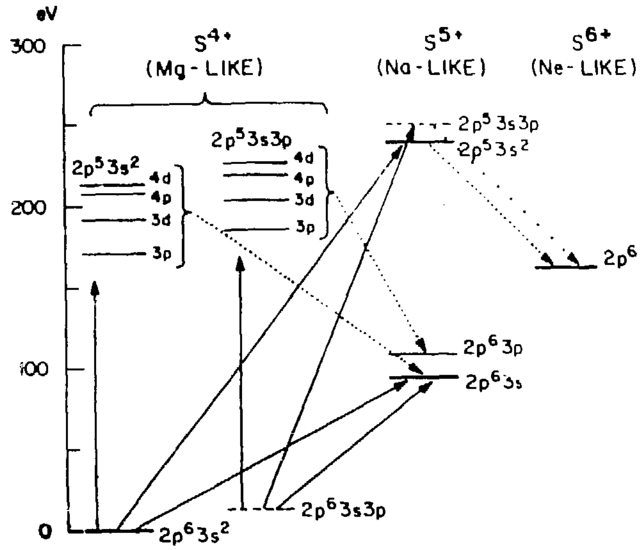

Figure 3. Energy levels and important transitions in the electron-impact ionization of Mg-like S^{4+}, from Ref. 6. Vertical arrows denote excitation processes, inclined solid arrows denote direct ionization, and dotted arrows denote autoionization pathways. Dashed energy levels indicate metastability against radiative or Auger decay.

states. Subsequent studies of electron-impact ionization of multiply charged iron ions suggest that metastable levels play an important role in the ionization balance in the plasmas where they are created.

V. THE IRON ISONUCLEAR SEQUENCE

Atomic processes involving iron and its ions are of considerable current interest because of its importance as an impurity in magnetically-confined fusion plasmas. Recently, a series of experiments on electron-impact ionization of Fe ions was completed using the ORNL-ECR multicharged ion source. This investigation included cross-section measurements for ion charge states 5, 6, 9, 11, 13 and 15. A parallel theoretical study was also carried out, with the goal of establishing the validity of a configuration-average model for these partially-stripped ions with complex

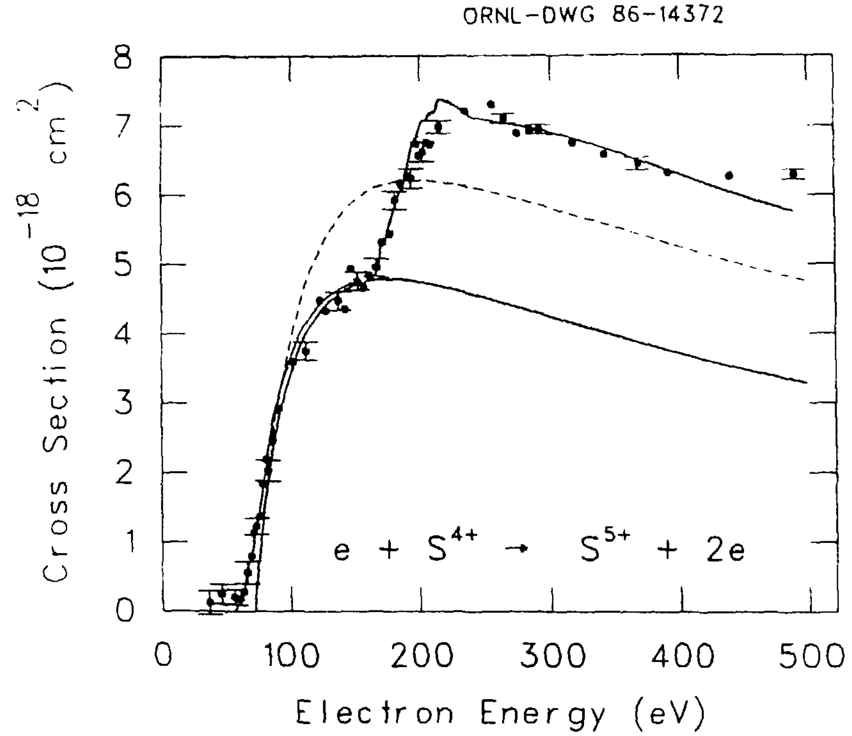

Figure 4. Comparison of theoretical and experimental results for electron-impact ionization of S^{4+}, from Ref. 7. The calculation represents ionization from the $2p^63s3p$ metastable levels. The dashed curve represents direct ionization only, while the solid curve includes contributions due to 2p–nl excitation-autoionization.

electronic structures. A primary objective of these investigations was to quantify the role of excitation-autoionization processes along the iron sequence. Reports have been prepared for publication on the experimental results[2] for charge states 5, 6 and 9 and on the calculations[9] for charges 5, 6, 9, 11 and 13.

Experimental cross-section data and theoretical calculations for electron-impact ionization of Fe^{9+}, Fe^{11+} and Fe^{13+} are compared in Figure 5. The calculations are based on the configuration-average-distorted-wave-statistical model, and include contributions of indirect 2p–nl inner-shell

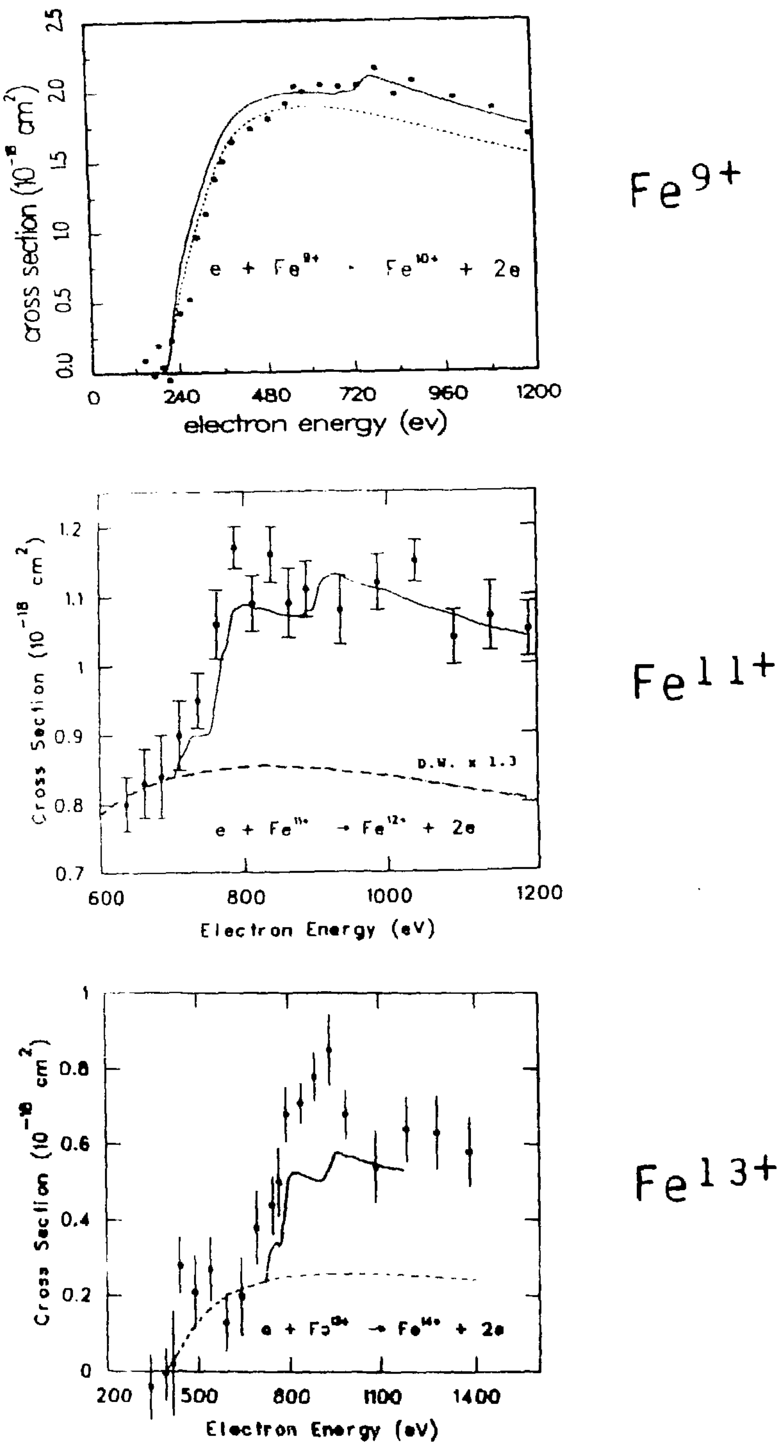

Figure 5. Comparison of experimental[2] and theoretical[9] cross sections for electron-impact ionization of three members of the Fe-isonuclear sequence. Dashed curves represent distorted-wave calculations for direct ionization only, and solid curves represent the total ionization cross section, which includes contributions due to indirect excitation-autoionization calculated in the average-configuration-distorted-wave-statistical model.

excitation-autoionization channels. The determination of these contributions is based on the assumption of a statistical distribution of the excitation collision strength among the various levels, and on the calculated fraction of excited levels which are autoionizing.

The theoretical calculations for these ions are in generally good agreement with experiment, and provide some additional information about the measurements. For Fe^{9+}, the theoretical result shown is for ionization from $3s^2 3p^4 3d$ metastable configuration only, and suggests that predominantly metastable Fe^{9+} ions are produced by the ECR source, as was found to be the case for Mg-like ions. Statistical weights and calculations of decay lifetimes of the levels in the $3s^2 3p^4 3d$ configuration suggest that a significant fraction of the incident ion beam would be expected to be in metastable states. The fractional contribution of excitation-autoionization (2p-nl, 3s-nl) in this case is only 10-15%.

The situations for Fe^{11+} and Fe^{13+} are somewhat different. The comparison between theory and experiment suggests that the Fe^{11+} and Fe^{13+} ion beams are predominantly in their respective ground $3s^2 3p^3$ and $3s^2 3p$ configurations. The fractional contributions of excitation-autoionization (2s-3d, 2p-nl) are also larger than for Fe^{9+}, about 30% for Fe^{11+} and 60% for Fe^{13+}.

Similar comparisons between experiment and theory for the other members of the Fe-isonuclear sequence which have been investigated show that there is no clear pattern in the relative magnitudes of excitation-autoionization contributions along the sequence. The configuration-average-distorted-wave calculations in general give relatively accurate predictions of these indirect-ionization contributions. However, a careful analysis must be made to determine which of the hundreds of individual excited levels are autoionizing, and which are bound. Each ionization stage must be considered individually in order to obtain accurate predictions of ionization cross sections for ions with such complex electronic structures. Experiments currently under way for Ni^{q+} and Cr^{q+} will establish whether the information gained from the Fe-isonuclear sequence may be reliably extended to neighboring ions of different charge which are isoelectronic to the Fe^{q+} cases studied.

VI. RESONANCES IN ELECTRON-IMPACT IONIZATION

Substantial resonance contributions to the cross section for electron-impact ionization were first predicted by LaGattuta and Hahn[1] in 1981. In a series of electron-impact ionization measurements in the Xe-isonuclear sequence[10], some evidence may have been obtained for this so-called resonant-excitation-double-autoionization (REDA) process, although no individual resonances could be resolved. The experimental data were characterized by an apparent smearing of calculated 4d-nl excitation-autoionization contributions[10] to energies some tens of eV below the calculated threshold energies, which was suggestive of the REDA process. Such a broad feature would be expected for these ions because of the number of individual 4d-nl level excitations involved would cause the associated resonances to overlap. The calculations could not however account for the observed magnitudes of this effect, and thus this study remains inconclusive as to whether or not the REDA mechanism is responsible.

In an effort to better characterize the role of the REDA process in ionization, the measurements at ORNL of cross sections for electron-impact ionization along the Fe-isonuclear sequence have very recently been extended to the Na-like Fe^{15+} case.[11] The results of this experiment are compared to the predictions of Lagattuta and Hahn[1] for this ion in Figure 6. The theory includes direct ionization, 2p-nl excitation-autoionization and also the REDA process (indicated by the hatched region). The electron energy resolution of the crossed-beams experiment is less than 2 eV, while the calculated

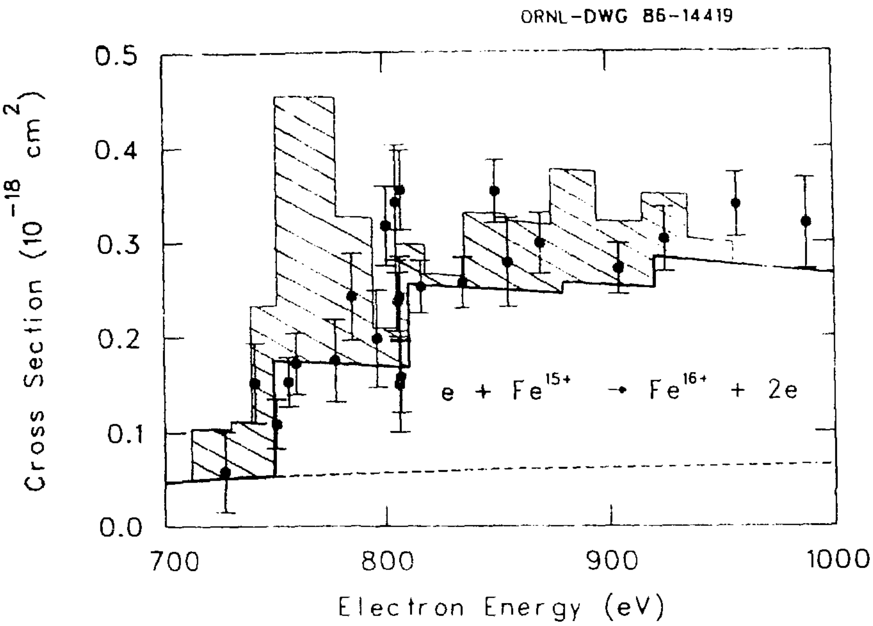

Figure 6. Comparison of predicted[1] and measured[11] cross sections for electron-impact ionization of Na-like Fe^{15+}. The dashed curve is the calculated direct-ionization cross section, to which the calculated contributions of 2p-nl excitation-autoionization and the REDA process have been added. The cross-hatched region indicates the predicted resonance contributions, which have been averaged over 20-eV energy bins. The experimental energy resolution is estimated to be 2 eV.

resonances have been averaged over 20-eV energy bins. Thus the experiment would have been expected to show even larger effects due to resonances than shown in Figure 6. The precision of the measurements was limited by the available ion beam intensity, but they suggest that while the excitation-autoionization contributions have been calculated quite reliably, the REDA contributions are smaller than predicted. The measurements do however strongly suggest a resonance feature near 800 eV electron energy, which is close to the calculated series limit for the $2p^53pnl$ resonances.

It is possible that other resonances may not have been distinguished as clearly in these measurements because of the 2-eV energy resolution in the experiment, and the fact that the resonances are expected to be distributed over a 250-eV energy range. Since each of the experimental points shown represents at least several hours of data-taking time, calculations of the exact positions of the individual resonances would have been extremely helpful in the experimental search for resonance structure in the ionization cross section. It would be instructive to repeat the experiment with such information in hand.

Considerable experimental and theoretical attention has been paid to the accurate determination of 1s-nl excitation-autoionization contributions to electron-impact ionization of Li-like ions[12]. This indirect process is now quite well understood for ions of B through Ne in the Li-isoelectronic sequence. These most recent measurements[12] of the cross section for electron-impact ionization of O^{5+} showed some possible indication of structure in the cross section about 40-50 eV below the 1s-2s excitation threshold. Ionization cross-section measurements for O^{5+} were repeated[13] recently at ORNL with a fine energy grid in the electron-energy interval between 430 and 460 eV. The experimental results are shown in Figure 7, along with the theoretical direct-ionization cross section. Calculated energies[12] for the $1s2s^22p$ and $1s2s2p^2$ resonances are also indicated. The resonance structure is resolved in the experiment, and the correlation with the predicted energies is well within the combined accuracies of the calculations[12] and experiment (approximately 1 eV each).

The observation of these particular resonances in electron-impact ionization is especially significant, since they must involve a simultaneous double Auger process, which Henry and Msezane[14] have termed "auto-double ionization." There are no successive single-step Auger pathways for decay

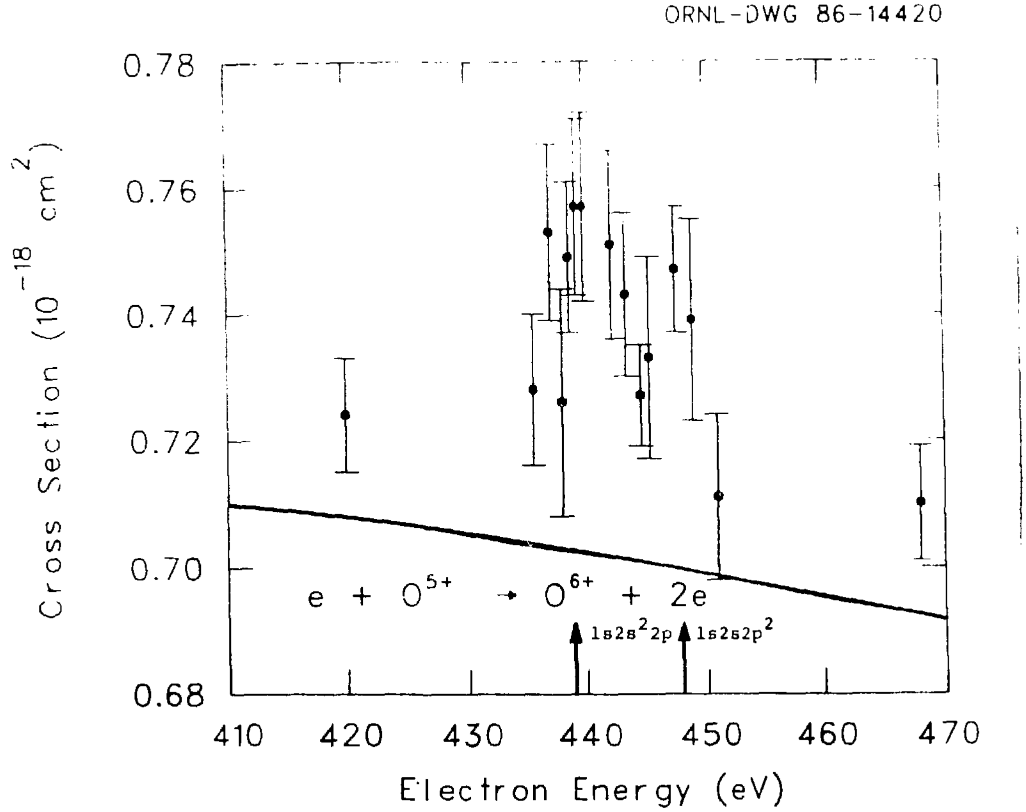

Figure 7. High-resolution measurements by Rinn and coworkers[13] of total cross section for electron-impact ionization of Li-like O^{5+} in the 420-470 eV electron-energy range. The solid curve is a distorted-wave direct ionization cross-section calculation and the vertical arrows at 439 and 448 eV indicate the calculated energies of the $1s2s^22p$ and $1s2s2p^2$ resonances[12]. Error bars represent one standard deviation on counting statistics, and are indicative of relative uncertainties.

of the $1s2s^2$ and $1s2s^22p$ resonances which result in the release of two electrons, and thus would cause a contribution to the measured cross section for single ionization of O^{5+}. Efforts are currently under way to calculate branching ratios for the decay of these doubly excited states[15]. These require the development of new theoretical methods to calculate the autoionization rate and branching ratio for the auto-double channel.

Further experiments are also planned to investigate other members of the Li-sequence in finer detail in the energy regions where these resonances are expected to be.

VII. SUMMARY

We have seen that indirect ionization mechanisms such as inner-shell excitation-autoionization can play an important role in the electron-impact ionization of multiply charged ions. Even for ions with complex electronic structures, this role has been shown to be quite accurately quantified by average-configuration-distorted-wave theoretical methods. Detailed comparisons between theory and experiment have also pointed to the potentially important role of metastable levels for some ionization stages in the ionization balance of high-temperature plasmas.

Definitive experimental evidence has also been presented for the first time for the contribution of individual resonances to the total cross section for electron-impact ionization of Li-like O^{5+}. In addition, experimental evidence has been presented for resonance structure in the cross section for electron-impact ionization of Na-like Fe^{15+}.

ACKNOWLEDGMENTS

The authors are grateful to many coworkers who have contributed significantly to this research, and whose names are included in the references to specific work. A number of the experiments were conducted in continuing collaboration with G.H. Dunn and members of his JILA research group. C. Bottcher, D.C. Griffin and M.S. Pindzola continue to provide the theoretical background and much of the inspiration for this research.

This work was supported by the Office of Fusion Energy of the United States Department of Energy under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

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