PROGRESS IN COLLISIONS OF MULTIPLY CHARGED IONS

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ABSTRACT

The increasing power and availability of supercomputers during the last decade led to significant progress in the theory of multiply charged ion interactions. However, important tests of many theoretical predictions were lacking, and have become possible only quite recently as new capabilities have been realized in the laboratory. This paper broadly surveys some of these experimental developments, and their impact on our understanding of collisional interactions of multiply charged ions. The scope is limited to measurements made with monoenergetic beams.

INTRODUCTION

Collisional interactions of multiply charged ions are in general dominated by enhanced Coulomb or polarization forces, and/or by the appreciable neutralization energy that such ions carry into the interaction. This neutralization energy is equal to the sum of the binding energies of the q electrons that have been removed from an atom to produce an ion of net positive charge q . This may be thought of as a reservoir of stored electronic potential energy for certain classes of interactions. In experimental studies with low-energy ion beams, these q electrons are removed from the atom in an ion source prior to acceleration.

The large Coulomb forces between highly charged ions and other charged particles dominate long-range interactions, modify particle trajectories, and produce large cross sections for some processes. The large neutralization energy of the ion can overshadow kinetic effects in slow collisions. For multiply charged ions, the reduced screening of the nucleus due to missing electrons increases the binding energy of the outer valence electrons, but has a lesser effect on inner-shell electrons. Thus processes involving inner-shell electrons may become relatively more important as the ionic charge increases. For example, in electron-impact ionization of multiply charged ions, inner-shell excitation-autoionization can dominate direct outer-shell ionization by an order of magnitude or more in some cases.

Polarization forces due to ion-induced dipole interactions are also enhanced in multiply charged ion interactions with neutral particles, since the attractive potential is proportional to the square of the ionic charge. This interaction is relatively weak and short-range compared to the Coulomb interaction, but can play a major role in multiply charged ion-neutral collisions at electron-volt energies and below.

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The neutralization of multiply charged ions at solid surfaces has become a topic of intense worldwide activity during the past two years. This process is believed to preferentially populate outer shells of the ion, leading to the production of exotic "hollow" or "superexcited" atoms, whose relaxation has been studied by both photon and electron spectroscopy. This is one example in which theory currently lags experiment, but this advantage may be short-lived as more theorists are attracted to this new inter-disciplinary area.

Finally, the atomic structure of highly charged ions also provides perhaps the most critical testing ground for quantum electrodynamics, since relativistic effects and radiative corrections scale as Z^4 . High-Z ions with simple electronic structures (e.g., H, He, Li or Na-like) provide particularly good cases for evaluating theory.

ADVANCED SOURCES OF MULTIPLY CHARGED IONS FOR RESEARCH

Our ability to study interactions of multiply charged ions in the laboratory is limited to a large extent by our ability to produce them under well-defined conditions. Powerful new sources of multiply charged ions have recently become available for the study of atomic interactions in many laboratories throughout the world, and have had a major impact on research in this area.

In the electron-cyclotron-resonance (ECR) ion source,¹ electrons confined by external magnetic fields are resonantly heated by absorption of microwave power, producing multicharged ions by successive ionization of atoms. This source produces intense, continuous ion beams of high charge which are accelerated to form beams for experiments. Improved performance in terms of higher beam intensity and charge states has recently been demonstrated at higher microwave frequencies and magnetic fields.² The technology of the ECR source is relatively straightforward, and many are currently in use throughout the world for atomic physics research. Without question, the ECR ion source has had the most widespread impact of any experimental device on the study of low energy collisions of multiply charged ions.

In the electron-beam ion source³ (EBIS), multicharged ions are created by and trapped in the space charge well of an intense, high-energy, magnetically confined electron beam. An electrostatic drift-tube structure traps the ions axially, and is pulsed periodically to expel ions for acceleration. This source is capable of producing pulsed ion beams of very high charge for experiments. A novel feature is that the monoenergetic electron beam also permits electron-ion collision studies to be performed within the ion source itself.⁴ However, the technology of the EBIS is demanding, and only a relatively small number are being used for atomic physics research at the present time.

Sophisticated new experiments have been devised to take advantage of the ion beams from ECR and EBIS sources to probe interactions of multicharged ions with electrons and atoms. The techniques employed involve crossed and merged beams of multicharged ions with electrons and neutral atoms, and utilize charge analysis, energy-loss or ejected-electron spectroscopy to deduce quantitative data on collision processes such as ionization, excitation and electron capture.

Collisions of electrons with very highly ionized ions ($q \leq 80$) are now readily accessible to quantitative study using X-ray spectroscopy in an electron-beam ion trap⁵ (EBIT). The EBIT is a compact version of the EBIS which employs an electron beam of very high energy, and is capable of producing and trapping ions of extremely high charge. A planned "super EBIT" will employ electron beams with energies as high as 250 keV. Ion

beams have now also been successfully extracted from an EBIT for further collision studies.⁶

Another significant advance has been the application of ring accelerator technology to the storage of highly charged heavy-ion beams.⁷ This has been made possible by effective ion-beam cooling obtained by merging an intense, monoenergetic electron beam with an ion beam circulating in a synchrotron ring at the same speed. The dramatically enhanced beam luminosity due to ion recirculation (by as much as a factor of 10^6) opens up a whole new spectrum of research possibilities. A number of heavy-ion storage-ring facilities have recently or will soon come on-line in Europe. Their raw power for high-resolution studies of electron-ion recombination has already been demonstrated by using the cooler electron beams part of the time to perform collision studies.⁸ This same electron-cooler beam technology has also been successfully adapted to single-pass merged-beams experiments on conventional ion-beam accelerators in several laboratories.^{9,10} These long-awaited high-resolution experiments have substantiated electron-ion recombination theory for low-Z multicharged ions, and planned measurements on higher-Z ions will provide more critical tests of theory.

The following sections document some experimental results obtained using these powerful new devices, and reported since the last meeting of this series in 1989. It should be borne in mind that there has been vigorous activity in this field, and only a few representative examples have been selected.

PROGRESS IN ELECTRON-IMPACT IONIZATION

An illustrative example to demonstrate the impact of multicharged ion source development on the study of electron-ion collisions is the electron-impact ionization of a simple Li-like (3-electron) ion, O^{5+} . The first crossed-beams measurements on this ion were reported in 1979 by Crandall et al.¹¹ using an O^{5+} ion beam of 10-40 nA produced in a Penning source at ORNL. These were the first cross-section measurements for ionization of a multicharged ion which clearly showed a contribution due to inner-shell excitation-autoionization superimposed on the direct ionization "background." The contribution of this indirect ionization process:

to the total cross section was measured to be $20 \pm 10\%$ near the threshold for the process at 550 eV, with the uncertainty due mainly to counting statistics on the ionization signal. The experiment was repeated by Crandall et al.¹² in 1984 using an improved crossed-beams apparatus and an O^{5+} ion beam of 300 nA from the newly-developed ORNL-ECR ion source operating in a preliminary mode at a microwave frequency of 2.45 GHz. In these new experiments, the $1s-nl$ excitation-autoionization contribution was measured to be $14 \pm 2\%$, providing one of the first definitive confirmations of theory for the inner-shell excitation cross section.

A year later, when the ORNL-ECR source became operational at its design frequency of 10.6 GHz, the O^{5+} ionization measurement was once again repeated by Rinn et al.¹³ using a still more intense beam of O^{5+} ions. In these measurements, the statistical error bars of the measurements were reduced to less than 0.5%, and individual steps due to excitation-autoionization via the $1s2s^2$ and $1s2s2p$ levels were clearly visible for the first time. In this experiment, a search was also made for the predicted

contribution of a more exotic process to the ionization cross section, the so-called resonant excitation auto-double ionization (READI) process.^{14,15} The first step in this ionization mechanism is actually a capture of the incident electron into an autoionizing level, followed by the simultaneous ejection of two Auger electrons, for example:

The most likely decay mechanism for the triply excited O^{4+} intermediate state is single autoionization, which produces only resonances in the elastic scattering cross section, since further autoionization is not possible for this level. This resonant process can only contribute to the ionization cross section via the relatively unlikely auto-double ionization process, for which the branching ratio is difficult to calculate, but is predicted to be extremely small. A detailed search was made in the 430–455 eV energy region, where these resonances are predicted to occur. While the data were suggestive, no clear evidence for the process could be claimed, due in part to the 2 eV energy spread in the electron beam.

The next chapter in the O^{5+} ionization story was written in 1990 in Giessen, Germany, using a newly-developed ECR ion source. The group led by A. Müller applied a rapid energy-scanning technique and a high-intensity space-charge-neutralized electron beam to the problem, and were successful in clearly demonstrating the READI process for the first time, as well as a rich resonance structure in the excitation-autoionization cross section due to the resonant excitation double ionization (REDA) process.¹⁶ The latter process can occur at energies above the 1s-2s excitation-autoionization threshold at 550 eV, where decay of an intermediate triply excited O^{4+} state by two sequential single autoionization processes becomes possible. An example of the REDA process is as follows:

The Giessen measurements for O^{5+} , which have statistical uncertainties as low as 0.1%, are shown in Fig. 1. These data provide a definitive test of the theory for inner-shell excitation, as well as for the role of REDA resonances in a simple ion.

A comparison of similarly precise measurements¹⁶ for ionization of Li-like C^{3+} with a recent close-coupling calculation¹⁷ shows excellent agreement with the observed excitation-autoionization and REDA features in the cross section, providing the most stringent test of excitation theory to date. The measured READI contributions are also consistent with theoretical predictions¹⁵ for the auto-double ionization process, although the latter are less accurate for this process.

Yet another chapter in the continuing O^{5+} ionization story is in progress, as reported by A. Müller at this conference.¹⁰ An improved ECR ion source in Giessen is permitting such resonance structures to be measured in still finer detail! Such detailed studies of the indirect ionization process in simple few-electron ions are important in furthering our basic understanding of electron-ion interactions.

The presence of an inner electronic shell is a prerequisite for indirect ionization to occur, and Li-like ions are the simplest systems that satisfy this criterion, having only two tightly-bound 1s electrons in the inner shell. It is therefore no surprise that indirect contributions to the ionization cross section for Li-like ions are relatively small. However, this is not the case for ions with inner shells containing more, less-tightly-bound inner-shell electrons, where indirect ionization mechanisms may dominate the total ionization

Fig. 1. Measured cross sections for electron-impact ionization of Li-like O^{5+} by Hoffman et al.,¹⁶ showing contributions due to $1s-nl$ excitation-autoionization (EA), beginning at 550 eV in upper figure. Resonance features due to the REDA process are also evident in the 550-650 eV ranges. Lower figure shows resonance features due to READI process in the 430-460 eV range (box in upper figure). Figure is taken from Ref. 16.

cross section.¹⁸ A case in point is ionization of Na-like Fe^{15+} ($2p^63s$) by electron impact. Figure 2 shows a comparison of experimental cross-section measurements of Gregory et al.¹⁹ with theoretical calculations of Chen et al.²⁰ At energies above 800 eV, the contribution due to $2p^53snl$ excitation-autoionization is 4 times the direct ionization, and the REDA contributions ($2p^53snln'l'$) are comparable in magnitude to the direct ionization cross section. This calculation represents by far the most comprehensive treatment of electron-impact ionization to date, accounting for the excitation and decay of more than 10,000 different autoionizing levels, and requiring a tremendous computational effort. Unfortunately, the statistical precision of the experiment is insufficient to map out the resonance features in this case, but further measurements on Fe^{15+} are planned when an upgraded ECR ion source becomes available at ORNL.

PROGRESS IN ELECTRON-IMPACT EXCITATION

Much experimental data has been obtained on inner-shell excitation processes via their signatures in the ionization cross section, but far less experimental data is available for outer-shell excitation processes. All existing absolute excitation cross-section

Fig. 2. Comparison of experimental cross-section measurements of Gregory et al.¹⁹ with theoretical calculations of Chen et al.²⁰ for electron-impact ionization of Na-like Fe^{15+} . The dashed curve shows the direct ionization cross section; the middle solid curve includes the 2p-nl excitation-autoionization contributions, and the upper curve also includes contributions due to the REDA process. Figure is taken from Ref. 20.

measurements for multiply charged ions have been based on the crossed-beams approach and on absolute intensity measurements of the radiation emitted as the excited states decay.¹⁸ Such measurements are severely limited by low photon intensities and detection efficiencies ($\sim 10^{-4}$), difficulties with absolute optical calibration, as well as low electron energy resolution ($\sim 2\text{ eV}$). Cascade contributions from excitation of higher levels may also contribute to the measured photon emission, complicating such measurements.

A different experimental approach²¹ which circumvents these limitations has been developed by the group of G.H. Dunn at JILA for use in conjunction with the ORNL-ECR multicharged ion source. The technique, illustrated in Fig. 3, involves merging fast beams of electrons and multiply charged ions in a uniform axial magnetic field, and detecting electron-impact excitation events via electron energy-loss spectroscopy. Trochoidal ($\mathbf{E} \times \mathbf{B}$) analyzers are used to merge and demerge the beams. The magnetic field ensures complete collection of inelastically scattered electrons, which are directed onto a position-sensitive detector. The first absolute cross-section measurements on a multiply charged ion with this apparatus were reported recently²¹ for 3s-3p excitation of Si^{3+} . This system was chosen because cross-section calculations for Na-like ions are expected to be of high reliability, and thus a comparison of experiment and theory serves to test the validity of both. The observed experimental energy resolution of 0.2 eV is an order of magnitude better than what has previously been achieved for excitation of multiply charged ions. Although the energy-loss method employed here is applicable only at energies near the excitation threshold, this is the energy range where the cross section and resonance effects are usually largest, and theory is most critically tested. This energy range also dominates the excitation rate in most plasma applications.

Another approach to the measurement of cross sections for electron-impact excitation of multiply charged ions has been developed by Huber et al.²² at the LAGRIPPA ECR ion source in Grenoble. Energy- and angle-resolved measurements of elastically and inelastically scattered electrons are performed in a crossed-beams geometry. Absolute differential cross-section measurements have recently been reported for 3s-3p excitation of Na-like Ar^{7+} at scattering angles between 10° and 27° . Such measurements

Fig. 3. JILA merged-beams electron-energy-loss apparatus for cross-section measurements of electron-impact excitation of multiply charged ions near threshold.

are complementary to total excitation cross-section measurements, and provide stringent tests of electron scattering theory.

PROGRESS IN ELECTRON-ION RECOMBINATION

Significant progress has been made during the past two years in high-resolution measurements of cross sections for dielectronic and radiative recombination. The successful application of several different technologies to this problem has provided long-awaited definitive tests (and verifications) of theoretical predictions for these processes for ions with simple electronic structure. Three of these were reported in *Physical Review Letters* during February 1990.

The first in this series was a measurement of the dielectronic recombination cross section for He-like Ar^{16+} by Ali et al.⁴ at Kansas State University, using a newly commissioned EBIS. They measured the ion source yields of Ar^{16+} and Ar^{15+} as a function of electron beam energy in the 2.0-3.5 keV range, and determined the $n=1 \rightarrow n=2$ dielectronic recombination cross section from their ratio, as shown in Fig. 4. Individual $1s2ln'$ ($n \leq 4$) resonances were resolved in their measurements, which were placed on an absolute scale by normalization to theory for electron-impact ionization of Ar^{16+} . Reasonable agreement with theory was obtained when the calculated cross section was convoluted with an experimental electron energy resolution of 61 eV, although the theory appears to overestimate the contributions of the higher- n resonances.

The first atomic physics cross-section measurements in a heavy-ion storage ring were reported in *Physical Review Letters* that same month by Kilgus et al.⁸ They used the merged electron-cooler beam in the Test Storage Ring at Heidelberg in Germany to

Fig. 4. Measured yields of Ar^{16+} and Ar^{15+} , and their ratio, as a function of electron-beam energy in the Kansas State EBIS, from Ref. 4. The ratio is proportional to the dielectronic recombination cross section.

measure absolute cross sections for dielectronic recombination of H-like O^{7+} in the 450-850 eV energy range. The laboratory energy of the O^{7+} beam in the ring was 143 MeV. With a center-of-mass energy resolution of 2 eV, they were able to resolve individual $2l2l'$ O^{6+} resonances as well as $2lnl'$ resonances with $n \geq 3$. The latter account for more than 90% of the total dielectronic recombination cross section. The measurements are in reasonable agreement with theoretical calculations of Griffin and Pindzola,²³ both with respect to the energies of the doubly-excited states, and the relative cross sections. They predict that by using separate cooling and target electron beams, the energy resolution could be improved by more than an order of magnitude. In that same issue, Andersen et al.⁹ reported absolute rate coefficient measurements for radiative recombination of electrons with C^{6+} ions by merging an similarly intense, high-quality electron beam with a 24-MeV C^{6+} beam from a tandem accelerator. The relative energy range of these measurements was 0-1 eV, with an estimated resolution of 0.15 eV. Their data agree well with theoretical predictions for the process.

Measurements of X rays emitted as a function of the electron-beam energy in EBIT have permitted dielectronic recombination cross sections to be determined²⁴ for ions in such extremely high charge states such as Ne-like Xe^{44+} , Au^{69+} and Th^{80+} . The data analysis is complicated somewhat by the superposition of resonances of nearby charge states, but since the Ne-like stages dominate, it is possible to fit such data to predictions of Dirac Fock theory in order to extract individual cross sections, which are placed on an absolute scale by normalization to those for radiative recombination. The electron energy resolution of such measurements is of the order of 50 eV. The EBIT has also been applied to precision X-ray spectroscopy of very highly ionized systems,²⁵ such as Na-like Pt^{67+} , providing sensitive tests of QED theory.

PROGRESS IN ION-ATOM COLLISIONS

With the successful application of optical and translational-energy spectroscopic methods, much progress has been made in the quantitative study of state-selective electron-capture collisions of multiply charged ions during the last several years.²⁶ High-resolution Auger-electron spectroscopy has also been used as a sensitive probe of the double-capture process, which is of intrinsic interest in understanding ion-atom collision mechanisms, but of lesser direct importance in plasma applications.

Fig. 5. Comparison of merged-beams electron-capture cross-section measurements with other measurements and theoretical calculations for $\text{O}^{5+} + \text{H}$ collisions, from Ref. 28.

As noted earlier, the ion-induced-dipole (polarization) interaction between a highly charged ion and a neutral particle can play an important role in collision dynamics at low energies, such as those which prevail in the edge or scrape-off plasma of magnetic fusion devices. A new experimental approach²⁷ has been developed in which intense multicharged ion beams from the ORNL-ECR ion source are merged with a 5-10 keV beam of ground-state H atoms formed by laser photodetachment of a H^- beam. By varying the relative energies of the beams, absolute electron-capture cross sections have been measured at center-of-mass collision energies in the 0.1 - 2000 eV/amu range. Representative data²⁸ are shown in Fig. 5 for $\text{O}^{5+} + \text{H}$ collisions. The rise in the cross section at energies below 2 eV/amu is due in part to the opening of the 4p capture channel, and in part to trajectory effects resulting from the attractive potential in the initial state. For more highly charged ions, such effects are expected to occur at higher energies, and resonances²⁹ may also appear in the total cross section due to population of quasi-bound levels in the attractive potential well in the initial state.

MULTICHARGED ION-SURFACE INTERACTIONS

A remarkable and unexpected result of recent research on the interaction of slow, highly charged ions with solid surfaces has been the extremely rapid neutralization which is observed to occur. Our understanding of such processes follows from the pioneering work of Hagstrum,³⁰ who first proposed the so-called resonance-neutralization model nearly 40 years ago, and from work in the 1970's by Arifov, Parilis and co-workers.³¹ The process is illustrated in Fig. 6. As a highly charged bare (or nearly bare) ion approaches a metal surface, the potential barrier between the ion and the surface is lowered. At some critical distance, electrons flow from the valence band of the metal into empty excited levels of the ion, forming so-called "hollow" or "super-excited" atoms. Of course,

Fig. 6. Formation of a "hollow" or "super-excited" N atom by the resonance neutralization of a N^{6+} ion at a metal surface.

quantum-mechanical tunnelling through the barrier is possible at larger distances. As this "hollow" atom de-excites near the surface by Auger electron emission, electrons continue to flow from the metal to keep the ion neutralized. The emission of projectile K X-rays,³² or of K-Auger electrons^{33,34} as the inner-most vacancy is filled has been used as a "clock" to provide information about the formation and relaxation of such exotic atoms.

Both the K X-ray and K-Auger measurements indicate substantial occupation of the L shell when the 1s vacancy is filled, which cannot be explained by a complex step-wise decay of the hollow atom above the surface, since insufficient time is available for this to occur.³⁴ An explanation has been given recently by Meyer et al.³⁵ based on detailed measurements of ejected projectile K-Auger electrons from grazing interactions of N^{6+} ions with Au and Cu surfaces. Analysis of the measured electron energy spectra at different incidence angles and comparison with Monte-Carlo simulations permitted the identification of spectral features due to both above-surface and subsurface neutralization. The faster subsurface neutralization process dominates, populating the L shell directly, while the slower above-surface mechanism contributes only at grazing ion incidence angles, where sufficient time is available. The manifestation of hollow atoms may be more subtle than was originally thought!

SUMMARY AND OUTLOOK

The last two years have witnessed major advances in our understanding of collisional interactions of multiply charged ions which are important in high-temperature plasmas. This understanding has come from comparison of detailed experimental and theoretical results for simple interacting systems. Such few-electron systems are, for the most part, adequately described by theoretical approximations which have been implemented on existing computational facilities. The next challenge to our understanding will come as this arsenal of new experimental techniques and facilities is applied to progressively more complex interacting systems, where many-body interactions begin to dominate, and a myriad of surprises awaits us. This is sure to place severe demands on both the ingenuity of theorists and on supercomputer development.

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