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EXPERIMENTAL STUDY OF INTERACTIONS OF HIGHLY CHARGED IONS WITH ATOMS AT keV ENERGIES

Progress Report

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Abstract

This Progress Report describes the experimental work carried out, and the work in progress, at the Cornell EBIS Laboratory during the period 7/1/1991 to 6/30/1992. During this period, a number of experiments were carried out.

The absolute values of the total, one, two and three electron transfer cross sections for highly charged argon ions ($8 \leq q \leq 16$) colliding with argon at 2.3 qkeV laboratory energy were measured.

The distribution of recoil ions and molecular fragments formed in highly charged ion atom and molecule collisions was measured in order to help the interpretation of electron spectra in the 40-320 eV energy range emitted in Ar^{q+}+Ar ($8 \leq q \leq 16$) collisions at 2.3 qkeV that were measured in our laboratory. The interpretation of the electron spectra is still under way.

A new collision chamber was built which contains an ion decelerating lens system and a high resolution monochromator-analyzer combination. Ions extracted from the Cornell Electron Beam Ion Source were successfully decelerated from 2.3 qkeV down to 30 qeV. Preliminary 0^{o} translational energy spectra for Ar^{12+} on Ar at a collision energy of 38.6 qeV show a 0.56 qeV resolution.

Work is in progress to extend measurements of cross sections and recoil ion charge state distributions down to collision energies in the 10 eV/amu range.

Work Accomplished

During the past year, three experiments utilizing the Cornell superconducting solenoid cryogenic electron beam ion source (CEBIS) were completed and are being written up. In the first experiment, we measured the total charge changing cross section for Ar^{q+} on Ar ($8 \leq q \leq 16$) at 2.3 qkeV. The method and apparatus were similar to that described in our Ar^{q+} on H_2 and He total cross section experiment. [1,2] The results are shown in figs. 1a, b and c.

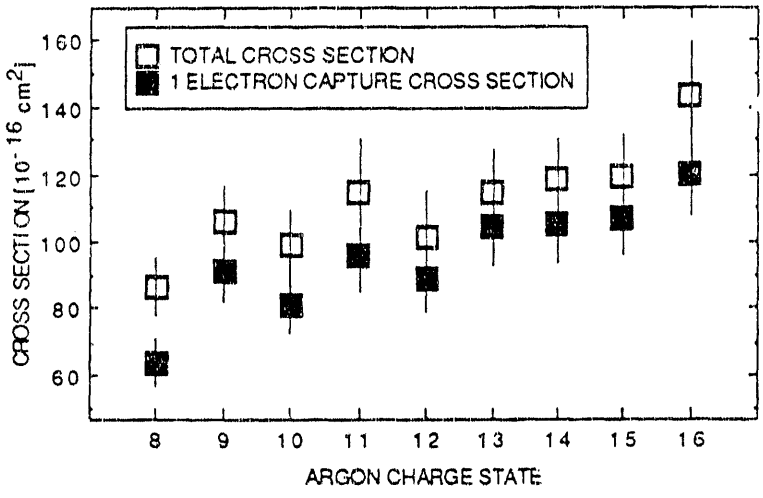

Figure 1a. Total and one electron transfer cross section for Ar^{q+} on Ar at 2.3 qkeV.

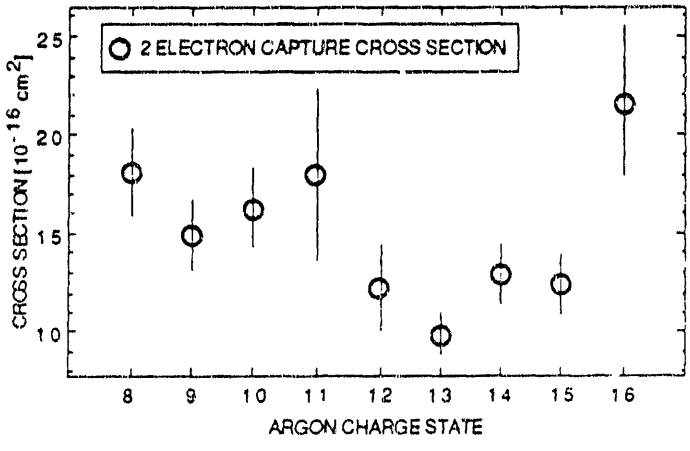

Figure 1b. Two electron transfer cross section for Ar^{q+} on Ar at 2.3 qkeV.

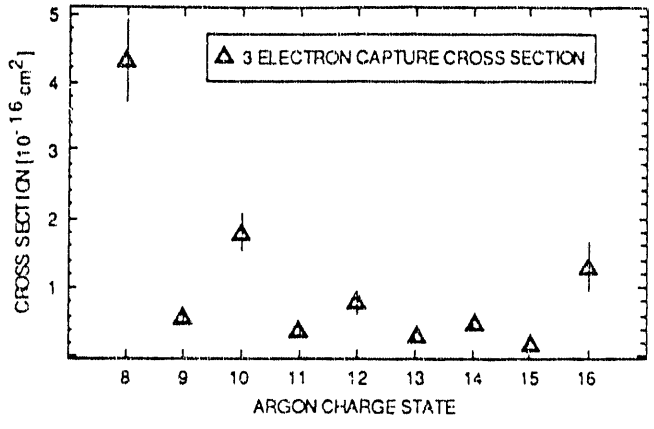

Figure 1c. Three electron transfer cross section for Ar^{q+} on Ar at 2.3 qkeV

The interesting thing to note is that the observed three electron transfer to the projectile is very small, and may even be due to sequential single electron capture.

In the second experiment, we measured the charge state distribution of the recoiling target ions. This distribution was measured using a 2.5 m long time of flight spectrometer with a central guide wire. (The guide wire, slightly biased negatively with respect to the time of flight spectrometer guide tube, guides the ions along the long drift region.) Typical spectra obtained are shown in figure 2a for Ar^{8+} on Ar, and in figure 2b for Ar^{16+} on Ar.

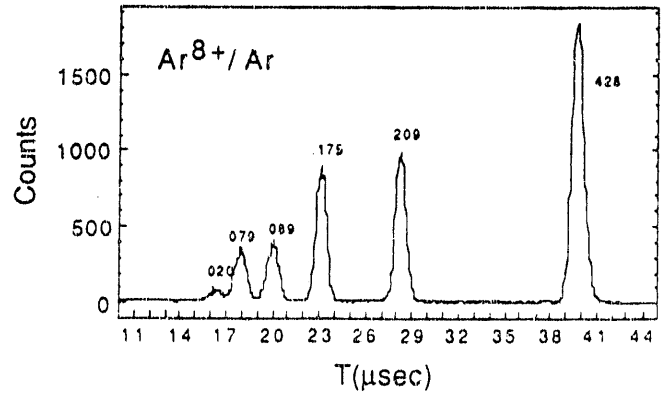

Figure 2a. Recoil ion charge state distribution measured by a time of flight spectrometer. The numbers shown indicate the fraction of a particular charge state present. The lowest charge state, 1+, occurs at the longest time of flight.

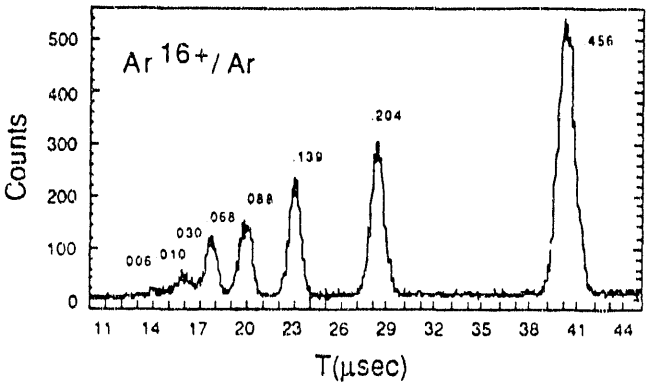

Figure 2b. Same as for Ar^{8+} on Ar in figure 2a. Charge state 7+ is clearly visible.

The main difference between the two spectra is that for Ar^{8+} on Ar, the maximum observed recoil ion charge state is 6+, corresponding to an emptied argon 3p subshell, while for Ar^{16+} there is a hint of charge state 8+, indicating that the 3s subshell in the target is also emptied.

In order to better understand the mechanism of multielectron transfer from many electron targets, we also investigated collisions of highly charged argon ions with other atomic and molecular targets (neon, CO, NO, O_2, N_2, etc.). Figure 3 shows the Ne ion recoil charge state spectrum observed in Ar^{9+} on Ne collisions. The recoil ion charge state distribution is clearly different from that observed in the symmetric Ar case.

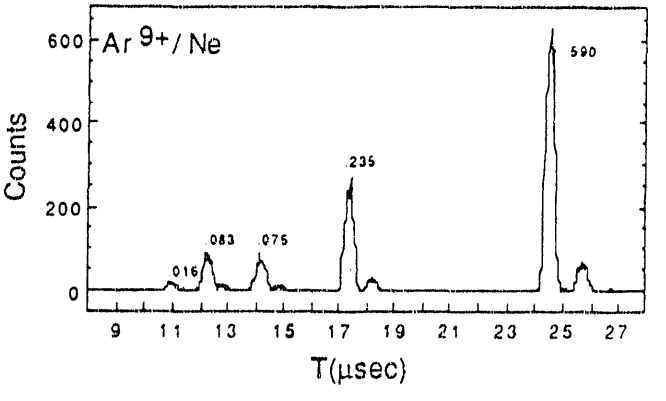

Figure 3. Ne recoil ion charge state distribution in Ar^{q+} on Ne collisions measured by time of flight. Note the heavier isotope ^{22}Ne that appears to the right of the more common ^{20}Ne peaks. The ion charge state 5+ is clearly visible.

Figure 4 shows a typical time of flight spectrum of molecular ions and dissociation fragments in Ar^{11+} on CO collisions. (Small amounts of H_2 were introduced to help calibrate the time of flight spectrometer).

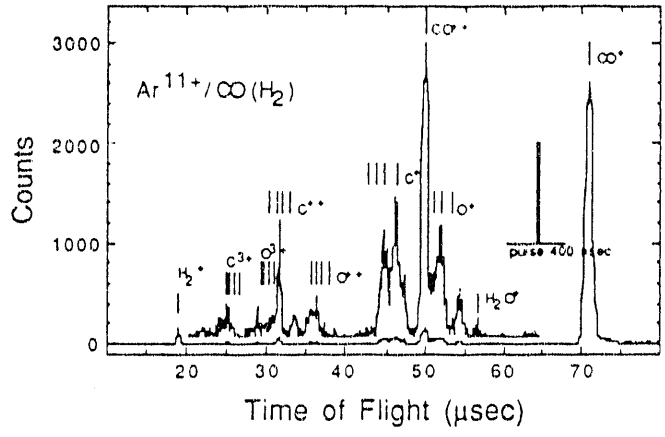

Figure 4. Time of flight spectrum of recoil ions and fragments in Ar^{11+} on CO collisions at 2.3 qkeV.

Similar spectra were observed for different argon charge states incident on CO and N_2. All measured spectra exhibit more or less the same features regardless of the incident argon ion charge state. The spectra are dominated by by the singly charged ion, in this case CO^+. This is in contrast with highly charged ion - atom collisions, in which case the singly charged target ion accounts for only half of all the target ion charge states. The stable/long lived metastable doubly charged molecular ion CO^{++} is the most prominent feature in the spectrum (x10).

The corresponding N_2^{++}, which cannot be distinguished from N^+ is also presumed to be present in the highly charged ion-nitrogen spectra. The remaining peaks are associated with the dissociation of the CO^{q+} molecule. For example, for CO^+, the $C^+ + O$ channel (C^+ with zero initial kinetic energy) is clearly seen in all the spectra. The $C + O^+$ channel on the other hand does not occur with any appreciable intensity. The dissociation of CO^{++} into $C^+ + O^+$ is evident in all spectra, with no hint of the $C^{2+} + O$ or $C + O^{2+}$ channels. The main channel in the dissociation of CO^{3+} is $C^{2+} + O^+$. A smaller peak corresponding to the complementary $C^+ + O^{2+}$ dissociation is also observed. The smaller intensity of the latter could be explained by the possible alignment of the CO^{3+} along the (repelling) electric field between the first two grids of the spectrometer before dissociation. The $C^{3+} + O$, and $C + O^{3+}$ dissociation channels are either absent or negligible. The dominant dissociation channel for CO^{4+} appears to be the symmetric breakup $C^{2+} + O^{2+}$ with all other channels negligible. CO^{5+} dissociates into $C^{3+} + O^{2+}$ and $C^{2+} + O^{3+}$. No other

channels are evident. Finally, CO^{6+} dissociates into $C^{3+} + O^{3+}$.

In the last two experiments described above, the low energy, highly charged Ar^{q+} ions, ($8 \leq q \leq 16$), were produced by the Cornell superconducting solenoid, cryogenic electron beam ion source CEBIS. The ions were extracted at 2.3 qkeV, charge state analyzed by a 90° magnet and crossed with a gas jet target. The latter was located between the first two grids of a 2.5 m long Wiley-McLaren time of flight spectrometer [3] which is at 90° to the incident beam direction. The incident beam was chopped into a 0.4 μs wide pulse, and the recoil target ions and/or fragments analyzed by time of flight.

During the past year, we have also finished the construction of a versatile collision chamber. The collision chamber is a stainless steel vacuum chamber, 24" in diameter and 24" long. It is pumped on by a 170 l/s Balzers turbomolecular pump, a 2400 l/s (air) Varian VHS-6 and a 1200 l/s (air) VHS-4 diffusion pump. The base pressure with all apparatus in place is 3×10^{-8} Torr. It contains ion decelerating optics, four 180° spherical electrostatic analyzers, and a gas target. The chamber is versatile in that all connections, electrical and mechanical, are made inside the vacuum chamber, and hence it is very easy to change from one experiment to another in a matter of hours. A 2m long time of flight spectrometer can also be attached to the collision chamber.

Two of the 180° spherical analyzers are connected in tandem to monochromatize the beam and the other two, also connected in tandem are used to analyze the beam after it has passed through the interaction region. The experimental arrangement is similar to that commonly used in translational spectroscopy experiments.[4] The 180° spherical analyzers have a mean radius of 3.5" and the separation between the spherical surfaces is 0.4 inches. For a 2 mm wide beam, the ΔE/E of the analyzers is 0.011. We have been able to slow down the incident ions from 2.3 qkeV to 30 qeV and record translational energy spectra. An example of a typical 0° translational spectrum obtained is shown in figure 5 for Ar^{12+} on Ar at an incident energy of 38.6 qeV.

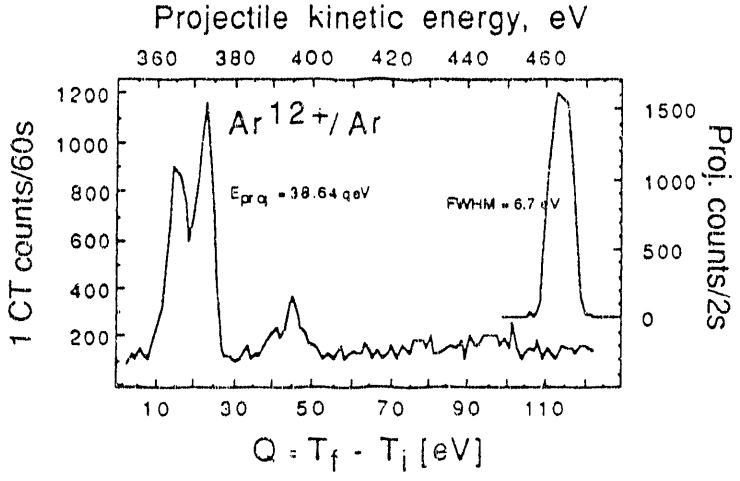

Figure 5. 0^{o} translational energy spectrum for Ar^{12+} on Ar at 464 eV collision energy. The FWHM of the direct beam, (intensity read on the right hand scale) is 6.7 eV, or 0.56 qeV! The one electron capture peaks, left hand scale, correspond to a Q value around 15 and 24 eV respectively, and the so called transfer ionization peaks extend up to 110 eV.

Work in Progress

At present we are still analyzing and trying to understand the Ar^{q+} on Ar collision data, in particular the recoil ion charge state distribution and the observed electron spectra. The correct interpretation of such collisions has to explain several observations. One, the charge state distribution of the target ions, which ranges from one to six, possibly eight for the higher charge state projectiles, two, the fact that the projectile charge state changes mostly by one unit while charge changing collisions involving two units are a factor of 10 smaller, and those involving three units are 100 times smaller, and three, the electron spectra emitted in such collisions.

The large electron transfer crossections, around 100 x 10^{-16} cm^2 imply that the characteristic distance at which the reaction takes place is around 5-6 Å. This means that the two potential wells, of the ion and atom, are sufficiently far apart so that interatomic Auger transitions involving the inner shells are unlikely, and cannot explain multiple ionization of the target.

The transfer of several electrons to some excited state of the collision system is possible. This state can in turn de-excite by emitting Auger electrons. Whether or not this happens during the collision, as the 3s and 3p electrons of argon form a quasimolecule, or whether these electrons are stranded on the projectile, has yet to be determined. In any case, the de-excitation of this excited state of the system gets rid of all but one (in most cases), and sometimes two and even three electrons so that one is left with a multiply ionized target ion, and a projectile whose chare state has changed by one, perhaps by two or even three units of charge.

Whether or not the Auger electrons are emitted during the formation of the quasi molecule, or are stranded on the projectile and then emitted, can perhaps be determined from the electron spectra. Both points of view are consistent with the observation of so called "hollow" atoms.[5] In this case, the capture of many electrons by a slow bare or one-electron nucleus near a metallic surface leads to the formation of multiexcited bound states with many electrons in the outermost shells. The observed K x-ray hypersatellite spectra indicate Lyman-α transitions due to a different number of L and M electrons present. (While x-ray spectra due to "hollow" atoms have not been observed to date because of the tenuous density of gas targets (as far as we know), there is no reason to suspect that such systems are not formed.)

With this as the basic model of the process, it remains to be seen if one can make some sense out of the electron spectra. Before the collision, when the target and projectile are

infinitely far apart, the total electronic energy of the system is given by $E(Ar^{q+}) + E(Ar)$. After the collision, the total electronic energy is $E(Ar^{q'+}) + E(Ar^{s+})$, where q' is the charge on the projectile and s is that on the target. The difference between these two energies is the energy transferred to nuclear motion plus that carried away by electrons and photons. One can place an upper limit on the energy of any Auger electrons emitted as follows. The energy transferred to nuclear coordinates is generally quite small and can be neglected to a first approximation. If we also neglect photon emission at this stage, then the difference in the above mentioned energies is the total energy carried away by the electrons. A more detailed interpretation of the emitted spectra, i.e. Auger electron transition rates and energies, will require use of our computer code TRANSIT [6] to calculate the different Auger transition rates for the many possible excited state configurations.

Regarding the state of the projectile into which the electrons have been captured, we can look at it as follows. The pertinent electrons in the neutral argon target are in the 3s and 3p state. This state remains pretty much unaffected by the incident projectile until the charged ion approaches the atom within a certain distance. At this point, the 3s state goes over into the $4p\sigma$ molecular state which correlates with the 4p state of the united atom, and the 3p splits into $4d\pi$ and $5f\sigma$ states which go over into the 4d and 5f states of the united atom. However, since at the low energies at which these collisions occur, we do not approach the united atom case by any means, it may be more useful to look at the multiparticle energy states in the projectile that are roughly of the same energy as the 3s and 3p states in the target. This means that the states of interest will be linear combinations of the 4s,p, 5s,p, 6s,p 7s,p of the Ar^{q+} ion with different electron populations, and the occupied 3s and 3p states of the argon atom. With the help of the Froese-Fischer general Hartree-Fock program [7], we are analyzing the different possibilities in order to get some understanding of the observed electron and ion recoil ion charge state distribution spectra in low energy, Ar^{q+} on Ar collisions.

We are in the process of measuring total cross sections and target ion charge state distributions at collision energies below 100 qeV for various collision systems. These include Ar^{q+} ($8 \leq q \leq 16$) on H_2, He, Ar, Ne and molecular gases such as N_2, O_2, CO_2, N_2, CO, NO etc.

At the moment we are finishing construction of a set of three stable and programmable power supplies that can float at a potential of several kilovolts. These power supplies will replace the battery supplies we are currently using to control the electrostatic monochromator and analyzer, and allow us to take real data. Once the power supplies are available, we will be able to measure potentials accurately and record 0^o translational energy spectra for a wide range of collision systems at a very high resolution.

Personnel Associated with the Project

The work described in this proposal was carried out by Jan Vancura, postdoctoral associate, myself, Jerry Flidr, graduate research assistant, and James Perotti, research technician.

Publications

Articles published in refereed journals

1. "An Electron Beam Ion Source for Laboratory Experiments", Rev. Sci. Instrum., 63, 3399 (1992). (E.N. Beebe and V.O. Kostroun)

Articles accepted for publication in refereed journals

1. "External Ion Injection into an Electron Beam Ion Source", Nucl. Instr. and Meth. in Phys. Research, (L. Assoufid and V.O. Kostroun).

Articles submitted for publication in refereed journals

1. "Absolute Total and One and Two Electron Transfer Cross Sections for $Ar^{q+}(8:\leq q\leq 16)$ on He and H_2 at 2.3 qkeV", Submitted to Phys. Rev A, (J. Vancura, V. Marchetti, J. Perotti and V.O. Kostroun).

Articles in preparation for submission to refereed journals

1. "Absolute Total and One and Two Electron Transfer Cross Sections for $Ar^{q+}(8:\leq q\leq 16)$ on Ar at 2.3 qkeV", (J. Vancura, V. Marchetti and V.O.Kostroun).

2. "Electron Emissions in $Ar^{q+}(8:\leq q\leq 16)$ on Ar Collisions at 2.3 qkeV", (J. Vancura and V.O. Kostroun).

3. "Ionization and Dissociation of CO and N_2 in Collisions of Low Energy Highly Charged Argon Ions", (J. Vancura and V.O. Kostroun).

4. " Many Body Theory of Atomic Transitions", (C. P. Holmes and V.O. Kostroun).

5. "The Energy Dependence of Transition Rates", (C. P. Holmes and V.O. Kostroun).

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