Charge neutralization of F^- ions in thin rare-gas targets

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The σ_{-0} cross section for F^- ions in rare-gas targets is found to be energy independent for helium and neon between 25 and 120 keV, and to increase with target mass and with beam energy for heavier gas targets. Extrapolation to existing lower-energy data suggests that in the few-keV energy region either double-electron detachment is equally as important as single-electron detachment, or that the single-electron-detachment cross section rises to a maximum and then decreases to our observed values.

I. INTRODUCTION

Several processes may result in the production of a neutral atom from a negative ion during a single collision with an atom. Simple electron detachment can occur in which both atoms are left uncharged and in their ground states

$$A^- + B \rightarrow A + B + e \ .$$

On the other hand, the electron-transfer process which dominates most positive-ion charge-changing cross sections is probably not important even for target atoms which have stable negative ions, and cannot occur otherwise. However a negative ion may be formed by charge transfer to an intermediate state, which then decays by electron emission after the ion and atom have separated. At kinetic energies up to about 1 keV, it has been possible to distinguish between simple detachment and the formation of an intermediate state by using energy-loss spectroscopy on the fast ions and atoms from the collision.[1] A general conclusion from these measurements is that the nonelastic total (angle-integrated) cross section appears to be dominated by the simple detachment process, and this assumption had led to the successful application of the complex potential model to explain quantitatively both the magnitude of the cross section and its increase with energy up to about 100 eV.[2] The complex potential model predicts that the cross sections should peak at an energy of a few keV, and then decrease at higher energies. Measurements up to 3.6 keV for F^- ions have been reported by Hasted[3] on neon krypton and xenon, by Bydin and Dukel'skii[4] on all the rare gases up to 1.8 keV, and by Huq *et al.*[5] up to 250 eV on helium, neon, and argon. There have been no previous measurements in our energy range.

II. EXPERIMENTAL METHOD

A schematic layout of the apparatus is shown in Fig. 1. An rf ion source was used to produce the F^- ions from freon-12 gas. A simple reversal in the polarities of the voltage to the accelerator, of the extraction voltage from the ion source, and of all the magnetic fields, from that when the apparatus was previously used for positive-ion measurements,[6] produced sufficient negative-ion intensity so that no other mechanical changes were neccessary. However the optimum ion source conditions required somewhat higher than normal gas pressures and higher anode voltages, and their adjustment was found to be more critical to obtain the optimum beam than was the case for positive ions.

The F^- ions from the accelerator were selected by a 66-cm radius, 90° deflection, uniform field, double focusing magnet, which had an energy resolution of about 0.1%. An adjustable slit 4 m from the accelerator and two radii upstream from the magnet was used to control the intensity of the ions entering a beam collimator at the conjugate focus downstream from the magnet. This collimator consisted of two apertures, both 0.8 mm in diameter and 20 cm apart. The second of these apertures was also the entrance to the gas target region. The collimator defined the directions of the ions entering the target to $\pm 0.2°$, but this full cone of angles was not filled because of the small angular spread in the ions from the accelerator which were able to pass through the adjustable slit. The target exit aperture was 3.81 cm from the target entrance aperture, and this distance was taken as the effective length of the gas target. The exit aperture was made 1.5 mm in diameter so that all the ions and atoms from detachment collisions which were scattered through

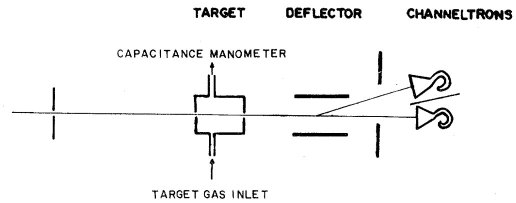

FIG. 1. Schematic layout of target and detectors.

angles up to 2° from the center region of the target could escape and were counted by the detectors. The pressure in the target was measured by a capacitance manometer through a pipe set in the side wall of the target. A second pipe in the side wall, which supplied the gas to the target through a thermal mechanical leak, was offset so that the pressure measurements were not affected by the gas flow.

Beyond the target, the ions were separated from the neutral atoms in a transverse electric field region which was produced by two parallel electrodes 5-cm long and biased symmetrically positive and negative. The ions and atoms were counted digitally by two channeltrons[7] set side by side and in separate grounded enclosures to prevent stray electrons from passing from one channeltron to another. The collimator geometry was such that no direct ions which had passed through the collimator could strike the target exit aperture, but it was found that a severely misaligned beam could produce a background about equal to the dark current in the deflected channeltron when zero deflection voltage was used. This background was probably due to multiple scattering from the edges of the apertures, and was negligible when the beam was optimized through the collimator. Since the geometry of the channeltrons has considerable symmetry, it is likely that there was a similarly small background of particles scattered from the exit aperture into the undeflected channeltron. The channeltrons were operated with their cones at ground potential and a sufficiently large positive potential on the collectors to give pulse saturation. It has been found[8] that when channeltrons are operated with a negative potential on the cone so as to have the collector near ground potential, the loss of electrons to grounded external regions reduces the effective aperture of the cone unless a negatively biased guard ring is used to repel these electrons back into the cone. To investigate any similar loss the cone was biased 200 V positive, and the positive collector voltage increased by the same amount. No significant increase in the effective area of particle detection was found when a small-size ion beam was traversed across a diameter of the cone, and the count rate was constant out to near the geometric edge. The discriminator following the amplifier was set at a level just above the noise so that the smallest pulses from the incident ions or atoms were above the discriminator threshold. Counting rates below 1 kHz were used to avoid gain variations. The detection efficiency for energetic neutral atoms and negative ions has not been measured, but there are indications that when used with pulse saturation the counting efficiency is near to 100%.[9] Measurements with positive ions above a few keV show no evidence for any systematic dependence of the counting efficiency on either the charge state or on the ion energy. Efficiencies in the range 80 to 100% have been measured by Fricke *et al.*[8] We have made the assumption that the detection efficiency is the same for the negative ions as for neutral atoms of the same energy so that the ratio of the counting rates is a true measure of the particle intensity ratio.

The initial growth method was used with a thin target and the cross section was calculated from

$$T\sigma_{-0}=\frac{R}{1+\frac{1}{2}R}\left\{1+\frac{1}{2}\left[(\sigma_{0-}-\sigma_{-+}\left[1+\frac{\sigma_{+0}}{\sigma_{-0}}\right]+\sigma_{0+}\right]T\right\},$$

where R is the ratio of neutral atoms to negative ions emerging from a target of thickness $T=nL$, where n is the atomic density and L is the effective target length. The correction for multiple collisions in the target is valid to first order in the target thickness, neglecting double and higher charge states. Values of σ_{0+} and σ_{0-} are available from Fogel *et al.*[10] up to 60 keV, and their 60-keV values were assumed for the higher energies. No measurements have been reported for σ_{-+}. By restricting the target gas pressures to less than 10^{-3} Torr, the σ_{0+} and σ_{0-} correction terms were always kept below 2%. The σ_{-+} cross section is probably not larger than the σ_{0+} or the σ_{0-} cross section, and σ_{+0} is probably of the same order as σ_{-0} so that it is unlikely that an error greater than 2% was introduced by neglecting the σ_{-+} term in the multiple collision correction. During each measurement, a plot was made of the ratio R as a function of target pressure to detect any serious departure from linearity, and at frequent intervals the deflection voltage was switched off in order to measure the background and so detect beam misalignment. Accurate values for the cross section were then calculated by making a linear least-squares fit to the right side of the equation as a function of the target pressure. The equation assumes that the beam entering the target contained only negative ions, but in the measurements there was a small fraction of neutrals from residual gas collisions in the drift space between the magnet and the target. The much lower background pressure of about 3×10^{-7} Torr being compensated by the much longer path length of 1.5 m. The effect shows up as a nonzero intercept in the linear relation with pressure. A correction was made assuming that the σ_{-0} cross section was the same for the residual gas as for the target gas so that the effective target thickness and therefore the correction terms were slightly increased.

III. RESULTS AND DISCUSSION

The cross sections obtained from the present measurements are shown in Table I and Fig. 2, where they are plotted as a linear function of the ion velocity. The errors shown are those obtained from the least-squares fit. Other errors from the length of the target and the target pressure measurement were smaller. Also shown in Fig. 2 are the previous measurements of Hasted[3] and of Bydin and Dukel'skii.[4] The lower-energy data of Huq *et al.*[5] for helium, neon, and argon gives a good extrapolation to the Bydin and Dukel'skii data, and

TABLE I. Charge neutralization cross sections σ_{-0} for F^- ions in rare-gas targets (10^{-16} cm^2).

Energy (keV)	He	Ne	Target Ar	Kr	Xe
25	3.25	3.06	5.00	5.66	4.87
30		2.73			
35		2.89			
37.5	3.12	2.84	4.92	5.57	5.58
45		2.94			
50	3.30	2.94	5.69	5.57	5.81
55		3.12			5.63
60	3.35	3.18	6.17	5.92	5.67
65		3.20			6.43
70	3.61	3.12	6.63	6.67	7.13
75		3.04			
80		3.03			
85	3.67	2.89	6.88	7.27	7.52
90		3.14			
95	3.39	3.13	6.91	7.89	8.18
100		3.07			
107.5	3.64		7.48		8.07
108		3.28		8.07	
112.5					8.60
115		3.02			
120	3.22	3.10	7.55	8.38	9.14

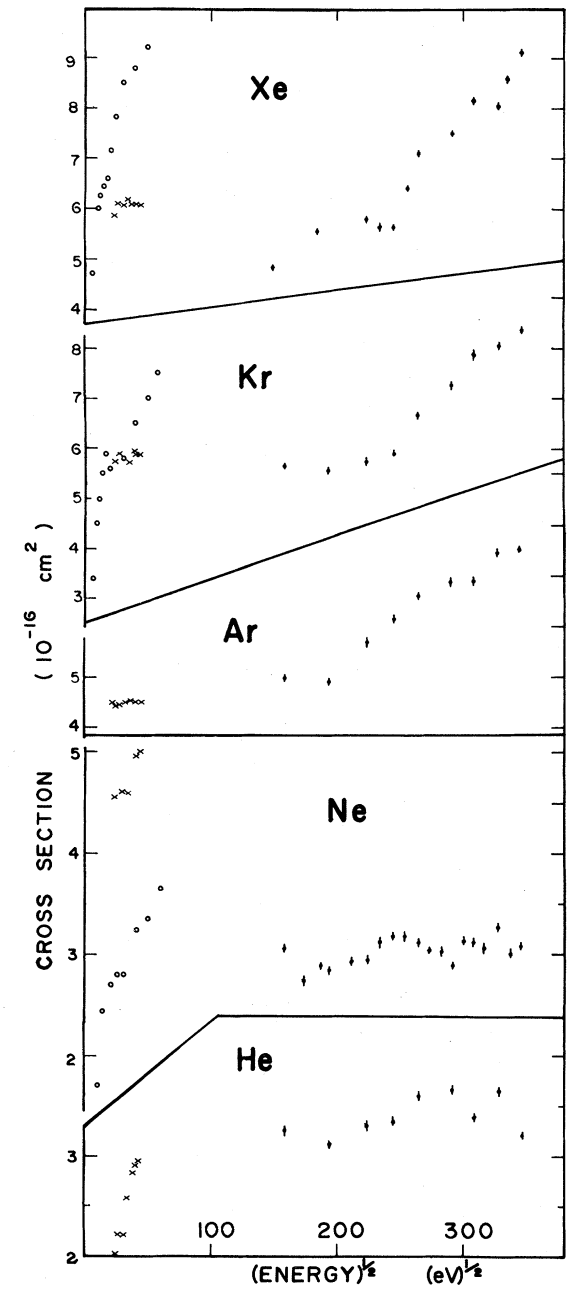

FIG. 2. Cross section for the removal of one electron from a beam of F^- ions by rare-gas atoms. Errors shown represent statistical variation only. Circles are the measurements of Hasted[3] and crosses the measurements of Bydin and Dukel'skii,[4] who both include contributions from double-electron detachment.

since Hasted found some dependence of his results on the source gas which he used to generate the F^- beam, it is likely that the Bydin and Dukel'skii data are the more accurate. For the helium and neon targets the cross sections are roughly energy independent, but for the heavier targets there is a continuous increase. The slight dips near 200 $(\mathrm{eV})^{1/2}$ and near 240 $(\mathrm{eV})^{1/2}$ in xenon may not be significant since they are at the limit of reproducibility of the data.

Even though the data of Bydin and Dukel'skii and of Huq increases to higher values than our 25 keV data, there may be no maximum in the σ_{-0} cross section for the neon target at an energy below the present measurements. All three previous experiments measured the slow electron current produced by the F^- beam in its passage through the target, so that their cross sections were effectively $\sigma_{-0}+2\sigma_{-+}$. At energies below about 100 eV, where good fits have been obtained with the simple detachment model, the contribution from σ_{-+} is probably quite small, but it may be a significant part of the cross section at a few keV. Smirnov[11] has suggested that the σ_{-+} cross section should be similar to the σ_{0+} cross section because of the small magnitude of the electron affinity compared to the ionization potential. In agreement with this, Matić and Čobić[12] found that σ_{-+} and σ_{0+} were roughly equal for C^- and 0^- detachment with the single detachment cross section being slightly smaller. The same comparison for F^- using the data of Fogel *et al.*[10] suggests that at our lowest energies the σ_{-+} cross section could be about 10^{-16} cm^2 for the helium and the neon targets, so that $\sigma_{-0}+2\sigma_{-+}$ could be about 5×10^{-16} cm^2 and provide a good extrapolation to the Bydin and Dukel'skii data without a maximum existing at a few keV. We have made rough measurements of σ_{-+} by reversing the deflection voltages, which confirm that it is about one third of σ_{-0} for neon at 50 keV. Fogel *et al.* found however that σ_{0+} decreased with increasing target mass throughout the rare gases so that it is only about 10^{-17} cm^2 for xenon, and if σ_{-+} is similar, then double detachment would make only a small contribution to the electron capture data, and it is not possible to extrapolate the low energy measurements to our data as was possible for the neon target. The σ_{-0} cross section for xenon must therefore reach a maximum and then decrease significantly between 3.6 and 25 keV.

A common property of the models of electron detachment (Demkov,[13] Lam *et al.*,[4] Smirnov and Firsov[15]) in which an initial molecular state becomes unbound as the two nuclei approach each other is that they predict cross sections which decrease with increasing ion energy once the ion energy becomes large compared to the electron orbit energy. This is because the cross section depends on the time which the unbound molecular state has to decay by electron emission and this time decreases as the ion velocity increases. For example, the detailed fit of Champion and Doverspike[2] for Cl^- detachment by a neon target when continued to higher energies shows a maximum at 300 eV and then a continuous decrease to small values at our energies. It may be that the inflection or slight peak in most of the Hasted or Bydin and Dukel'skii data around 300−400 eV is due to this type of decay process, but clearly other processes dominate.

Herzenberg and Ojha[16] pointed out that when the molecular state becomes unbound, if the electron has a strong *s*-wave component, then there is no angular momentum barrier to delay it escaping to infinity so that in their model the electron emission is not a decay process with a definite lifetime. Their model for H^-+He predicted a probability for free electron creation, which not only increased rapidly as the nuclear separation decreased, but it also increased as the square of the ion velocity. Their model therefore predicted that electron detachment occurs at larger impact parameters as the ion velocity increased, so that in contrast to the usual model of decaying unbound molecular state, the cross section increased continuously with the ion energy up to the breakdown of the adiabatic assumption. For H^- this was at a few keV, but for F^- it probably occurs somewhere in the region of 50 keV.

The cross sections reported here show general target atomic number dependence and energy dependence which is similar to that commonly found in electron transfer collisions such as σ_{0+}. This type of cross section is understood at least qualitatively in terms of an interaction between two discrete states. The Massey maximum in the cross sections, which occurs when the interaction time equals the oscillation period between the two states, therefore depends on the energy defect for the collision. To produce the rise in the cross section throughout the energy region of our measurements so that the Massey maximum is at still higher energies, the energy defect for the intermediate negative-ion state would have to be more than 10 eV.

[1]J. Fayeton, D. Dhuicq, and M. Barat, J. Phys. B 11, 1267 (1978).
[2]R. L. Champion and L. D. Doverspike, Phys. Rev. A 13, 609 (1976).
[3]J. B. Hasted, Proc. R. Soc. London, Ser. A 212, 235 (1952).
[4]Iu. F. Bydin and V. M. Dukel'skii, Zh. Eksp. Teor. Fiz. 31, 569 (1956) [Sov. Phys.—JETP 4, 474 (1957)].
[5]M. S. Huq, L. D. Doverspike, R. L. Champion, and V. A. Esaulov, J. Phys. B 15, 951 (1982).
[6]B. Hird and H. C. Suk, Phys. Rev. A 14, 928 (1976).
[7]Type 4039, Galileo Electo-Optics Corp.; type B419BL01, Phillips Electronics, Ltd.
[8]J. Fricke, A. Müller, and E. Salzborn, Nucl. Instrum. Methods 175, 379 (1980).
[9]M. Vujović, Nucl. Instrum. Methods 76, 171 (1969).
[10]Ia. M. Fogel, V. A. Ankudinov, and D. V. Pilipenko, Zh. Eksp. Teor. Fiz. 38, 26 (1960) [Sov. Phys.—JETP 11, 18 (1960)].
[11]B. M. Smirnov, *Atomnie Stolknovenia i Elementarnie Processi v Plazme* (Atomizdat, Moscow, 1968).
[12]M. Matić and B. Čobić, J. Phys. B 4, 111 (1971).
[13]Yu. N. Demkov, Zh. Eksp. Teor. Fiz. 46, 1126 (1964) [Sov. Phys.—JETP 19, 762 (1964)].
[14]S. K. Lam, J. B. Delos, R. L. Champion, and L. D. Doverspike, Phys. Rev. A 9, 1828 (1974).
[15]B. M. Smirnov and O. B. Firsov, Zh. Eksp. Teor. Fiz. 47, 232 (1964) [Sov. Phys.—JETP 20, 156 (1965)].
[16]A. Herzenberg and P. Ojha, Phys. Rev. A 20, 1905 (1979).