Bromine–rare-gas electron-transfer and electron-loss cross sections at 15–130-keV collision energies

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Measurements are reported of the cross sections for the capture and loss of an electron by 15–130-keV bromine atoms during single collisions with rare-gas atoms. The bromine data fit well with similar data for the other halogens. The electron-capture cross sections peak at collision velocities that agree with the simple two-state model. Competition between target ionization and beam ionization, which is determined by relative ionization potentials, seems to account for the observed differences in the electron-loss cross sections.

I. INTRODUCTION

The usual method of producing a fast atom beam for keV atom-atom collision measurements is to neutralize positive ions. Unfortunately this creates an uncertain proportion of the atoms in metastable states. Measurements with such mixed-state beams are of limited value since the cross section is different for each atomic state. We have previously reported a technique to produce fluorine,[1] chlorine,[2] and iodine[3] atomic beams, which are unlikely to contain any significant proportion of metastable atoms. These neutral beams were used to measure absolute atomic cross sections for charge-changing collisions with all the rare gases.

We report similar measurements with a bromine atomic beam. The cross sections measured were for the collision processes

$$\mathrm{Br}+R\rightarrow \mathrm{Br}^{+}+R+e \quad \text{for } \sigma_{0+} ,$$

$$\mathrm{Br}+R\rightarrow \mathrm{Br}^{-}+R^{+} \quad \text{for } \sigma_{0-}$$

where R stands for each of the rare gases. There are no previously published measurements of these cross sections to our knowledge.

II. NEUTRAL BEAM PRODUCTION

The atomic structure of the halogens is such that quartet states from the ^4P term form the lowest-energy excited states above the $^2P^0$ ground-state doublet. These quartet states, and possibly others at higher energies, are metastable with lifetimes long enough to survive for several meters in a fast atomic beam. If a neutral beam is produced by collisions of accelerated positive ions (in the ^3P state) with a gas, electron capture is allowed to both the quartet and doublet states. The energy defect is more favorable to excited-state rather than to ground-state production, and therefore a neutral beam with an uncertain mixture of doublet- and quartet-state atoms is created.

The method used here was to neutralize accelerated negative ions rather than positive ions. The electron detachment processes during negative-ion–gas collisions favor the ground-state atom production for the following reasons. Simple direct detachment dominates the detachment process at these energies.[4] This removes the valence electron, leaving behind the core state, which for the halogen negative ions is a member of the ground-state doublet. Excitation of autodetaching states also occurs in negative-ion–atom collisions. These appear as Feshbach resonances in the emitted electron spectrum. They have not been identified in bromine, but for chlorine[5] and fluorine[6] the most prominent resonance has a configuration $(^3P)4s^2\,^3P$, which has as its parent the $(^3P)4s\,^4P$ metastable state of the neutral atom. However, the resonance is at a lower energy than the metastable state so that decay to it is energetically impossible and the resonance decays to the ground state. Similarly, higher energy resonances have neutral atom parent states, which in each case are at a higher energy than the resonance. Thus metastable neutral atom production is unlikely in a negative-ion–gas-atom collision and nearly all the atoms should be in the ground-state doublet states. Some excitation to the metastable states will occur through subsequent collisions between the neutralized bromine atom and the neutralizing gas. Multiple collisions should therefore be minimized by using the lowest gas pressure that is sufficient to produce an adequate neutral beam.

III. EXPERIMENTAL METHOD

The same apparatus was used as for the previous measurements,[1–3] where it is described in detail. The bromine negative-ion beam was produced in an rf-type ion source. A suitable partial pressure of bromine was produced in the argon buffer gas by connecting into the gas supply a glass tube containing a drop of bromine that was cooled in liquid nitrogen. Negative ions were extracted from the ion-source canal where they were produced by electron-capture processes from positive ions. The accelerated and mass-analyzed beam was passed through two differentially pumped gas cells in series, which were carefully aligned with each other. In the good vacuum between them a clearing electric field removed all charged particles, leaving only the neutral beam to enter the second cell containing the target gas. Argon at 10^{-3} Torr was used as the neutralizer gas in the first cell. Three side-by-side channel-electron multipliers (CEM)

simultaneously counted the fast positive, neutral, and negative bromine atoms that left the second cell.

The measurement consisted of determining the three CEM counts, over the same time interval, for different pressures of the gas in the second cell. In the initial growth method,[7] assuming single collision conditions, the ratio of the charged ions to neutral atoms at the end of the second gas cell is linearly proportional to the gas pressure there, and the constant of proportionality depends only on the effective length of the target and the cross section.

Corrections were made for the breakdown of this simple linear relation due to double collisions of the particle as it traversed the target.[8] By keeping the target pressure below 8×10^{-4} Torr the corrections never exceeded 3%. This was necessary because the various cross sections in the correction terms are not all known. The σ_{-0} and σ_{0+} values have been measured[9] but σ_{+0} and σ_{+2} had to be estimated from the Xe^+-Ar data of Brackman and Fite.[10] The σ_{2+} cross section and all multiple-electron-transfer cross sections were neglected in the calculation of the correction terms.

Preliminary tests with no gas in either cell established that the same count rate was produced in each CEM when the negative-ion beam was directed to it. As a further check, a few cross-section measurements were repeated with the deflection field reversed, so that the roles of the negative- and positive-ion CEM's were interchanged. The results were identical within statistical error.

The absolute error was estimated at about 5%, or 3×10^{-18} cm^2, whichever was greater. This includes estimates of errors in the measurement of the pressure in the target and its effective length. Sufficient numbers of counts were recorded to keep counting statistics a small part of the total error, except for the smallest cross-section values.

IV. RESULTS AND DISCUSSION

A. Fast negative-ion creation

The σ_{0-} cross sections are listed in Table I. The cross section was too small to measure for the He target over the energy range of our measurements. This was also true for Ne except at 120 keV, the highest energy measured, for which a detectable cross section of $(5\pm3)\times10^{-18}$ cm^2 was found.

The initial and final states are probably well defined in this electron-capture process. If we neglect two electron processes, then the electron which is captured to form the negative ion must be the result of simple target ionization. The very small cross section in He and Ne is probably due to the large ionization potentials of these atoms. In the heavier targets the ionization potentials, though smaller, are still much larger than the electron affinity so that the reaction is always endothermic. Since the electron affinities of all the halogens are quite similar it seems likely that the differences in the cross section should correlate with differences in the ionization potential of the target. To illustrate this the previous measurements

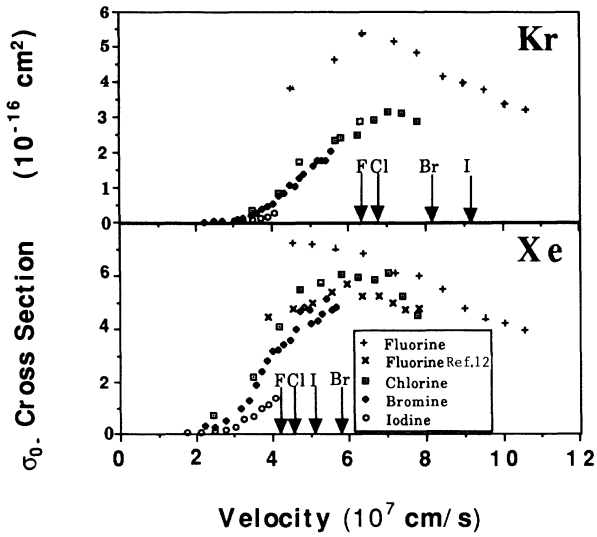

FIG. 1. The electron-transfer cross sections σ_{0-} for bromine-krypton and bromine-xenon collisions plotted as a function of the collision velocity. The arrows show the predicted velocities for the maximum cross section using the two-state model of Hasted and Lee (Ref. 12).

on the xenon and krypton targets, together with the present results, are shown plotted as a function of ion velocity in Fig. 1. All the cross sections seem to lie roughly on the same curve with the exception of our fluorine data which are significantly larger. On the other hand the earlier measurements of Fogel', Ankudinov, and Pilipenko,[11]

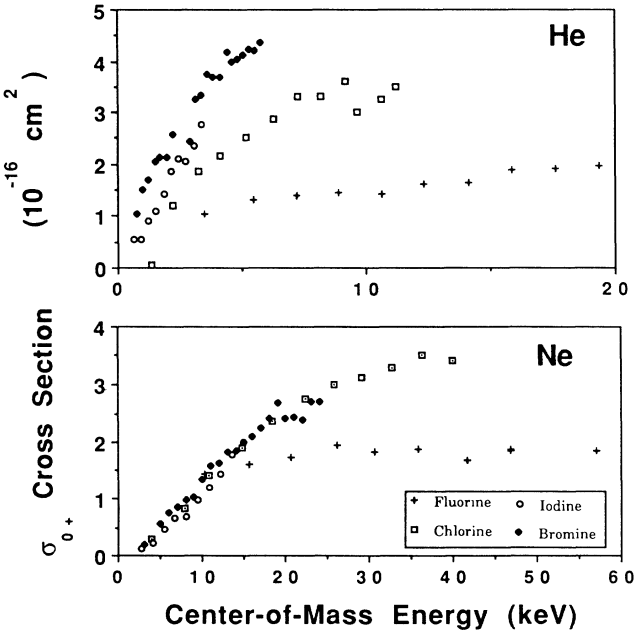

FIG. 2. The electron-loss cross sections σ_{0+} for halogen-helium and halogen-neon collisions plotted as a function of the center-of-mass energy.

though they were obtained using a neutralized F^+ beam and may therefore contain metastable states, provide a good fit.

The simple two-state model should apply to this electron-capture process. The model predicts that the maximum cross section should occur at a collision energy such that the interaction time equals the time for half a quantum-mechanical oscillation between the initial and final states. The collision velocity for the maximum cross section can then be expressed simply by $v_m = \alpha|\Delta E|/h$, where α is a characteristic interaction length and ΔE is the energy defect for the collision process. Hasted and Lee[12] have made a correction to the energy defect which improves the general agreement of this model with experiment. At infinity ΔE is the difference between the ionization potential of the rare gas and the electron affinity of the halogen, but at the internuclear separations at which the electron is transferred there are corrections for the Coulomb and for the polarization energies. With these corrections they found that for most collisions α is about 6.7 Å. A calculation using their relation gives the maximum cross sections as occurring at the velocities shown by the arrows on Fig. 1., in reasonable agreement with the observed maxima in the cross-section data.

TABLE I. Cross sections for the production of fast negative bromine ions σ_{0-}, and for the production of fast positive bromine ions σ_{0+} during a single collision with a rare-gas atom.

Energy	$\sigma_{0-}(10^{-17}$ cm$^2)$				$\sigma_{0+}(10^{-16}$ cm$^2)$			
(keV)	Ar	Kr	Xe	He	Ne	Ar	Kr	Xe
15.7				1.04	0.21	1.43	.081	
20.7		0.17	3.36	1.49		1.63	1.31	0.64
25.7		0.24	2.73	1.70	0.57	1.95	1.44	0.75
30.2	0.12				0.76	2.07		
31.7		0.50	4.83	2.04			1.97	0.67
35.2	0.04			2.14	0.86	2.37	2.56	
36.9			7.28					0.88
40.2					0.97	2.62		
41.3		0.97		2.13			2.37	1.22
45.2	0.18	1.48			1.03	2.94		
46.2			12.7	2.58			2.50	1.10
50.2	0.36	1.90			1.35	3.07	2.43	
51.9			18.8					1.62
55.3	0.48	2.40				3.23	2.47	
56.9			23.8					1.82
60.2	0.78	3.65			1.64	3.40	2.79	
61.0			28.3	2.44				1.84
65.2	0.96	4.26			1.82	3.55	2.96	
66.2			31.7	3.25				
70.2	1.13	5.22			1.84	3.61	2.96	
71.2			32.4	3.33				
75.2	1.81	7.50			2.01	3.75	2.39	
76.2			34.2	3.74				1.99
80.2	2.03	8.42			2.09	3.85	3.44	
81.0			36.1	3.68				2.54
85.2	2.75	10.8			2.23	3.87	3.92	
86.2			39.8	3.69				2.62
90.2	3.11	10.5			2.40	4.01		
92.2			46.8	4.18			3.58	3.05
95.2	4.11	12.5	48.6		2.69	4.22	3.62	3.07
101.0			47.4	4.04				2.93
100.2	4.55	13.9	42.1		2.41	4.27	3.52	
105.2	5.56				2.45	4.16		2.59
110.2	5.73		43.5		2.38	4.61		
111.2		16.3		4.23			3.81	2.61
115.2	5.84				2.72	4.54		
116.2		17.4	45.6	4.22			3.83	2.70
120.2	5.78				2.70	4.67		
121.0		17.5	51.2	4.36			3.70	2.77
125.3	8.65	17.4	47.4			5.09	3.68	2.86
130.3	8.15	20.2	48.3			5.21	4.18	2.75

B. Fast positive-ion creation

The σ_{0+} cross-section data are listed in Table I. A systematic trend of the σ_{0+} cross sections which correlated with the ionization potentials was found in the previously reported measurements. For a fluorine atomic beam the cross section decreased with increasing target mass even though an increase might be expected from the increasing atomic radius.[1] This was interpreted as increasing competition from simple target ionization; a collision process which does not produce fast ions, so that it is not detected by our measurements. Ne and He are the only rare gases with higher ionization potentials than fluorine so that only for these targets can competition from target ionization be neglected. The smaller ionization potentials of Ar and Kr seems to allow some competition from target ionization, and it probably dominates in F-Xe, which has the smallest target ionization potential. Iodine has a smaller ionization potential than any of the rare gases, so there should be much less competition from target ionization. However the Xe cross section[3] was not larger than that for Kr, which suggests that since the iodine ionization potential is only 1.7 eV less than that of Xe, some target ionization may occur for this target.

The present bromine σ_{0+} data show the expected increase between Ne and Kr where target ionization should be negligible. Between Kr and Xe there is a significant drop, which is also expected, since the ionization potential of bromine is only 0.3 eV less than that of Xe, and target ionization should be more significant than with the iodine beam.

Comparing only σ_{0+} cross sections in which target ionization is negligible within the model, the cross sections for the same target show a very similar energy dependence. Figure 2 shows the data for a Ne target and for a He target plotted as a function of the center of mass energy. With the exception of fluorine all the data for each target seem to lie on roughly the same curve.

In a general comparison between the halogen-beam–rare-gas electron transfer and between the loss cross sections, fluorine shows a different behavior from the other halogens for both the σ_{0-} and the σ_{0+} data. This suggests that a different collision mechanism for both electron capture and for ionization may occur for this atom as compared to that of the other halogens.

[1]B. Hird, F. Rahman, and M. W. Orakzai, Can. J. Phys. **66**, 972 (1988).
[2]B. Hird, F. Rahman, and M. W. Orakzai, Phys. Rev. A **37**, 4620 (1988).
[3]B. Hird, M. W. Orakzai, and F. Rahman, Phys. Rev. A **39**, 5010 (1989).
[4]R. L. Champion, Adv. Electron. Electron Phys. **59**, 143 (1982).
[5]D. L. Cunningham and A. K. Edwards, Phys. Rev. A **8**, 2960 (1973).
[6]A. K. Edwards and D. L. Cunningham, Phys. Rev. A **9**, 1011 (1974).
[7]E. Horsdal Pedersen and P. Hvelplund, J. Phys. B London **7**, 132 (1974).
[8]B. Hird and F. Rahman, Phys. Rev. A **30**, 2940 (1984).
[9]B. Hird and I. A. Abbas, Phys. Rev. A **31**, 3974 (1985).
[10]R. T. Brackman and W. L. Fite, United States Weapons Laboratory Technical Report No. AFWL-TR-68-96, Kirtland Air Force Base, NM, 1968.
[11]Ya. M. Fogel', V. A. Ankudinov, and D. V. Pilipenko, Zh. Eksp. Teor. Fiz. **38**, 26 (1960) [Sov. Phys.—JETP **11**, 18 (1960)].
[12]J. B. Hasted and A. R. Lee, Proc. Phys. Soc. London, **79**, 702 (1962).