

in place of g_s/g_I , we finally have

$$g_J/g_s = 1 + 1.2059 \times 10^{-3} - (\nu_s - \sum \nu) / \nu_s.$$

With our preliminary experimental value for

$$(\nu_s - \sum \nu) / \nu_s = (118 \pm 3) \times 10^{-5},$$

we now obtain,

$$g_J/g_s = 1.000026 \pm 0.00003,$$

showing no difference in our limit of accuracy between the g factors of the free electron and the sodium ground state. Further experiments with the aim of improving

the experimental accuracy and extending the method to much lower buffer gas pressures and eventually to near vacuum are in progress.

ACKNOWLEDGMENTS

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Charge Exchange Cross Sections for Helium Ions in Gases*

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The charge exchange cross sections have been determined for a helium ion beam in several stopping gases. The cross sections for electron loss by a fast helium atom (σ_{01}) and for electron capture by an ion (σ_{10}) are reported for energies between 4 and 200 kev. The target gases studied were hydrogen, helium, nitrogen, oxygen, neon, and argon. σ_{01} increases monotonically throughout the energy range for all gases studied, obtaining values of 10^{-6} cm² at 200 kev. In all stopping gases except helium, σ_{10} passes through a maximum of approximately 3×10^{-16} cm² near 50 kev, whereas for helium this cross section decreases throughout the energy range as expected for the resonant exchange reaction. Evidence is presented that the metastable excited state of the helium atom is of importance in the charge exchange process.

INTRODUCTION

PREVIOUS investigations of charge exchange collisions for fast particles in gases which have been reported by this laboratory include determinations of the equilibrium charge distribution of a particle beam after traversing a thick gas target¹ and measurements of the absolute cross sections for electron capture and loss by fast hydrogen atoms and ions.² The target gases were hydrogen, helium, nitrogen, oxygen, neon, and argon, and the energy range was from 4 to 200 kev. The present paper reports measurements of the electron loss cross sections for fast helium atoms passing through the above gases and in the same energy range.

The literature of charge exchange, prior to 1952, has been summarized by Massey and Burhop³ and was also reviewed by Allison and Warshaw⁴ in 1953. As is evident from these reviews, large discrepancies fre-

quently exist between the results of the various investigators. Since publication of these reviews, there have appeared several reports of measurements of the charge exchange cross sections for energetic helium atoms and ions. Stedeford and Hasted,⁵ repeating the work of Keene,⁶ obtained somewhat different results and emphasized the difficulty of this type of measurement. Working at the University of Chicago, Snitzer⁷ has reported measurements of the ratio of the cross sections for electron capture and electron loss in gases and Krasner⁸ determined the cross section for electron loss by fast helium atoms in the energy range from 100 to 450 kev. More recently, Allison *et al.*⁹ have reported measurements of σ_{10} , σ_{12} , σ_{21} , and σ_{20} where the usual notation σ_{if} is used, with i denoting the initial charge state and f the final charge state. Fedorenko¹⁰ has reported values for the cross sections σ_{12} in several gases for energies less than 40 kev. This energy region has also been investigated by de Heer¹¹ who studied

* Work done under the auspices of the U. S. Atomic Energy Commission.

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¹ Stier, Barnett, and Evans, *Phys. Rev.* **96**, 973 (1954); hereafter referred to as I.

² P. M. Stier and C. F. Barnett, *Phys. Rev.* **103**, 896 (1956); hereafter referred to as II.

³ H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Clarendon Press, Oxford, 1952).

⁴ S. K. Allison and S. D. Warshaw, *Revs. Modern Phys.* **25**, 779 (1953).

⁵ J. B. H. Stedeford and J. B. Hasted, *Proc. Roy. Soc. (London)* **A227**, 466 (1955).

⁶ J. P. Keene, *Phil. Mag.* **40**, 369 (1949).

⁷ E. Snitzer, *Phys. Rev.* **89**, 1237 (1953).

⁸ S. Krasner, *Phys. Rev.* **99**, 520 (1955).

⁹ Allison, Cuevas, and Murphy, *Phys. Rev.* **102**, 1041 (1956).

¹⁰ N. V. Fedorenko, *Zhur. Tekh. Fiz.* **24**, 769 (1954).

¹¹ F. J. de Heer, Ph.D. thesis, University of Leiden, Amsterdam, 1956 (unpublished).

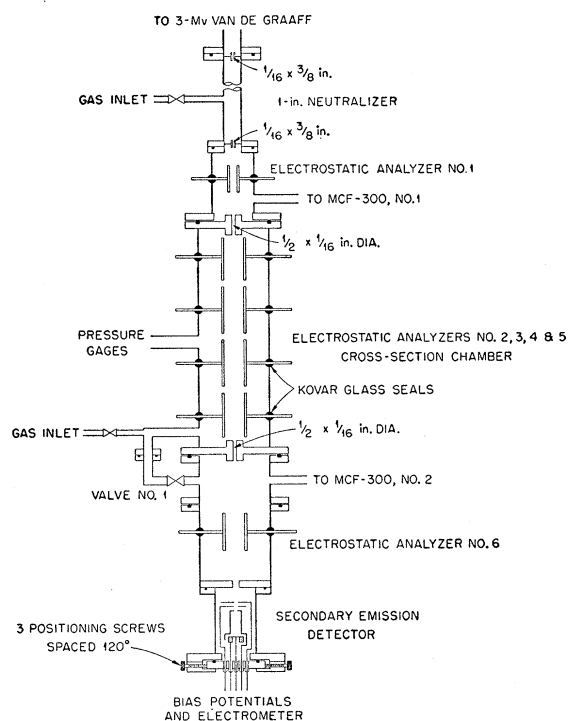


FIG. 1. Apparatus for electron loss cross-section measurements (20–250 kev).

charge transfer cross sections for various heavy ions traversing gases. Dissainaike¹² has measured the equilibrium charge distributions in a helium beam emerging from metal foils for the energy region 130 kev through 1.1 Mev. Jackson,¹³ Schiff,¹⁴ and Moiseiwitsch¹⁵ have treated theoretically the problem of electron capture by the helium ion in helium gas. The results of these calculations and the more recent experimental data will be included in the graphs which display the results of the measurements reported in this paper.

METHOD

In many respects, the equipment and techniques used in the present measurements are the same or equivalent to those previously described (I,II)¹ and only a brief description will be given here. The beam of positive ions was supplied by a Phillips ionization gauge type ion source and was accelerated to the desired energy by a conventional Cockcroft-Walton accelerator. After mass analysis, the beam of positive ions was partially converted to fast helium atoms by passage through a differentially pumped gas cell in

which the target gas pressure was maintained at a few microns of mercury. Positive ions were removed from the emergent beam by passage through an electrostatic field such that only neutral particles were incident on a second gas cell. The electron loss cross sections were computed from the attenuation observed in the transmitted beam as a transverse electric field is applied within this second gas cell. It is apparent that, when ions are removed as formed by the electric field, the transmitted beam will decrease exponentially with the length of the electric field and with the density, i.e., the pressure of the target gas.

A characteristic charge distribution is established in the particle beam after passage through a gas target which is sufficiently thick to insure equilibrium between competing electron capture and loss processes. The existence of equilibrium can be demonstrated by the independence of the transmitted beam on the initial charge state of the particle. If we neglect cross sections involving double electron transfer at equilibrium pressures we may write:

$$\phi_0 = \sigma_{10}\sigma_{21}/D_0, \quad \phi_1 = \sigma_{01}\sigma_{21}/D_0, \quad \phi_2 = \sigma_{01}\sigma_{12}/D_0,$$

where ϕ_0 , ϕ_1 , and ϕ_2 are the fraction of the beam in the charge state 0, 1, and 2, respectively, and

$$D_0 = \sigma_{01}(\sigma_{12} + \sigma_{21}) + \sigma_{10}\sigma_{21}.$$

From this it is apparent that

$$\phi_0/\phi_1 = \sigma_{10}/\sigma_{01}.$$

In this manner, the cross section for electron capture σ_{10} may be computed from the experimentally measured quantities σ_{01} , ϕ_0 , and ϕ_1 .

APPARATUS

A schematic diagram of the equipment employed for the energy range 20 to 200 kev is shown in Fig. 1. For experimental reasons, a different but equivalent apparatus was used for the measurement at energies less than 20 kev. The ion beam from the accelerator was magnetically analyzed yielding a monoenergetic He^+ beam which was incident on the first gas cell designated as the neutralizer. The apertures were chosen to give a pressure differential of approximately 100:1. The first set of electrostatic deflection plates could remove all ions from the emergent beam such that only neutral particles would be incident upon the second gas cell in which the electron loss cross section was measured. Since ions must be deflected through a finite angle to be blocked by the boundaries of the exit aperture, the electric field in the second gas cell was segmented to allow an experimental verification of the computed length of the electric field effective in removing ions from the emergent beam. Effective field lengths determined in this manner agreed well with calculated values and the cross sections have been computed using these effective lengths. The maximum difference

¹² G. A. Dissainaike, *Phil. Mag.* **44**, 1051 (1953).

¹³ J. D. Jackson, *Can. J. Phys.* **32**, 60 (1954).

¹⁴ H. Schiff, *Can. J. Phys.* **32**, 393 (1954).

¹⁵ B. L. Moiseiwitsch, *Proc. Phys. Soc. (London)* **A69**, 653 (1956). See also U. H. Demkov, *Uchenye Zap. Iski Leningrad. Gosudarst. Univ. A. A. Zhdanova Ser. Fiz. Nauk*, **74**, 146 (1952); and O. B. Firsov, *J. Exptl. Theoret. Phys. U.S.S.R.* **21**, 1001 (1951).

between the effective and geometric field lengths was approximately 5% at 200 keV.

All pressures were measured with an accurately calibrated, liquid-nitrogen-trapped McLeod gauge. Several gauges were used during the course of this work and no systematic error due to gauge calibration could be detected. The target gases used in these experiments were passed over a liquid nitrogen cold trap to remove condensable impurities and were normally introduced into the target chamber through an appropriate purifier. Helium was passed through activated charcoal maintained at liquid nitrogen temperature, hydrogen was introduced through a palladium leak, and argon was purified by a bed of calcium and copper shavings maintained at 600°C. The cross sections were not changed by use of these purifiers, indicating the relative purity of the gases, and the lack of sensitivity of the experiment to trace impurities.

The detector used was similar to the differential thermocouple and secondary electron emission instrument described in (I). As previously described, the secondary electron emission for helium atoms is 3% greater than the emission from singly charged ions and the results given are corrected for this difference.

TESTS FOR CONSISTENCY AND REPRODUCIBILITY

In preliminary experiments, it was found that the cross section varied erratically and appeared to be a function of the length of the electrostatic field used in the determination. In order to eliminate possible effects of elastic scattering, the lower aperture of the cross-section chamber was replaced by a movable 0.005 inch slit. This slit was moved linearly across the beam and the emergent flux of particles was integrated. Cross sections determined in this manner were indistinguishable from those obtained with the aperture, indicating that elastic scattering was not important in the measurements of the cross sections. The nonreproducibility of the cross sections was found to be caused by differences in the pressure established in the neutralizer. Fig. 2 shows the dependence of the apparent cross sections on the neutralizer pressure for a He⁰ beam in hydrogen at 100 keV. Since only neutral helium atoms were incident on the charge exchange chamber, it must be concluded that the character of the neutral beam was dependent on the amount of the gas in the neutralizer. The observations indicated that an appreciable number of electron capture collisions in the neutralizer left the fast helium atoms in the metastable state, 1s2s ³S₀. For low neutralizer pressures, when the mean free path was comparable to the dimensions, there was a high probability of the fast helium atoms emerging in this metastable state. As the pressure was increased, the small-angle elastic scattering subsequent to the capture collision de-excited the helium atoms to the ground state, 1s² ¹S₀. This explanation is supported by the additional data displayed in Fig. 2 where the electron loss cross section for fast atoms of nitrogen,

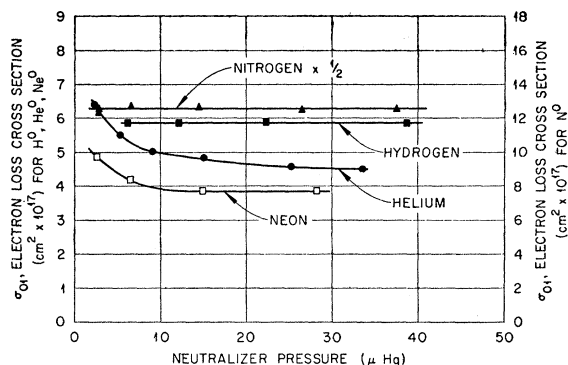


FIG. 2. The measured electron loss cross section as a function of the neutralizer pressure. H⁰, He⁰, Ne⁰, N⁰ in nitrogen gas.

hydrogen, and neon are plotted as a function of neutralizer pressure for a target gas of hydrogen. It is seen that the measured cross sections depend on the neutralizer pressure for Ne and He, but not for H and N. This corresponds to the existence of relatively strong metastable states for He and Ne atoms, but no highly populated metastable state in H and N atoms. At higher pressures, equilibrium must be established between the flux of ions, atoms in the ground state, and atoms in the metastable state. The cross sections reported are for electron loss by this equilibrium distribution of metastable state and ground state atoms.

It is apparent, from Fig. 1, that the apparatus used for measurements of the charge exchange cross section may be used to determine the fraction of the helium beam which is neutral after passage through a thick gas target. Since charge equilibrium could be approached in the charge exchange chamber from an incident neutral beam or an incident ion beam, the existence of equilibrium was readily demonstrated. In this manner, ϕ_0 was measured throughout the energy range from 4 to 200 keV.

ERRORS

The absolute error of the reported cross section at the higher energies is believed to be less than 10%. This estimate represents the sum of the individual errors arising in the measurements of pressure, energy, path length, etc. At the lower energies (less than 10 keV), larger errors are introduced by the uncertainty in the energy of the ion beam from the accelerator due to the energy distribution of the ions from the source. At an energy of 4 keV the particle energy may be less than that recorded by 5%, and since the loss cross section is a steep function of the energy the error in the reported cross section may be somewhat larger than 10%. The relative accuracy and internal consistency of the data obtained in these experiments is best demonstrated by the small scatter of the individual determinations about the smooth curve and by the smooth joining of the results of the separate low- and high-energy experiments.

TABLE 1. Equilibrium fraction of a fast helium beam in the charge states 0, +1, and +2 passing through various gases.

Energy keV	Hydrogen			Helium			Nitrogen			Oxygen			Neon			Argon		
	ϕ_0	ϕ_1	ϕ_2	ϕ_0	ϕ_1	ϕ_2	ϕ_0	ϕ_1	ϕ_2	ϕ_0	ϕ_1	ϕ_2	ϕ_0	ϕ_1	ϕ_2	ϕ_0	ϕ_1	ϕ_2
4							0.980	0.020										
8	0.849	0.151		0.972	0.028		9.963	0.037		0.973	0.027		0.982	0.018		0.995	0.005	
12	0.839	0.161		0.955	0.045		9.946	0.054		0.946	0.054		0.970	0.030		0.984	0.016	
16	0.834	0.166		0.938	0.062		9.928	0.072		0.929	0.071		0.959	0.041		0.976	0.024	
20	0.840	0.166		0.925	0.075		9.906	0.094		0.903	0.097		0.940	0.060		0.972	0.028	
40	0.825	0.175		0.848	0.150	0.0023	0.810	0.190		0.780	0.220		0.835	0.164	0.0012	0.885	0.115	
60	0.805	0.195		0.792	0.204	0.0039	0.725	0.275		0.705	0.293	0.0021	0.745	0.251	0.0014	0.800	0.200	
80	0.760	0.240		0.747	0.248	0.0048	0.650	0.348	0.0023	0.640	0.356	0.0040	0.672	0.320	0.0078	0.720	0.279	0.0012
100	0.710	0.289	0.0009	0.705	0.289	0.0059	0.590	0.406	0.0041	0.585	0.408	0.0069	0.609	0.379	0.012	0.652	0.345	0.0026
120	0.652	0.346	0.0017	0.660	0.333	0.0074	0.530	0.463	0.0072	0.535	0.454	0.011	0.559	0.425	0.016	0.582	0.413	0.0045
140	0.592	0.405	0.0032	0.623	0.368	0.0092	0.480	0.508	0.012	0.492	0.493	0.015	0.515	0.464	0.021	0.520	0.473	0.0073
160	0.535	0.460	0.0053	0.580	0.408	0.012	0.430	0.552	0.018	0.450	0.529	0.021	0.475	0.500	0.025	0.452	0.527	0.011
180	0.485	0.507	0.0081	0.540	0.445	0.015	0.385	0.589	0.026	0.412	0.565	0.032	0.441	0.529	0.030	0.412	0.571	0.017
200	0.445	0.543	0.012	0.580	0.473	0.019	0.340	0.625	0.035	0.378	0.588	0.034	0.414	0.551	0.035	0.372	0.604	0.024

RESULTS AND DISCUSSION

The results of all measurements of the equilibrium fraction of the helium beam in the 0, +1, and +2 charge states are presented in Table I. The present data for ϕ_0 are nearly indistinguishable from the earlier results [Fig. 8 of (I)], and therefore represent an improvement in precision and an extension to lower energies. It should be pointed out that the energy at which $\sigma_{01} = \sigma_{10}$ has been redetermined and is the same as reported in (I) within experimental error.

The values of the loss cross section σ_{01} obtained in the present experiment are plotted in Figs. 3 through 8. In these figures, curves are shown of the calculated values of the electron capture cross section σ_{10} determined from the equilibrium fraction ϕ_0 and the loss cross section. In Fig. 3, the results obtained for a

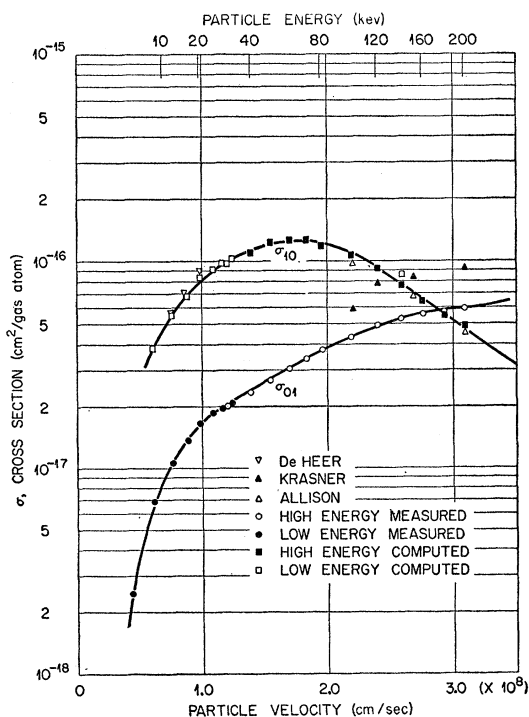


FIG. 3. The charge transfer cross section per atom of gas traversed as a function of particle velocity and energy. Helium atoms and ions in hydrogen gas.

hydrogen gas target are displayed. It is seen that the loss cross section increases monotonically throughout the energy range as is true for all the target gases studied. The results obtained by the group at the University of Chicago⁷⁻⁹ are included in this figure. The data of Allison *et al.*⁹ for σ_{10} are in relatively good agreement with the cross sections reported here although the data of Krasner^{8†} for σ_{01} are approximately 60% larger than the present results. Since the cross sections reported by Krasner could be obtained in this experiment by establishing a relatively low pressure

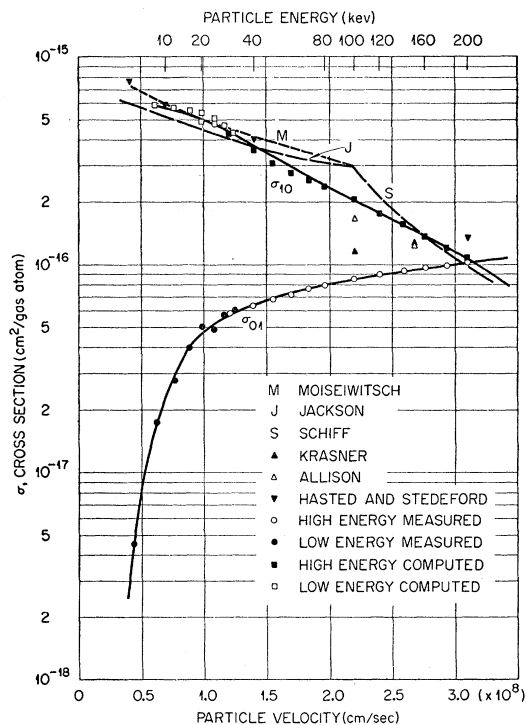


FIG. 4. The charge transfer cross sections per atom of gas traversed as a function of particle velocity and energy. Helium atoms and ions in helium gas.

† Note added in proof.—Prof. S. K. Allison of the University of Chicago has recently repeated the measurements of Krasner. He informs us that his redetermination of the loss cross sections coincides with the measurements made at Oak Ridge National Laboratory.

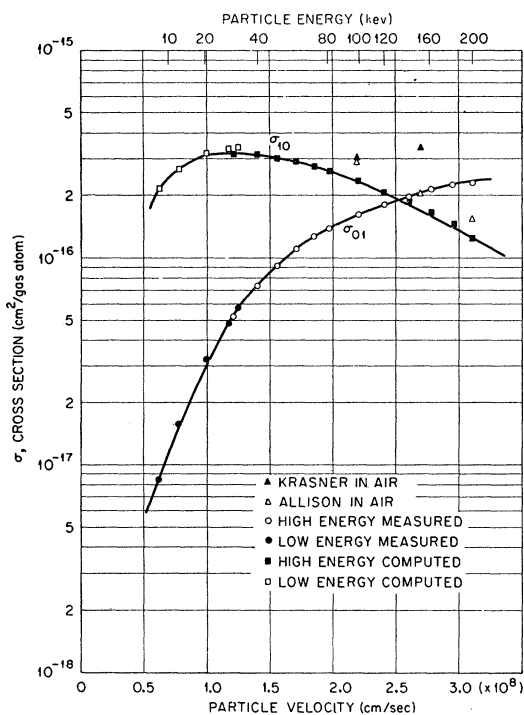


FIG. 5. The charge transfer cross sections per atom of gas traversed as a function of particle velocity or energy. Helium atoms and ions in nitrogen gas.

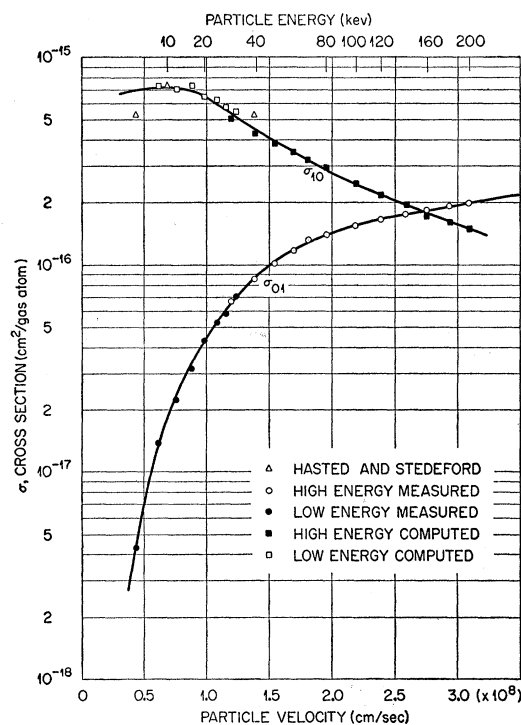


FIG. 7. The charge transfer cross section per atom of gas traversed as a function of particle velocity and energy. Helium atoms and ions in neon gas.

in the neutralizer, it seems possible that the discrepancy arises from the metastable component of the neutral

beam. As mentioned previously, in lieu of being able to perform the experiment with completely de-excited

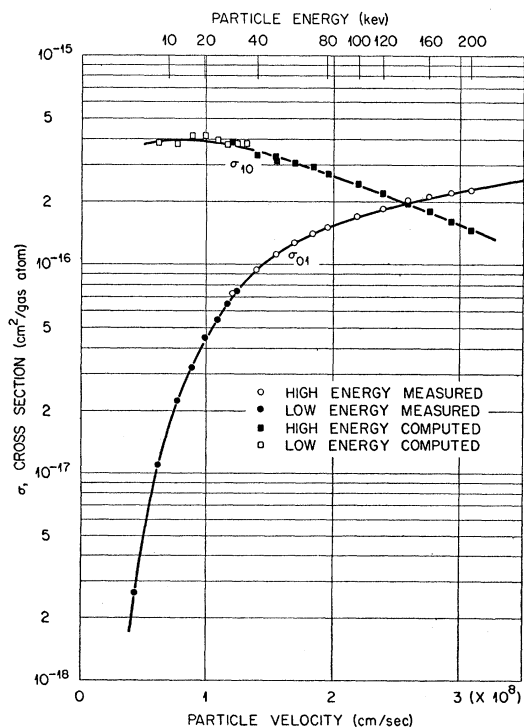


FIG. 6. The charge transfer cross section per atom of gas traversed as a function of particle velocity and energy. Helium atoms and ions in oxygen gas.

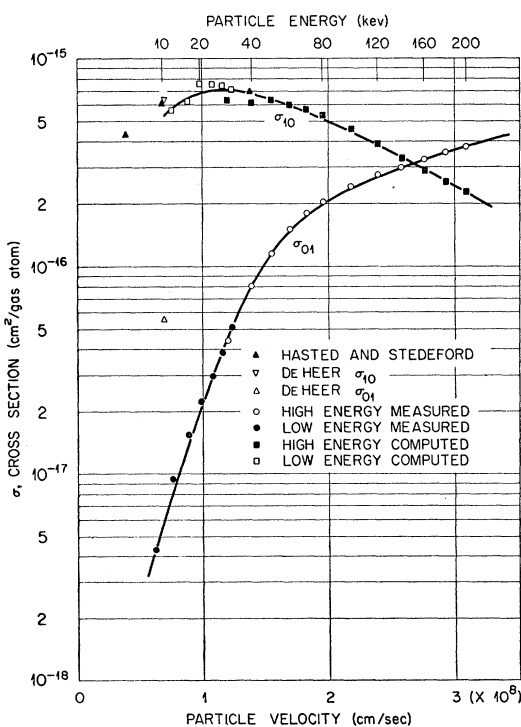


FIG. 8. The charge transfer cross section per atom of gas traversed as a function of particle velocity and energy. Helium atoms and ions in argon gas.

fast helium atoms, it seemed most useful to use an incident beam in which equilibrium existed between the excited and ground state atoms. At the lower energies (10–20 kev) the values of σ_{10} obtained by de Heer¹¹ are seen to be in good agreement with the present results.

In Fig. 4, the corresponding cross sections are plotted for a target gas of helium and again a comparison is made with values obtained by other experimenters. The values of σ_{10} are in substantial agreement at higher energies with those of Allison and at lower energy there is good agreement with the values obtained by Stedeford⁵; however, the discrepancy with Krasner's results for σ_{01} is also present for helium gas. The capture or charge exchange cross section σ_{10} for helium ions in helium gas represents a resonant process in that the change in the total internal energy of the colliding particles during the collision is zero. From classical arguments, it is expected that the cross section will decrease as the relative velocity increases. From an inspection of the various σ_{10} -velocity curves, it is noted that in gases other than helium the cross section passes through a maximum, whereas in helium it decreases throughout the energy range from 4 to 200 kev. The theoretical results of Jackson,¹³ Schiff,¹⁴ and Moiseiwitsch¹⁵ are shown as the Curves *J*, *S*, and *M* respectively. Using an impact-parameter method,

Jackson and Moiseiwitsch have calculated the cross sections for energy less than 100 kev. The primary difference in these calculations is the form of the He_2^+ wave function used. At higher energies, the impact method is not applicable and Schiff has used an extension of the Born approximation. In the energy regions where these separate methods are appropriate, the agreements between the theoretical and experimental results are satisfactory.

The cross sections shown in Figs. 5 and 6 for the target gases nitrogen and oxygen are substantially equal except in the low-energy region where the slope of the σ_{10} curve for nitrogen is much greater than that for oxygen. These results are compared in Fig. 6 with those obtained by the Chicago group using air as the target gas. It is seen that reasonable agreement was obtained with Allison for σ_{10} , but that the σ_{01} curves differ by nearly a factor of two. For the gases neon and argon, the results are plotted in Figs. 7 and 8. No data are available for comparison at high energies, but the values obtained by Stedeford in the energy range 4 to 30 kev are in good agreement with present values.

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Hyperfine Structure Measurements on Plutonium-239*

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The atomic hyperfine structure of plutonium-239 has been investigated by the atomic-beam magnetic resonance method. Research has centered about a relatively highly populated electronic energy level having unit angular momentum which is probably the first excited state of a ground-state 7F term arising from the configuration $(5f)^6(6d)^0(7s)^2$. Atomic and hyperfine structure constants for this level are found to be $I = \frac{1}{2}$, $J = 1$, $g_J = 1.4975 \pm 0.0010$, $\Delta\nu = 7.683 \pm 0.060$ Mc/sec.

From the measured hyperfine structure separation and $5f$ wave functions derived from a Hartree relativistic calculation, the nuclear magnetic moment is inferred to be ± 0.02 nm under the assumption that the $J = 1$ level under investigation arises from a pure 7F term belonging to the configuration $(5f)^6$.

INTRODUCTION

THE heavy elements, those elements toward the end of the periodic table as it is now known, are particularly interesting as a class because of the characteristic nuclear properties exhibited in this region, because of the transition-type electronic systems found here, and because of the opportunities for a more sensitive test of the relativistic theory of the atomic system.

* This work was done under the auspices of the U. S. Atomic Energy Commission.

Plutonium has been investigated previously with respect to nuclear and atomic properties by the methods of optical spectroscopy and paramagnetic resonance. Van den Berg and Klinkenberg¹ first observed the optical spectrum and found the nuclear spin to be $\frac{1}{2}$. Later Conway² observed the furnace spectrum reporting on approximately 500 lines; as yet no term analysis has been made. McNally and Griffin³ have investigated

¹ M. Van den Berg and P. F. A. Klinkenberg, *Physica* **20**, 461 (1954).

² J. G. Conway, *J. Opt. Soc. Am.* **44**, 276 (1954).

³ J. A. McNally and P. M. Griffin in Stable Isotopes Division