Total Ionization in Argon by Heavy Ions of Energies 8 to 100 keV*

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The total ionization produced by ions stopped in argon gas has been measured in the energy range 8–100 keV. The experimental method employed an ion accelerator and a cylindrical ionization chamber with a differentially pumped open window. The following ions were studied: hydrogen, helium, carbon, nitrogen, oxygen, and argon. The average energy loss per ion pair (W value) was found to be dependent on both the velocity and mass of the incident ions at low ion velocities. The W value, in electron volts per ion pair, ranges from 26.8 for 50-keV protons to 90.9 for 25-keV argon ions.

INTRODUCTION

A LARGE part of the existing experimental information on the ionization produced by ions stopped in gases has been obtained by using as incident particles alpha and beta particles from radioactive sources.[1] This information is thus available only for a limited range of ion masses and ion energies, the energies usually being greater than 1 MeV. The work described here provides information on ionization in argon gas for ions with masses in the range 1–40 amu and with energies in the range 8–100 keV. In this energy range the incident ion velocities are comparable to the electron orbital velocities. It is at these velocities that one expects the processes of elastic scattering and electron capture and loss by the ion to become significant compared to the ionization and excitation which predominate at higher velocities. The ionizing efficiency of the ion, as represented by the average energy required to produce an ion pair (W), might therefore be expected to be strongly dependent on the velocity and type of the incident ion, in contrast to the lack of dependence observed in argon for high velocity ions.

EXPERIMENTAL METHOD

The experimental apparatus and technique was essentially the same as that used by Lowry and Miller[2] for measuring the ionization by protons in nitrogen and argon. An ion beam from a low-energy accelerator was analyzed magnetically and electrostatically and allowed to enter a gas-filled cylindrical ionization chamber through a 0.0065-in.-diam differentially pumped aperture. The chamber was operated alternately as: (1) an ionization chamber to measure the ionization current and (2) a proportional counter to measure the incident particle rate. Periodic alternation of these measurements served to reduce uncertainties due to variations present in the incident particle rate. The polarity of the center electrode was negative during ionization chamber operation and was positive during proportional counter operation. The chamber aperture as used by Lowry and Miller was modified to that shown in Fig. 1 in order to decrease the number of ions scattered into the chamber and to reduce the amount of ionization occurring outside the ionization chamber. Ionization loss outside the aperture was investigated by operating the beam stop both negative and positive with respect to the outer electrode of the chamber. Since the measured ionization for these two cases was the same within the experimental uncertainty, it was concluded that aperture loss was not significant. The beam stop was regularly operated negative with respect to the outer electrode.

The collection characteristics of the chamber are indicated by Fig. 2, which shows the relative ionization collected as a function of collecting potential for 50-keV protons, 100-keV Ar^{++} ions, and 25-keV Ar^{+} ions. These curves are representative of those taken for various ions, energies, and pressures. Since the ranges and ionization patterns of the other ions studied lie between those of protons and argon ions,[3] their collection efficiencies should also be intermediate. To further study possible ionization loss due to recombination and losses in the entrance aperture and to the chamber electrodes,

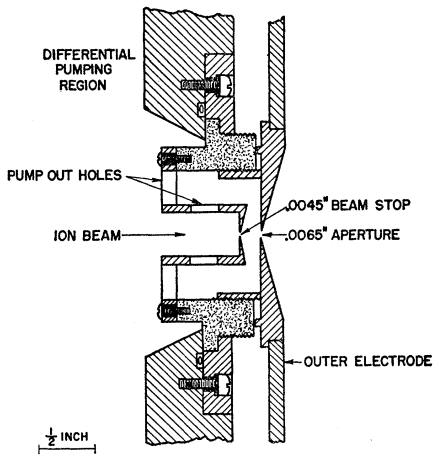

FIG. 1. Aperture assembly.

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[1] W. P. Jesse and J. Sadauskis, Phys. Rev. **90**, 1120 (1953); T. E. Bortner and G. S. Hurst, Phys. Rev. **93**, 1236 (1954); S. C. Curran, J. Angus, and A. L. Cockroft, Phil. Mag. **40**, 36 (1949); W. P. Jesse and J. Sadauskis, Phys. Rev. **107**, 766 (1957).

[2] R. A. Lowry and G. H. Miller, Phys. Rev. **109**, 826 (1958).

[3] G. E. Evans, P. M. Stier, and C. F. Barnett, Phys. Rev. **90**, 825 (1953).

the ionization yield was measured at three chamber pressures for protons and argon ions. From extrapolation of the saturation curves according to the theory of Jaffe[4] and from the measurements at different chamber pressures, it is estimated that the ionization losses for the conditions of the present measurements are less than 2%.

The energy of the ions was determined by using an electrostatic analyzer with cylindrical electrodes. From its geometry the absolute calibration of this instrument was expected to be accurate to 0.3% and was checked to within this accuracy by observing the 163-keV resonance in the $B^{11}(p,\gamma)C^{12}$ reaction. The energy spread of the beam was 0.25%.

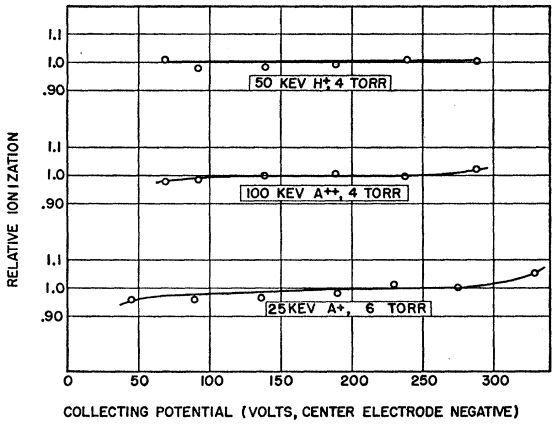

FIG. 2. Saturation curves for H^+, Ar^+, and Ar^{++}.

The chamber was supplied with Matheson argon gas with a stated purity of 99.998%. The gas was admitted into the chamber through a calcium purifier operated at 350°C, with subsequent circulation through the same purifier by convection. Mass spectrometer analysis of gas samples periodically taken from the chamber showed nitrogen to be the only impurity present in an amount greater than the minimum detectable (0.1%). The observed nitrogen contamination was usually less than 0.5%. The method of Bortner and Hurst[5] was used to correct the results for nitrogen contamination to obtain ionization values for pure argon. For these corrections their value of the empirical constant for the nitrogen-argon mixture ($a=0.53$) appropriate to 5-MeV alpha particles was used. The W values for the ions in nitrogen were taken from independent measurements with the present apparatus.

The measurements contain additional corrections for the following: (1) electrometer background current (1–5%); (2) pile-up of the pulses during proportional counter operation (1–5%); and (3) dissociation of the polyatomic ions outside the chamber, with one or more particles failing to enter the chamber (1–2%).

[4] G. Jaffe, Ann. Physik **42**, 303 (1913).
[5] T. E. Bortner and G. S. Hurst, Phys. Rev. **93**, 1236 (1954).

RESULTS

The results of the ionization measurements are presented in Tables I–VI and Fig. 3 for ions of the following six elements: hydrogen, helium, carbon, nitrogen, oxygen, and argon. In Tables I–VI the following information is given for each measurement: (1) the type of ion; (2) the initial energy per incident atomic particle; (3) the ionization yield in ion pairs per atomic particle; (4) the average energy loss in eV/ion pair (W value); and (5) v_0/v, where $v_0=e^2/\hbar$ is

TABLE I. Ionization yield of hydrogen ions in argon.

Initial ions	E keV/atom	I i.p./atom	W eV/i.p.	v_0/v	
H_3^+	8.28	294.8± 3.7	28.1±0.4	1.732	(3% N_2)
H_2^+	12.50	457.6± 4.9	27.3±0.3	1.409	
H_2^+	12.45	456.2± 8.9	27.3±0.6	1.412	
H_2^+	12.61	466.6± 7.4	27.0±0.4	1.403	
H_3^+	16.67	613.2± 5.3	27.2±0.3	1.221	
H_2^+	18.79	676.4± 3.3	27.8±0.2	1.148	
H^+	24.93	912.1± 8.0	27.3±0.3	0.998	(3% N_2)
H^+	24.95	937.2± 7.9	26.6±0.3	0.998	(3% N_2)
H^+	24.96	930.3± 6.2	26.8±0.2	0.998	(1.5% N_2)
H_2^+	25.01	922.0± 7.8	27.1±0.3	0.997	
H^+	37.32	1386.1±13.2	26.9±0.3	0.817	(8% N_2)
H^+	49.98	1859.3± 7.0	26.9±0.2	0.705	
H^+	50.00	1868.0± 9.5	26.8±0.2	0.704	

TABLE II. Ionization yield of helium ions in argon.

Initial ion	E keV/atom	I i.p./atom	W eV/i.p.	v_0/v
He^+	25.19	850.3±15.1	29.6±0.5	1.986
He^+	25.15	819.4±12.1	30.7±0.5	1.987
He^+	37.29	1219.9± 9.2	30.6±0.3	1.632
He^+	50.06	1740.6±19.6	28.8±0.4	1.408

TABLE III. Ionization yield of carbon ions in argon.

Initial ion	E keV/atom	I i.p./atom	W eV/i.p.	v_0/v
C^+	25.16	457.2± 5.3	55.0±0.7	3.441
C^+	37.48	776.1± 4.2	48.3±0.3	2.819
C^+	37.56	789.7±10.4	47.6±0.7	2.817
C^+	49.85	1138.6±11.3	43.8±0.5	2.445
C^+	50.01	1142.6±11.4	43.8±0.5	2.441
C^+	49.98	1099.7± 8.9	45.4±0.4	2.442

TABLE IV. Ionization yield of nitrogen ions in argon.

Initial ion	E keV/atom	I i.p./atom	W eV/i.p.	v_0/v	
N_2^+	12.48	142.0±2.1	87.9±1.3	5.280	
N_2^+	18.76	263.9±3.8	71.1±1.1	4.308	(8% N_2)
N^+	24.97	401.4±5.0	62.2±0.8	3.734	
N_2^+	25.00	401.3±3.4	62.3±0.6	3.371	
N^+	37.36	724.0±8.0	51.6±0.6	3.050	(8% N_2)
N^+	37.44	709.1±7.1	52.8±0.6	3.047	
N^+	50.00	1054.9±6.2	47.4±0.4	2.638	

TABLE V. Ionization yield of oxygen ions in argon.

Initial ion	E keV/atom	I i.p./atom	W eV/i.p.	v_0/v
O^+	25.40	366.1± 4.5	69.4±0.9	3.955
O^+	37.48	638.3±11.6	58.7±1.1	3.256
O^+	49.91	1010.2±10.6	49.4±0.6	2.821

TABLE VI. Ionization yield of argon ions in argon.

Initial ion	E keV/atom	I i.p./atom	W eV/i.p.	v_0/v	
Ar^+	24.90	273.9± 4.8	90.9±1.6	6.320	(3% N_2)
Ar^+	30.97	366.5± 3.3	84.5±0.8	5.670	
Ar^+	37.39	473.9± 6.2	78.9±1.1	5.154	(8% N_2)
Ar^+	37.38	466.0± 2.8	80.2±0.6	5.154	
Ar^+	37.49	466.8± 4.2	80.3±0.8	5.152	
Ar^+	41.95	529.7± 5.6	79.2±0.9	4.866	
Ar^{++}	49.84	654.9± 7.4	76.1±0.9	4.464	(3% N_2)
Ar^+	49.97	645.6± 5.6	77.4±0.7	4.458	
Ar^{++}	61.97	861.6± 6.4	71.9±0.6	4.004	
Ar^{++}	74.71	1078.1±12.0	69.3±0.8	3.646	(8% N_2)
Ar^{++}	74.68	1087.7±10.7	68.7±0.7	3.647	
Ar^{++}	83.87	1246.7±11.3	67.3±0.7	3.441	
Ar^{++}	100.01	1538.6± 8.9	65.0±0.5	3.151	

the velocity of an electron in the first Bohr orbit of hydrogen, and v is the initial velocity of the incident ion. All measurements were made at a chamber pressure of 4 Torr.

In some of the measurements polyatomic and doubly charged ions were used as incident particles. The results of these measurements have been correspondingly identified in Tables I–VI and Fig. 3. A few of the measurements were made at a time when the nitrogen contamination was relatively large. These are denoted in Tables I–VI by the percent nitrogen present during the measurement.

To provide a comparison of the results for the various ions with equal velocities, the values of W tabulated in

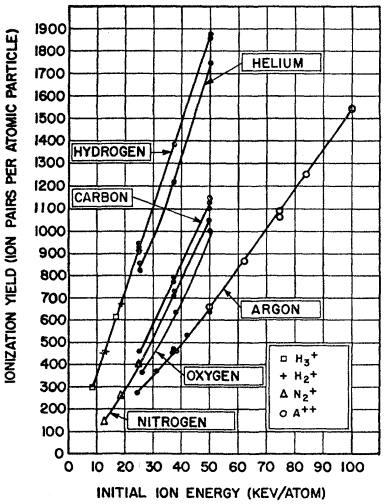

FIG. 3. Ionization yield for various ions.

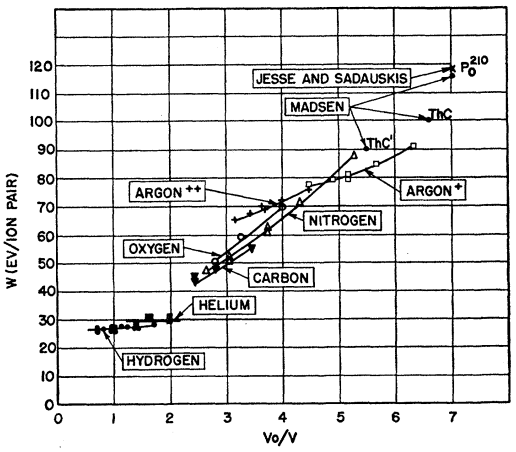

FIG. 4. Comparison of W values of different incident ions at various initial velocities v. $v_0=e^2/\hbar=2.2\times10^8$ cm/sec.

Tables I–VI have been plotted as a function of v_0/v in Fig. 4. The polyatomic ion measurements have not been separately identified in this figure.

The experimental uncertainties in the ionization yields tabulated in Tables I–VI include the following: (1) Statistical uncertainty in the yield as averaged from the individual ion current and counting rate determinations; (2) uncertainty in the electrometer background current; (3) uncertainty in the amount of dissociation of polyatomic ions in the chamber entrance; (4) uncertainty in the correction for nitrogen contamination of the chamber gas; and (5) uncertainty in the electrometer calibration. The maximum uncertainties from these five sources were: (1) ±2%, (2) ±1%, (3) ±0.7%, (4) ±0.7%, and (5) ±0.3%. The tabulated uncertainties for the W values in addition to those listed above for the ionization yield, include an uncertainty in the beam energy of 0.3%. The uncertainties listed do not include any that may be associated with chamber saturation or possible loss of ionization in the entrance aperture or to the electrodes of the chamber. These factors have been discussed in the previous section.

DISCUSSION

The curves of Fig. 4 illustrate the velocity dependence of the W value for the various ions. The values of W, at least for the lighter ions, appear to be approaching at high velocities a common value in the neighborhood of 26.4 eV/i.p., a value generally quoted for high-energy alpha particles in argon.[6] The present results for protons generally agree within the experimental errors with those of Lowry and Miller[2] for 25- to 250-keV protons. The W value of 26.66 eV/i.p. quoted by Larson[7] for 1.826-MeV protons seems quite consistent with the present results.

[6] W. P. Jesse and J. Sadauskis, Phys. Rev. **90**, 1120 (1953); T. E. Bortner and G. S. Hurst, Phys. Rev. **93**, 1236 (1954).
[7] H. V. Larson, Phys. Rev. **112**, 1927 (1958).

The results of Madsen[8] and of Jesse and Sadauskis[9] for ionization in argon by recoils from alpha decay have been included in Fig. 4. The Madsen points have been corrected for the presence of 5% air in the argon gas. The results of the alpha recoil measurements seem to agree with the general trend of the present results.

W values for alpha particles having kinetic energies of several MeV have been observed[10] in argon and found to be independent of the particle energy within 0.5%. It is seen from Fig. 4 that for the heavier ions W is quite dependent on the particle velocity, increasing as the velocity decreases. It is also seen that for a given initial ion velocity, W increases as the mass of the ion increases, with the exception of argon ions at the lower velocities.

[8] B. S. Madsen, Kgl. Danske Videnskab. Selskab. Mat-Fys. Medd. **23**, No. 8 (1945).
[9] W. P. Jesse and J. Sadauskis, Phys. Rev. **102**, 389 (1956).
[10] W. P. Jesse, H. Forstat, and J. Sadauskis, Phys. Rev. **77**, 782 (1950).

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Magnetic Resonance with Large Angular Momentum*

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Multiple quantum transitions present in molecular-beam magnetic resonance of molecules of high rotational angular momentum are treated in terms of a purely classical model. The probability of a change $(\Delta m_J/J)$ in rotational magnetic quantum number per unit angular momentum is shown to be given by $P(\Delta m_J/J)=1/[4\sin(\frac{1}{2}\epsilon)]$ if $|\Delta m_J/J|<2\sin(\frac{1}{2}\epsilon)$ and $P(\Delta m_J/J)=0$ if $|\Delta m_J/J|>2\sin(\frac{1}{2}\epsilon)$, where $P(\Delta m_J/J)\,d(\Delta m_J/J)$ is the fraction of molecules for which this change lies between $\Delta m_J/J$ and $\Delta m_J/J+d(\Delta m_J/J)$, and ϵ is the angle which the angular momentum makes with the magnetic field after the transition for a molecule whose angular momentum is initially along the field. By way of comparison, the transition probability of a spin-$\frac{1}{2}$ particle under the same conditions is $\sin^2(\frac{1}{2}\epsilon)$. This probability is weighted according to the probability of detecting a transition with a change in magnetic moment of $\mu_J\Delta m_J$ and averaged over the thermal distributions of J and V to give a theoretical line shape. The theory is applied to rotational magnetic moments of molecules. The calculated line shape is shown to agree reasonably well with an experimental curve of the rotational magnetic-moment resonance of the molecule OCS.

I. INTRODUCTION

RECENT work by Lawrence,[1] Anderson,[2] and Pinkerton[3] has indicated that in molecular-beam magnetic resonance of magnetic moments associated with large angular momenta an important role is played by multiple quantum transitions. The purpose of this paper is to present a completely classical treatment of these transitions, as should be valid in the limit of large angular momenta. The system considered consists of a molecule with a magnetic moment proportional to the angular momentum J, so that the gyromagnetic ratio g_J is independent both of J and its projection m_J along the field axis. This is very nearly true for all rotational moments. The molecule passes through a standard flop-out molecular-beam apparatus,[4] with an oscillating field perpendicular to a homogeneous magnetic field to produce a transition. A complete calculation has been made of the theoretical line shape, which agrees reasonably well with the experimental line shape of the rotational moment resonance of the molecule OCS. The values of the rotational moments measured in several molecules and the experimental techniques of measurement are discussed in another paper.[5]

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[1] T. R. Lawrence, C. H. Anderson, and N. F. Ramsey, Phys. Rev. **130**, 1864 (1963).
[2] C. H. Anderson, Ph.D. thesis, Harvard University (1961) (unpublished).
[3] J. N. Pinkerton, Ph.D. thesis, Harvard University (1961) (unpublished).

II. SINGLE COIL DERIVATION

The system consists of a molecule with a magnetic moment $\mathbf{u}_J$ proportional to its angular momentum $\mathbf{J}\hbar$. The molecule passes through a rotating field inside of a homogeneous field region. The rotating field is given by

$$\mathbf{H}_1=H_1\cos\omega t\,\mathbf{i}-H_1\sin\omega t\,\mathbf{j},$$

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