Collisional Breakup of High-Energy H_2^+ Ions*

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The gas collisional cross sections for breakup of 20.9-MeV H_2^+ ions into H^0+H^+ and H^++H^++e have been measured in H_2, He, N_2, and Ar. Total dissociation cross sections in the same gases and in the energy range 10 to 65 MeV have been deduced from radial-attenuation measurements on the internal beam of the Berkeley 88-in. cyclotron. These cross sections have also been calculated for each of the H_2^+ molecular-ion vibrational levels and for an average over the vibrational levels, with an approach proposed by Salpeter. The measured and calculated average cross sections are in good agreement.

I. INTRODUCTION

THE gas collisional cross sections for breakup of H_2^+ ions have been studied fairly extensively in the past decade, particularly below about 250 keV. In attempts to explain the rather large differences among the earlier measurements, experimental attention has been focused on the effects of possibly differing, vibrational-population distributions. It is now generally agreed that measured dissociation cross sections are essentially independent of the type of ion source from which the H_2^+ beams are obtained. On the other hand, variations attributable to special vibrational distributions have been observed with H_2^+ beams produced by gas collisional breakup of H_3^+ ions[1,2] (up to 40% greater than from an ion source), or by ionization of H_2 gas with electrons having energies within a few electron volts of the H_2 ionization threshold[3,4] (cross-section variations of about a factor of 4).

No theoretical estimates of dissociation cross sections for energies below approximately 1 MeV have been published. In the high-energy range, Salpeter's approximate calculations, made a number of years ago for the H_2^+ ground-vibrational-state ion,[5] gave quite good agreement with total cross-section values obtained by Effat.[6] (Effat measured the radial attenuation of a cyclotron's internal beam in the energy range from 9 to 18 MeV in N_2 and Ar gases.) Recently, Peek, Weihofen, and Green have reported more exact calculations for specific electronic transitions in a molecular-hydrogen target,[7] and have obtained a total dissociation cross section using a closure approximation. The approaches of Salpeter and of Peek *el at.* give results that are in very good agreement with each other.

In this paper we report measurements of the total and partial gas collisional cross sections in H_2, He, N_2, and Ar at a sufficiently high energy, 20.9 MeV, that the Salpeter model should apply in all cases. The ion source was a high-voltage PIG type that is assumed to produce ions with an initial vibrational distribution calculable by use of the Franck-Condon principle. We have used Salpeter's approach to calculate breakup cross sections for all of the bound vibrational levels of the H_2^+ molecule.

Several measurements with gas-cell targets have been reported for energies above 1 MeV (approximately the energy for which the model should begin to give reasonable results). Barnett,[8] Sweetman and Riviere,[9] and Pivovar, Tubaev, and Novikov[10] have reported H_2, He, N_2, and Ar data at maximum energies of 2.25, 3, and 1.2 MeV, respectively. Goldring *el at.*[11] made measurements in H_2 and N_2 at 3 MeV, and Röpke and Spehl[12] went up to 3.5 MeV in Ar. Their results are included for comparison with our measurements.

As an alternative to gas-cell measurements, it is possible to deduce total cross sections by observing the variation with radius of the internal beam of a cyclotron,[6] provided certain assumptions are made about the dependence of the cross section on the H_2^+ energy and about the operating parameters of the cyclotron. We mention briefly some internal-cyclotron-beam

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[1] A. C. Riviere and D. R. Sweetman, Proc. Phys. Soc. (London) **78**, 1215 (1961).

[2] C. F. Barnett and J. A. Ray, in *Atomic Collision Processes*, edited by M. R. C. McDowell (North-Holland Publishing Company, Amsterdam, 1964), p. 743.

[3] J. W. McGowan and L. Kerwin, Can. J. Phys. **42**, 972 (1964).

[4] N. N. Tunitskii, P. M. Smirnova, and M. V. Tknomirnov, Dokl. Akad. Nauk SSSR **101**, 1083 (1955).

[5] E. E. Salpeter, Proc. Phys. Soc. (London) **A63**, 1297 (1950).

[6] K. E. A. Effat, Proc. Phys. Soc. (London) **A65**, 29 (1952).

[7] James M. Peek, W. H. Weihofen, and Thomas A. Green, in *Abstracts of the Fourth International Conference on the Physics of Electronic and Atomic Collisions, Quebec 1965* (Science Bookcrafters, Hastings-on-Hudson, New York, 1965), Paper NB 3; and James M. Peek, Sandia Laboratory, Albuquerque, New Mexico (private communication). See also Phys. Rev. **140**, A11 (1965).

[8] C. F. Barnett in *Proceedings of Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 32, p. 398.

[9] D. R. Sweetman, Proc. Roy. Soc. (London) **A256**, 416 (1960); and A. C. Riviere and D. R. Sweetman (private communication).

[10] L. I. Pivovar, V. M. Tubaev, and M. T. Novikov, Zh. Eksperim. i Teor. Fiz. **40**, 34 (1961) [English transl.: Soviet Phys.—JETP **13**, 23 (1961)].

[11] G. Goldring, D. Kedem, U. Smilansky, and Z. Vager, Nucl. Instr. Methods **23**, 231 (1963).

[12] H. Röpke and H. Spehl, Nucl. Instr. Methods **17**, 169 (1962).

measurements in the Berkeley 88-in. cyclotron for H_2^+ ions in the energy range 10 to 65 MeV, but our main emphasis is on the gas-cell measurements in the Berkeley heavy-ion linear accelerator (Hilac) beam; no assumptions about energy dependence and cyclotron parameters enter these calculations.

II. THEORY

The gas collisional breakup of H_2^+ can result in dissociation (denoted by the cross section σ_1)

$$\sigma_1: H_2^+ \rightarrow H^+ + H^0 \tag{1}$$

or ionization (denoted by σ_2)

$$\sigma_2: H_2^+ \rightarrow H^+ + H^+ + e. \tag{2}$$

Electron capture is not important for H_2^+ energies above a few hundred keV. The contributions to σ_1 from the $1s\sigma_g$-$2p\sigma_u$ electronic transition have been investigated by Peek[7] in the first Born approximation for collisions with H; collisions with H_2 are being investigated by Peek, Weihofen, and Green.[7] We know of no theoretical calculations for collisions with other gases; however, a method for estimating these cross sections at incident speeds that are large compared to the orbital speed of the bound electron in the H_2^+ ion has been prescribed by Salpeter[5]: The breakup of H_2^+ may be achieved either by electronic excitation to an unstable state or by excitation of nuclear vibrations. Salpeter reduced the problem at high-impact energies to a study of collisions in which (a) a free electron of the same velocity as the H_2^+ ion experiences a change of momentum K sufficient either to excite an unstable electronic state of the H_2^+ ion ($K^2/2m_e > 12.5$ eV) or to remove the bound electron ($K^2/2m_e > 30$ eV); and (b) a free proton of the same velocity as the H_2^+ ion experiences a change of momentum K sufficient to dissociate H_2^+ ions ($K^2/2M_p > 2.65$ eV). (The numerical values are for the ground vibrational state.) He calculated the cross sections for the necessary momentum transfers for collisions with atoms of H, N, O, and Ar, using Mott and Massey's results[13] for large-angle scattering in the first Born approximation.

Although the Salpeter calculations are for H_2^+ ions in the ground vibrational state ($v=0$), the H_2^+ ions used in our experiment were undoubtedly distributed over various vibrational states. To determine the effects of such a distribution we have used the Salpeter prescription to calculate σ_1 and σ_2 for all vibrational states ($v=0$ to 18) of an H_2^+ ion in the lowest rotational state. The effect of a distribution over rotational states has not been estimated.

The breakup resulting from electronic excitation of H_2^+ was determined as follows: For a particular internuclear separation R, we calculated the probability that a free electron of the same velocity as the H_2^+ ion undergoes a change of momentum K sufficient to excite the $2p\sigma_u$ state (leading to the total breakup cross section)

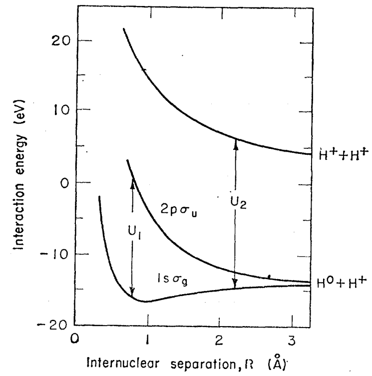

FIG. 1. Potential-energy diagram.

$$\frac{K^2}{2m_e} \geqslant U_1(R) \equiv |V(2p\sigma_u) - V(1s\sigma_g)|_R \tag{3}$$

or the two-proton state (ionization)

$$\frac{K^2}{2m_e} \geqslant U_2(R) \equiv |V(H^+ + H^+) - V(1s\sigma_g)|_R. \tag{4}$$

The quantities U_1 and U_2 are indicated in Fig. 1.

Momentum transfer to the electron can be achieved by either elastic or inelastic scattering from the target. The elastic scattering contribution to the cross section for each of these processes was obtained by integrating the differential scattering cross sections given by Motz, Olsen, and Koch[14] from

$$\theta_{\min}(R) = \left(\frac{2U(R)}{m_e v^2}\right)^{1/2} \tag{5}$$

to π. Thus we have

$$\sigma_{2,\mathrm{el}}(R) = 2\pi \int_{\theta_{\min}(U_2)}^{\pi} I(\theta) \sin\theta \, d\theta, \tag{6}$$

and

$$\sigma_{T,\mathrm{el}}(R) = 2\pi \int_{\theta_{\min}(U_1)}^{\pi} I(\theta) \sin\theta \, d\theta, \tag{7}$$

where $I(\theta)$ is the differential elastic-scattering cross section and σ_T is the total breakup cross section, $\sigma_T \equiv \sigma_1 + \sigma_2$, due to electronic excitation of H_2^+.

Two different methods were used to calculate the contribution from inelastic scattering. One was to use the differential scattering cross sections of Marton and Schiff[15] (based on first Born approximation with

[13] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Clarendon Press, Oxford, England, 1949).

[14] J. W. Motz, H. Olsen, and H. W. Koch, Rev. Mod. Phys. **36**, 881 (1964).

[15] L. Marton and L. I. Schiff, J. Appl. Phys. **12**, 759 (1941).

Hartree-Fock wave functions) for small momentum transfer, and those of Morse[16] (first Born approximation with Thomas-Fermi screening) for large momentum transfer; a graphical interpolation was used for the intermediate region. Since the validity of our method is restricted to high impact energies, Eq. (5) was also used to relate the energy transfer to the scattering angle for inelastic collisions. Alternatively, Gryzinski's[17] "semiclassical" result for transferring energy greater than U [Eq. (10), Ref. 17] was used. The value of U used in evaluating this equation was the greater of $U_1(U_2)$ or the lowest excitation energy of the target molecule under consideration.

The contribution from momentum transfer to either of the two protons, producing dissociation by vibrational excitation ($K^2/2M \geqslant$ the binding energy of vibrational state v), was calculated from the approximate formula used by Salpeter:

$$\sigma_p = 4\pi(Z+Z^2)e^4[E_{\text{in}}E(v)]^{-1}, \tag{8}$$

where Z is the atomic number of the target, E_{in} is the H_2^+ energy, and $E(v)$ is the binding energy of the vibrational state v. For the lower lying levels, the contributions to the breakup cross sections from vibrational excitation are small. However, the implication by this formulation that cross sections for excitation to nearby vibrational levels are quite large thereby suggests the possibility of dissociation through cascade processes. The effects of cascading are not included in our analysis.

The cross section for the dissociation or ionization of a particular vibrational state v was then obtained by averaging $\sigma(R)$ over all internuclear positions described by the vibrational wave functions of Cohen, Hiskes, and Riddell[18]:

$$\sigma_2(v) = \sigma_{2,\text{el}}(v) + \sigma_{2,\text{inel}}(v) = \int \psi_v^2(R)[\sigma_{2,\text{el}}(R) + \sigma_{2,\text{inel}}(R)]R^2\, dR, \tag{9}$$

$$\sigma_T(v) = \sigma_{T,\text{el}}(v) + \sigma_{T,\text{inel}}(v) + \sigma_p = \int \psi_v^2(R)[\sigma_{T,\text{el}}(R) + \sigma_{T,\text{inel}}(R)]R^2\, dR + \sigma_p(v), \tag{10}$$

$$\sigma_1(v) = \sigma_{1,\text{el}}(v) + \sigma_{1,\text{inel}}(v) + \sigma_p = \sigma_T(v) - \sigma_2(v). \tag{11}$$

Although we have integrated over the internuclear separations [Eqs. (9) and (10)] to obtain the results given in this article, considerable computational effort can be saved by considering ψ_v to be localized at the classical outer turning points in the vibrational potential well. We find that with this approximation we tend to overestimate σ_2 by about 5 to 15% and σ_T by as much as 40%.

[16] P. M. Morse, Physik. Z. **33**, 443 (1932).
[17] M. Gryzinski, Phys. Rev. **138**, A336 (1965).
[18] S. Cohen, J. R. Hiskes, and R. J. Riddell, Jr., Lawrence Radiation Laboratory Report UCRL-8871, 1959 (unpublished).

III. EXPERIMENTAL PROCEDURE

The experimental arrangement is shown in Fig. 2. A momentum-analyzed beam of H_2^+, (nominally 20 MeV) from the Berkeley Hilac passed through the second 24-cm long gas cell (shown at the bottom of the figure) through a pair of magnets, and into plastic scintillator-photomultiplier counters. The signals from the counters were pulse-height analyzed to demonstrate that the proper particles were being counted; discriminators were set so as to make contributions from low-energy scattered particles negligible, and the pulses were scaled. The target gases were hydrogen, helium, nitrogen, and argon at pressures ranging from 1 to 100 mTorr. The $\frac{1}{4}$-in.-diam beam was easily contained in the two-in.-diam plastic scintillator counters. The method of taking data was to set a pressure, use the neutral hydrogen beam as a monitor, and count the H^+ and H_2^+ beams in the other counter by varying the magnetic field in the analyzing magnet. In a given run enough counts were obtained to give a maximum of 3% statistical uncertainty in a measurement, and measurements made at different times and with different apparatus over a period of one year gave consistent results.

Although the targets were fairly thin for beams of this energy, it was necessary to know the cross sections for ionizing neutral hydrogen atoms in interpreting the data. These cross sections were obtained by breaking up the H_2^+ atoms in the first gas target, sweeping the ions out of the way with immediately following magnet and determining ionization cross sections of the hydrogen atom with the second gas cell.

Background corrections were necessary for both the H^0 and H^+ count rates. The H^0 background was due to collisional breakup of H_2^+ ions in the low pressure but fairly long-drift tubes between the targets and magnets, and was always repeatable. The background in the H^+ count rate was also due in part to H_2^+ breakup on gas in the drift tubes, but to a greater degree was from grazing collisions with the collimators of the gas cell. Variation of the size of the contribution from the collimator collisions, due to beam steering from the accelerator, resulted in somewhat larger uncertainties in the σ_2 cross section than in σ_1. During the experiment gas pressures in the targets were monitored with Schulz-Phelps ionization gauges that were cross calibrated with McLeod gauges and an oil manometer. The Gaede effect[19] was determined by repeating the pressure calibrations with the mercury of the McLeod gauge cooled to 0°C.[19] The maximum effect was 5% for the pressures used in this experiment. From the internal consistency of the pressure calibrations, including day-to-day fluctuations, we set a maximum uncertainty of ±10% in target thickness.

[19] See, for example, Ch. Meinke and G. Reich, Vacuum **13**, 579 (1963), or Vacuum Tech. **12**, 79 (1963).

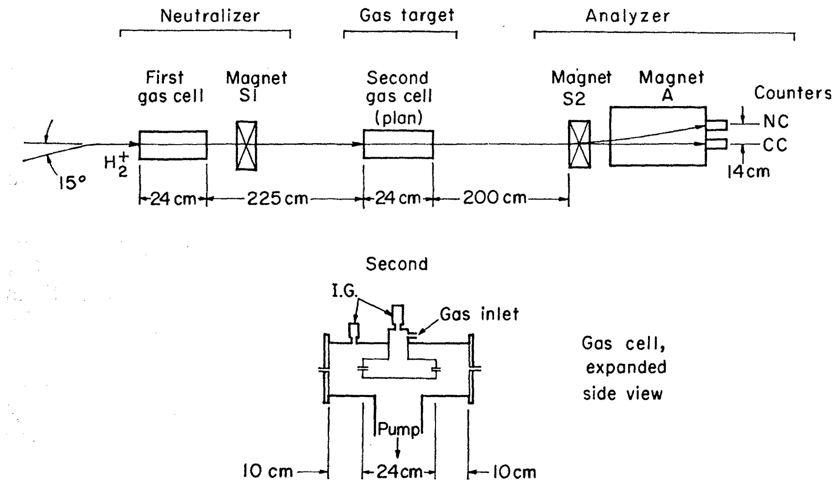

FIG. 2. Experimental arrangement.

After combining these various effects we estimate that the standard error in σ_1 is $\pm 15\%$ and in σ_2 is $\pm 20\%$.

Radial beam-attenuation measurements were also carried out in the Berkeley 88-in. cyclotron (with the cooperation of Hermann Grunder) for 10- to 65-MeV H_2^+ ions in hydrogen, helium, nitrogen, and argon. As the results of careful measurements of the beam phase with respect to the rf as a function of radius were available,[20] the energy gain per turn could be determined precisely. Attenuation measurements were made at a number of pressures for each gas; with the assumption that the cross sections decrease as $1/E$, it was possible to make simple corrections for the background gas of unknown composition inside the cyclotron. However, there was an uncertainty in the pressure measurements because the ion gauges were at the vacuum wall rather than in the cyclotron gap and, in addition, only total current to the probe was measured, so that contributions from low-energy particles may have been present. If we neglect the latter factor, we might conclude that the cross-section values obtained in this way should be good to about $\pm 30\%$, although an accurate estimate of the uncertainty is impossible. The total attenuation cross sections obtained with this technique agreed with the gas-cell results to within the 30% uncertainty estimate.

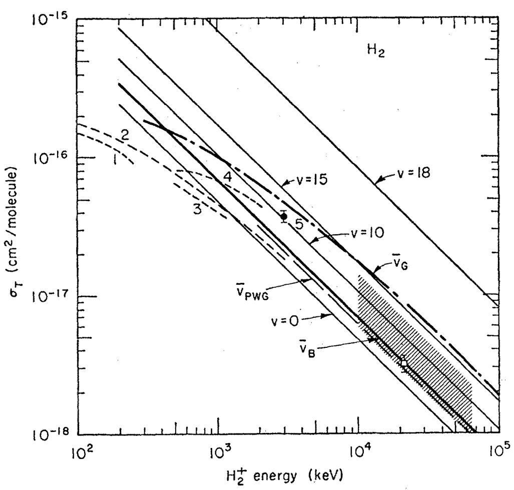

FIG. 3. Total breakup cross section, $\sigma_T = \sigma_1 + \sigma_2$, in hydrogen. The solid lines are the Born approximation results of Table II plus similar calculations at other energies. These results are inversely proportional to the H_2^+ kinetic energy. The broken line labeled $\bar{v}_G$ is for an average over vibrational levels, when the Gryzinski inelastic cross sections are used. The broken line labeled $\bar{v}_{PWG}$ is the averaged cross section obtained by Peek *et al.* in a closure approximation. The square at 20.9 MeV is our gas-cell measurement, and the cross-hatched area indicates the value obtained from our internal-cyclotron-beam measurement, with its uncertainty of $\pm 30\%$. Other experimental results are (1) J. Guidini, *Ionization Phenomena in Gases, Munich 1961* (North-Holland Publishing Company, Amsterdam, 1962), p. 1228; (2) Ref. 9; (3) Ref. 10; (4) Ref. 8; and (5) Ref. 11.

[20] These measurements were made concurrently by Alper Garren and Lloyd Smith for another purpose. We are indebted to them for their results and interpretation.

IV. RESULTS

The results of the 20.9-MeV H_2^+ and 10.4-MeV H^0 cross-section measurements are summarized in Table I. Values of the total cross sections from this table are plotted in Figs. 3 through 6. Also shown are the results of a number of other experiments at energies above 100 keV and the average values of σ_T in the energy range 10 to 65 MeV that were deduced from our internal-cyclotron-beam measurements and Effat's similar measurements at 9 to 18 MeV. It must be emphasized that we have no real way to estimate the

TABLE I. Cross sections per target molecule in units of 10^{-18} cm^2. The absolute uncertainties in σ_1 and σ_2 are estimated to be ± 15 and $\pm 20\%$, respectively. The uncertainty in the ratio σ_2/σ_1 is about 20%.

	20.9-MeV H_2^+				10.4-MeV H^0
	σ_1	σ_2	σ_T	σ_2/σ_1	σ_{01}
H_2	1.53	1.69	3.22	1.10	2.24
He	1.06	1.77	2.83	1.67	1.59
N_2	10.9	20.3	31.2	1.88	19.9
Ar	12.3	35.4	47.7	2.87	35.8

TABLE II. Calculated cross sections for 20-MeV H_2^+ ions in hydrogen, in units of 10^{-18} cm²/molecule.[a]

		$\sigma_{1,\text{inel}}$			σ_1			$\sigma_{2,\text{inel}}$		σ_2	
v	$\sigma_{1,\text{el}}$	B	G	σ_p	B	G	$\sigma_{2,\text{el}}$	B	G	B	G
0	0.24	1.07	6.42	0.02	1.33	6.68	0.51	0.63	1.68	1.14	2.19
1	0.26	1.31	6.60	0.02	1.59	6.88	0.54	0.68	1.84	1.22	2.38
2	0.25	1.42	6.34	0.02	1.70	6.61	0.52	0.66	1.83	1.18	2.35
3	0.26	1.58	6.21	0.03	1.87	6.50	0.52	0.66	1.87	1.18	2.39
4	0.28	1.82	6.06	0.03	2.13	6.37	0.54	0.71	2.02	1.24	2.56
5	0.29	2.02	6.42	0.04	2.35	6.75	0.55	0.72	2.10	1.27	2.65
6	0.30	2.24	6.41	0.04	2.58	6.75	0.56	0.74	2.20	1.30	2.75
7	0.31	2.47	6.41	0.05	2.83	6.76	0.56	0.76	2.30	1.33	2.86
8	0.32	2.72	6.23	0.06	3.10	6.60	0.57	0.79	2.41	1.36	2.98
9	0.31	2.95	6.22	0.07	3.33	6.60	0.56	0.79	2.44	1.35	3.00
10	0.32	3.42	6.28	0.09	3.82	6.68	0.59	0.84	2.67	1.46	3.26
11	0.33	3.64	6.19	0.11	4.07	6.63	0.58	0.84	2.68	1.41	3.26
12	0.34	4.18	6.32	0.15	4.68	6.82	0.60	0.89	2.90	1.49	3.50
13	0.34	4.61	6.19	0.20	5.16	6.74	0.60	0.90	3.00	1.51	3.61
14	0.35	5.40	6.05	0.31	6.06	6.71	0.62	0.94	3.25	1.55	3.86
15	0.36	6.17	6.03	0.51	7.04	6.90	0.63	0.98	3.39	1.60	4.01
16	0.36	7.42	5.93	1.05	8.84	7.34	0.62	1.02	3.58	1.66	4.22
17	0.36	9.12	5.62	3.15	12.63	9.14	0.65	1.07	3.92	1.72	4.57
18	0.35	10.3	5.20	29.1	39.8	34.7	0.67	1.15	4.42	1.82	5.10
Av.	0.28	1.96	6.37	0.05	2.29	6.70	0.54	0.71	2.06	1.25	2.60

[a] The elastic-scattering contributions $\sigma_{1,\text{el}}$ and $\sigma_{2,\text{el}}$ are calculated in the Born approximation. The inelastic contributions are calculated for both the Born and Gryzinski approximations. Total values of σ_1 and σ_2 are given in double entry indicating that Born (B) or Gryzinski (G) inelastic cross sections are used. The Gryzinski calculations are based on molecular values for the excitation and ionization energies. All other entries are twice the cross sections for a hydrogen atom.

TABLE III. Calculated cross sections for 20-MeV H_2^+ ions in helium, in units of 10^{-18} cm²/atom. See Table II footnote.

		$\sigma_{1,\text{inel}}$			σ_1			$\sigma_{2,\text{inel}}$		σ_2	
v	$\sigma_{1,\text{el}}$	B	G	σ_p	B	G	$\sigma_{2,\text{el}}$	B	G	B	G
0	0.12	0.60	1.48	0.029	0.75	1.63	0.55	0.68	2.08	1.23	2.63
1	0.13	0.68	1.46	0.032	0.84	1.62	0.58	0.73	2.29	1.31	2.87
2	0.13	0.69	1.26	0.036	0.86	1.43	0.55	0.71	2.28	1.26	2.83
3	0.13	0.73	1.13	0.041	0.90	1.30	0.55	0.71	2.35	1.25	2.89
4	0.14	0.81	1.06	0.047	0.99	1.24	0.57	0.75	2.53	1.32	3.10
5	0.14	0.87	0.94	0.054	1.06	1.13	0.57	0.76	2.65	1.33	3.22
6	0.14	0.93	0.82	0.062	1.13	1.02	0.58	0.78	2.77	1.35	3.35
7	0.14	1.00	0.70	0.073	1.21	0.92	0.58	0.79	2.89	1.37	3.47
8	0.15	1.08	0.58	0.087	1.32	0.82	0.58	0.82	3.00	1.40	3.58
9	0.14	1.14	0.52	0.105	1.38	0.76	0.57	0.80	2.98	1.38	3.55
10	0.16	1.29	0.40	0.130	1.58	0.69	0.59	0.86	3.19	1.44	3.77
11	0.14	1.35	0.40	0.166	1.66	0.71	0.58	0.84	3.12	1.43	3.70
12	0.15	1.52	0.35	0.220	1.89	0.72	0.60	0.89	3.27	1.49	3.88
13	0.15	1.66	0.30	0.307	2.12	0.75	0.60	0.89	3.29	1.49	3.88
14	0.15	1.92	0.24	0.461	2.53	0.85	0.61	0.92	3.35	1.53	3.96
15	0.15	2.15	0.18	0.772	3.07	1.10	0.62	0.94	3.42	1.56	4.04
16	0.15	2.51	0.12	1.58	4.24	1.86	0.62	0.97	3.48	1.60	4.10
17	0.15	2.96	0.07	4.73	7.84	4.95	0.63	1.00	3.52	1.63	4.14
18	0.14	3.26	0.02	43.7	47.1	43.9	0.64	1.05	3.57	1.69	4.21
Av.	0.14	0.85	1.02	0.079	1.07	1.24	0.57	0.75	2.55	1.32	3.12

uncertainties of our cyclotron values so the data are of only casual interest.

In Tables II through V are the calculated dissociation cross sections for all 19 vibrational levels. The results are broken down to show the contributions from each of the terms in Eqs. (9) and (11). The inelastic and total cross sections are tabulated for both a Born approximation (B) and a semiclassical, Gryzinski calculation (G) of the inelastic contribution. The tabulated cross sections for $\bar{v}$ were obtained by averaging over a vibrational-level population distribution calculated[21] according to the Franck-Condon principle. For H_2 and N_2 the Born approximation cross sections given are simply twice the calculated atomic cross sections. For the Gryzinski inelastic scattering probabilities, however, we interpreted the "minimum excitation energy U," used in his Eqs. (7) and (10) of Ref. 17, to be the lowest electronic excitation energy of the molecule.[22] The molecular targets could also have been treated as two atoms, as in the Born calculations. In this case, the only large difference is in $\sigma_{1,\text{inel}}$, the value of which is about 40% lower for two atoms for a molecule of N_2, and 15% higher for H_2.

[21] J. R. Hiskes, Nucl. Fusion 2, 38 (1962).

[22] For example, the six $2p$ electrons of N_2 were considered to have an excitation threshold of 6.1 eV, and the four $2s$ electrons 10.3 eV.

TABLE IV. Calculated cross sections for 20-MeV H_2^+ ions in nitrogen, in units of 10^{-18} cm²/molecule. See Table II footnote.

		$\sigma_{1,\text{inel}}$			σ_1			$\sigma_{2,\text{inel}}$		σ_2	
v	$\sigma_{1,\text{el}}$	B	G	σ_p	B	G	$\sigma_{2,\text{el}}$	B	G	B	G
0	4.9	3.8	36.0	0.5	9.3	41.4	14.0	5.5	8.4	19.5	22.4
1	5.3	4.3	48.9	0.6	10.2	54.8	14.9	5.9	9.2	20.8	23.2
2	5.2	4.4	62.0	0.7	10.3	68.0	14.2	5.7	9.1	19.9	23.3
3	5.2	4.6	70.9	0.8	10.5	76.7	14.1	5.7	9.3	19.7	23.4
4	5.5	5.1	74.5	0.9	11.6	80.9	14.7	6.0	10.1	20.6	24.7
5	5.7	5.5	78.6	1.0	12.2	85.3	14.8	6.1	10.5	20.9	25.3
6	5.8	6.0	81.4	1.2	12.8	88.3	14.9	6.1	11.0	21.2	25.9
7	5.9	6.4	83.9	1.4	13.6	91.2	15.1	6.3	11.4	21.4	26.6
8	6.0	6.9	86.4	1.6	14.5	94.1	15.4	6.4	12.0	21.8	27.4
9	5.9	7.2	86.4	2.0	15.1	94.3	15.0	6.3	12.2	21.3	27.2
10	6.3	8.2	92.6	2.4	16.9	101	15.8	6.7	13.3	22.5	29.1
11	6.2	8.6	90.2	3.1	17.8	100	15.4	6.5	13.4	22.0	28.8
12	6.4	9.7	94.4	4.1	20.2	105	16.0	6.9	14.5	22.9	30.5
13	6.4	10.5	94.8	5.7	22.7	107	16.0	6.9	15.0	22.9	31.0
14	6.5	12.2	96.1	8.6	27.4	111	16.2	7.1	16.2	23.3	32.4
15	6.6	13.5	98.0	14.4	34.5	119	16.5	7.2	16.9	23.7	33.4
16	6.6	15.0	96.8	29.5	51.2	133	16.7	7.4	17.8	24.2	34.6
17	6.7	16.1	99.2	88.3	111	194	16.8	7.6	19.5	24.4	36.3
18	6.4	17.1	98.7	816	840	920	16.4	7.9	22.1	25.3	39.4
Av.	5.5	5.4	71.3	1.5	12.4	78.3	14.7	6.0	10.3	20.7	25.9

TABLE V. Calculated cross sections for 20-MeV H_2^+ ions in argon, in units of 10^{-18} cm²/atom. See Table II footnote.

		$\sigma_{1,\text{inel}}$			σ_1			$\sigma_{2,\text{inel}}$		σ_2	
v	$\sigma_{1,\text{el}}$	B	G	σ_p	B	G	$\sigma_{2,\text{el}}$	B	G	B	G
0	11.0	3.8	16.6	1.7	16.4	29.2	31.6	5.8	7.4	37.4	38.9
1	10.3	4.2	16.4	1.8	16.4	28.5	33.4	6.2	7.9	39.6	41.4
2	11.6	4.3	15.2	2.1	18.0	28.9	32.0	6.0	7.8	38.0	39.8
3	11.7	4.6	14.8	2.3	18.7	28.9	31.7	6.0	7.8	37.7	39.6
4	12.4	5.0	15.1	2.7	20.1	30.2	33.1	6.3	8.3	39.4	41.5
5	12.6	5.4	14.9	3.1	21.1	30.6	33.4	6.4	8.6	39.8	42.0
6	13.0	5.8	14.8	3.5	22.3	31.3	33.8	6.5	8.9	40.2	42.7
7	13.2	6.2	14.7	4.2	23.6	32.1	34.1	6.6	9.2	40.7	43.3
8	13.5	6.8	14.7	4.9	25.2	33.2	34.7	6.7	9.5	41.4	44.3
9	13.3	7.1	13.9	6.0	26.4	33.3	33.8	6.6	9.6	40.4	43.4
10	14.2	8.1	14.3	7.4	29.6	35.9	35.6	7.0	10.3	42.6	46.0
11	13.8	8.4	13.6	9.5	31.7	37.1	34.7	6.8	10.3	41.6	45.0
12	14.5	9.5	13.8	12.5	36.5	40.8	36.1	7.1	11.0	43.6	47.2
13	14.4	10.2	13.4	17.5	42.0	45.2	36.1	7.2	11.3	43.3	47.4
14	14.6	11.2	12.8	26.3	52.1	53.7	36.6	7.3	12.1	43.9	48.7
15	14.7	11.4	12.6	44.0	70.1	71.3	37.2	7.5	12.5	44.7	49.7
16	14.8	11.7	12.2	90.1	117	117	37.8	7.7	12.5	45.4	50.9
17	14.7	12.4	11.2	270	287	296	38.2	7.9	14.1	46.1	52.3
18	14.4	12.7	9.8	2490	2520	2520	39.1	8.2	15.6	47.3	54.8
Av.	12.2	5.3	15.0	4.5	27.0	31.8	33.1	6.3	8.5	39.4	41.6

The solid lines of Figs. 3 through 6 are $1/E$ extrapolations of the Born-approximation results for representative values of v and $\bar{v}$. The $1/E$ dependence of the cross sections was confirmed by calculations at lower energies. Plotted as a broken line on each figure is the total cross section according to the Gryzinski calculation. For illustrative purposes the curves are extended to lower energies than the Born approximation warrants.

V. DISCUSSION

Our values for measured high-energy breakup cross sections are in good agreement with calculated values for averages over a Franck-Condon distribution of level populations,[23] when first-Born approximation values are used for both the elastic and inelastic electron-scattering probabilities. Inelastic-scattering contributions from the Gryzinski semiclassical approach give apparently excessive values for the molecular gases.

A successful model should also predict the relative values of σ_1 and σ_2. Examples of the ratio σ_2/σ_1 for a few vibrational levels are given in Table VI. The trend with Z, and the ratios for $v=0$ and $\bar{v}$ calculated on the basis of the Born approximation throughout, are in

[23] We are not suggesting that the experimental H_2^+ beam has a vibrational population distribution given by the Franck-Condon principle, even if it were created with one in the ion source. However, this distribution seems to be a plausible one to try.

TABLE VI. The ratio σ_2/σ_1 for several vibrational levels with the inelastic contributions calculated from both the Born and the Gryzinski formulas.

	v	H_2	He	N_2	Ar
Born	0	0.86	1.64	2.10	2.28
	5	0.54	1.25	1.71	1.88
	10	0.38	0.91	1.33	1.44
	18	0.05	0.04	0.03	0.02
	$\bar{v}_B$	0.55	1.23	1.67	1.79
Gryzinski	0	0.33	1.61	0.54	1.33
	5	0.39	2.85	0.30	1.37
	10	0.49	5.46	0.29	1.28
	18	0.15	0.10	0.04	0.02
	$\bar{v}_G$	0.39	2.52	0.33	1.31
	Exptl.	1.10 ±0.22	1.67 ±0.35	1.88 ±0.38	2.87 ±0.57

rough agreement with our measurements and with the results of the other experiments referenced in this paper. Unfortunately, neither the experimental results nor our knowledge of the vibrational distribution are precise enough to permit a good comparison.

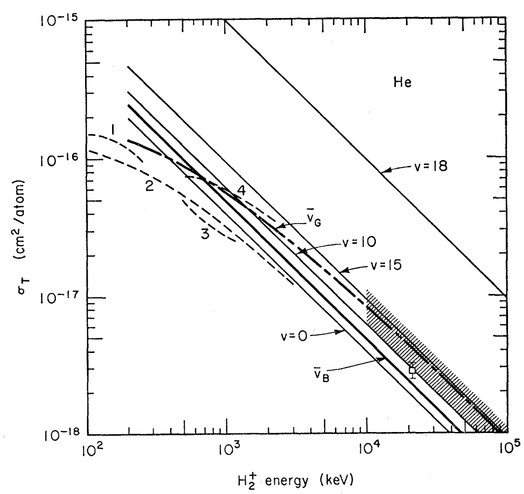

FIG. 4. The total breakup cross section, $\sigma_T=\sigma_1+\sigma_2$, in helium. See also Fig. 3 caption.

The excellent agreement of the average cross section in H_2 calculated by Peek *et al.*[7] (see Fig. 3) with that calculated in the Salpeter approximation, and the good agreement between our measured and calculated cross sections in H_2, He, N_2, and Ar inspired considerable confidence in the usefulness of Salpeter's approach. As mentioned earlier, the calculated values of the vibrational excitation terms (σ_p) do not make a significant contribution to the cross sections because they are relatively small for the lower lying vibrational states (the averaged cross sections correspond roughly to those for $v=4$ or 5, which are two of the most heavily weighted levels for Franck-Condon transitions). However, since σ_p varies inversely with threshold energy [Eq. (8)], and since the binding energy of the highest

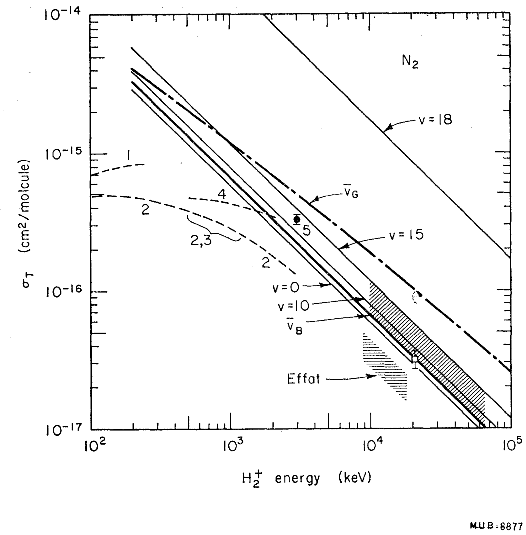

FIG. 5. The total breakup cross section, $\sigma_T=\sigma_1+\sigma_2$, in nitrogen. The cross-hatched area (Effat) shows the results of Ref. 6. See also Fig. 3 caption.

vibrational levels is quite small, this term becomes extremely large and, in fact, dominates the cross sections at the highest levels.

The only high-energy experiment that we are acquainted with that bears on collisional dissociation of the highly excited vibrational levels was reported by Riviere and Sweetman.[24] They measured the fraction of the total H_2^+ beam that was in the uppermost vibrational levels by electric field dissociation. After passing the beam through a gas target sufficiently

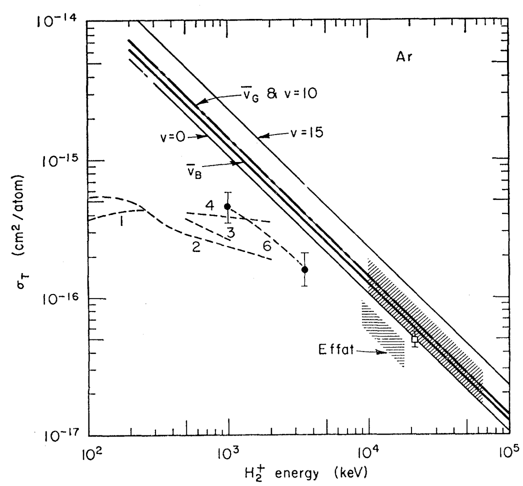

FIG. 6. The total breakup cross section, $\sigma_T=\sigma_1+\sigma_2$, in argon. The experimental curve labeled 6 is from Ref. 12. The cross-hatched area (Effat) shows the results of Ref. 6. See also Fig. 3 caption.

[24] A. C. Riviere and D. R. Sweetman, in *Proceedings of the Fifth International Conference on Ionization Phenomena in Gases, Munich, 1961* (North-Holland Publishing Company, Amsterdam, 1962), p. 1236.

thick to dissociate a large fraction of the incoming beam, they found that the fraction of the emerging beam in the uppermost levels had dropped only slightly. The small decrease does not necessarily imply a failure in the model, since the form of Eq. (8) and the philosophy in which it was used would also imply very large cross sections for excitation to neighboring vibrational levels —suggesting a mechanism for repopulation of the higher levels by an upward cascade. This characteristic of the cross section, however, remains to be demonstrated.

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Coincidence Measurements of Large-Angle Ar^+-on-Ar Collisions*

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Large-angle single collisions of keV-energy Ar^+ ions with Ar atoms are studied wherein both particles scattered from the same encounter are detected in coincidence. The scattered incident particle appears m-times ionized at angle θ and the recoiling target particle appears n-times ionized at angle ϕ. Values of m and n range from zero to eight. It is found that m and n are independent and uncorrelated, i.e., the distribution among charge states m is the same regardless of the charge state n seen in coincidence. An exception to this rule is seen in one region where the inelastic energy has multiple values. Relative probabilities for the (m,n) reaction are given for many data sets, with incident energies T_0 from 3 to 400 keV and for angles θ between 8° and 40°. The inelastic energy $\bar{Q}_{mn}$ associated with the (m,n) reaction is also measured for a number of values of m and n in each data set. It is found that a particular reaction does not have a fixed characteristic energy. Thus, for example, $\bar{Q}_{55}$ increases from 877 to 1473 eV depending on the violence of the collision. Average values of inelastic energy loss $\bar{Q}$ are plotted versus incident energy at various scattering angles, versus the average number of electrons lost in the collision, and versus the distance of closest approach. Values of $\bar{Q}$ range from 26 eV at $T_0=3$ keV, $\theta=8°$, to 2430 eV at $T_0=300$ keV, $\theta=40°$. The effects of thermal motion of the target atom, of finite instrumental resolution, and of a possible distribution in values of inelastic energy all combine to give "linewidth" effects. These are measured and discussed.

1. INTRODUCTION

COINCIDENCE-scattering measurements in atomic physics are a recent development. The first such experiment was reported by Afrosimov, Gordeev, Panov, and Fedorenko,[1] who studied Ar^+-Ar collisions at 12 and 50 keV by this technique and interpreted the phenomena observed. Preliminary results from a parallel experiment in our laboratory have been described[2,3] and these together with the present experiment offer evidence in support of a rather different interpretation.[4]

The reaction under study is

$$Ar^{+}+Ar \rightarrow Ar^{+m}+Ar^{+n}+(m+n-1)e. \qquad (1)$$

The incident ion is scattered to angle θ with charge $+m$ and the recoil target particle is found at angle ϕ with charge $+n$. The present experiment includes the energy range 3 to 400 keV at angles θ between 8° and 40° and measures m and n for particles originating from the same collision. The fractional probability $\bar{p}_{mn}$ of the (m,n) event is determined. Furthermore, the inelastie energy lost $\bar{Q}_{mn}$ is measured for reactions in which both m and n are specified.

The Ar^+-Ar collision has been the subject of many papers prior to these coincidence studies. Thus Fedorenko,[5] Kaminker and Fedorenko,[6] Carbone *et al.*,[7]

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[1] V. V. Afrosimov, Yu. S. Gordeev, M. N. Panov, and N. V. Fedorenko. Zh. Tekhn. Fiz. **34**, 1613 (1964); **34**, 1624 (1964); **34**, 1637 (1964) [English transl.: Soviet Phys.—Tech. Phys. **9**, 1248 (1965); **9**, 1256 (1965); **9**, 1265 (1965)]. Additional recent measurements by these same authors may be found in Zh. Tekhn. Fiz. **36**, 123 (1966) [English transl.: Soviet Phys.—Tech. Phys. **36** (to be published) (1966)], and in JETP Pis'ma v Redaktsiyu **2**, 153 (1965) [English transl.: JETP Letters **2**, 185 (1965)].

[2] E. Everhart and Q. C. Kessel, Phys. Rev. Letters **14**, 247 (1965).

[3] Q. C. Kessel, A. Russek, and E. Everhart, Phys. Rev. Letters **14**, 484 (1965).

[4] E. Everhart and Q. C. Kessel, following paper, Phys. Rev. **146**, 27 (1966).

[5] N. V. Fedorenko, Zh. Tekhn. Fiz. **24**, 784 (1954).

[6] D. M. Kaminker and N. V. Fedorenko, Zh. Tekn. Fiz. **25**, 2239 (1955).

[7] R. J. Carbone, E. N. Fuls, and E. Everhart, Phys. Rev. **102**, 1524 (1956).