Negative-ion formation from dissociative collisions of H_2^+, H_3^+, and HD_2^+ in H_2, He, and Xe

Ignacio Alvarez and Carmen Cisneros
Instituto de Fisica, México, 20, D.F.

C. F. Barnett and J. A. Ray
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830*
(Received 15 December 1975)

Negative-ion formation cross sections have been determined for dissociative collisions of H_2^+, H_3^+, and HD_2^+ in H_2, He, and Xe in the energy region from 40 to 600 keV. In addition, the maximum and equilibrium H^- or D^- fractions for the three molecular ions in H_2 gas were determined. For equivelocity incident HD_2^+ and H_3^+, D^- or H^- formation cross sections were the same. For H_2^+ the H^- formation cross sections were smaller than those of the triatomic incident beams for velocities greater than 3.3×10^8 cm/sec. Equilibrium fractions were independent of the incident particle at equal velocities and were 0.024 at 2×10^8 cm/sec.

I. INTRODUCTION

At the present time there is considerable interest in experimental techniques for the production of energetic intense beams of negative hydrogen or deuterium ions. The interest arises from the need to provide a source of 150–500-keV deuterium atoms to heat a low-temperature plasma to a sufficiently high temperature, to maintain and produce a thermonuclear reaction.[1] One of the more common techniques for production of energetic neutral beams is through charge-exchange collisional processes. However, in this energy range the production of D^0 from D^+ by charge-exchange collisions is inefficient from considerations of both energy and particle conservation.

An alternate to this method is the collisional electron detachment of D^- to form D^0. Efficiencies are approximately 80% in this energy range. Two experimental methods are available to form a beam of D^- ions: (i) formation of D^- in an ion source and acceleration to the desired energy; (ii) acceleration of molecular ions and the subsequent dissociation of these ions to form D^-.

Collisional dissociative processes present interesting possibilities for formation of negative ions. Several reviews[2-4] have been published of the various channels through which molecular ions may dissociate. In addition, the dissociative cross sections have been intensively investigated. Three investigations have been made of the formation of negative hydrogen ions from dissociative collisions of hydrogen molecular ions in gases in the energy range 2–50 keV. Fedorenko *et al.*[5] found the H^- formation cross section for 12-keV H_2^+ in argon to be approximately 2×10^{-18} cm^2. Williams and Dunbar[6] have measured the total H^- production cross sections for 2–50-keV H_2^+ and H_3^+ molecular ions in H_2. In a more recent paper Meyer and Anderson[7] have measured the D^- equilibrium fraction for 3–23-keV D_2^+ passing through Cs vapor. Intensive investigations[8,9] have been carried out for proton-formation cross sections of H_2^+ and H_3^+ in various gases, and it was found that the cross sections were dependent on the initial vibrational state of the molecular ion.

The importance of this cross section and the lack of information in the desired energy range have motivated the present work. Theoretical work is not available for predicting the negative-ion–formation cross sections. In this paper we report the measurements of the H^- and D^- cross sections, and the charge-state equilibrium fractions of 40–600-keV H_2^+, H_3^+, and HD_2^+ in H_2, He, and Xe gases.

II. APPARATUS

The apparatus used for the cross-section measurements consisted basically of a differentially pumped gas chamber, a set of electrostatic deflection plates to separate the dissociated products, and detectors as shown in Fig. 1.

Beams of HD_2^+, H_3^+, and H_2^+ were produced in an rf ion source and accelerated to 40–600 keV with a conventional type of accelerator. Two sets of apertures with knife edges were used as collimators. The first set were 0.05 and 0.15 cm in diameter, and the second set defined the geometric length of the dissociation cell with apertures also of 0.05 and 0.15 cm. To ensure that scattering from the aperture edges was not influencing the measurements, the exit gas cell aperture was changed from 0.15 to 0.25 and to 0.3 cm.

The geometrical length of the dissociation chamber was 30.5 cm. Pumping speeds were sufficient

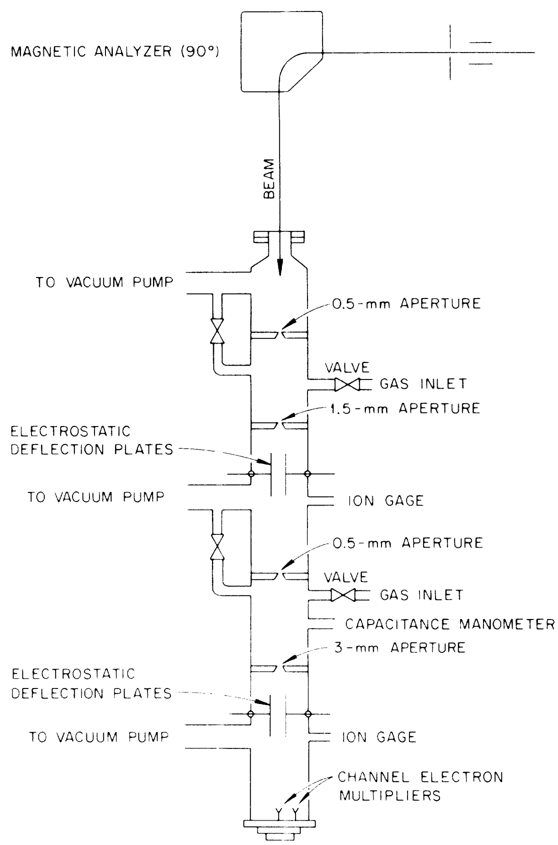

FIG. 1. Schematic drawing of the experimental apparatus.

so that a differential pressure of 1500 was maintained between the cell and the adjacent chamber. The entire bakable vacuum system, excluding the detector assembly, was made of stainless steel and assembled with metal O rings.

Funnel-type channel electron multipliers were used as detectors to measure both the neutral and charged components. The detectors were mounted so that they could be moved in both the vertical and horizontal position to ensure that the total scattered beam was collected by the 1-cm-diam detector. The detectors were operated in the charge saturation mode as suggested by Ray and Barnett.[10]

Target gas pressures were measured by means of a capacitance manometer which provided continuous pressure monitoring and measurement. The manometer was calibrated against a precision McLeod gauge, using H_2 gas in which the pumping effect was minimal.

III. EXPERIMENTAL PROCEDURE

Hydrogen gas of 99.98% purity was placed in the collision chamber where the molecular-ion beam undergoes dissociative collisions. The beams on emerging from the collision cell were passed through an electrostatic analyzer. By virtue of the dissociation products having less energy (e.g., D^- from HD_2^+ will have $\frac{2}{5}E_0$), the products are separated into the various components. Since an electron multiplier was used as the detector, it was impossible to separate the neutral components H_2, H, and D.

Dissociation cross sections have been calculated from the relation

$$I(D^-, H^-)/I_0 = 1 - e^{-\pi\sigma(D^-, H^-)}, \qquad (1)$$

where $I(D^-, H^-)$ represents the D^- or H^- intensity, I_0 is the incident molecular-ion intensity, π is the target thickness equal to $\int_{-\infty}^{\infty} d\pi = nl$, and n, l are the target density and effective length, respectively. The negative-ion fraction

$$F(D^-, H^-) \equiv I(D^-, H^-)/I_0, \qquad (2)$$

has two regions of interest: (i) a linear region for small n, where single collisions dominate; (ii) quadratic dependence for larger n, where multiple collisions are present. For single collisions Eq. (1) can be expanded, and by neglecting higher-order terms the fraction can be written as

$$F^- = \sigma(D^-, H^-)\pi .$$

As the target thickness π increases, the fraction F^- increases linearly and the cross section is obtained from the slope of the curve of F^- vs π. The slope was determined by fitting the curve to a least-squares fit. The cross-section values reported are the result of the average of several determinations. As gas cell pressure is increased beyond single-collision conditions, the F^- fraction is quadratic with pressure, reaches a maximum, and then decreases to an equilibrium value which is independent of any further increase in pressure. The equilibrium fraction is usually denoted by F_∞ and is defined as

$$F_\infty^- = \lim_{p\to\infty} F^- .$$

Equilibrium and peak fractions were measured for D^- and H^-, using HD_2^+, H_2^+, H_3^+, and the results have been compared for the same velocity of D^- and H^-.

IV. ERRORS

Barnett and Gilbody[11] have discussed in detail the errors in dissociative-type collisions. Systematic errors result mainly from: (i) the effective path length of the gas cell; (ii) deviations from thin-target conditions; (iii) impurities in the target gas; (iv) loss of reaction products on the exit

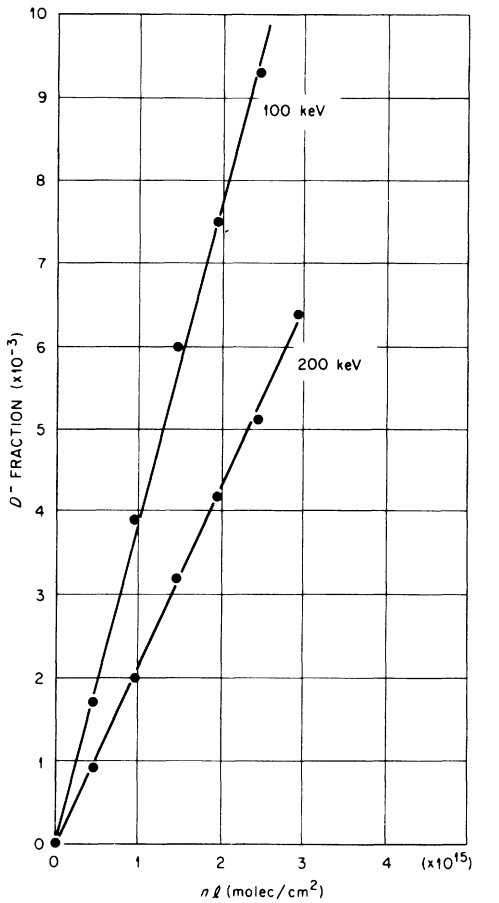

FIG. 2. Typical curves showing the linear growth of D^- at various energies of the HD_2^+ beam in hydrogen as a function of the target thickness, $\pi \equiv nl$.

aperture; (v) measurements of target gas cell pressure.

With the assumption of molecular flow, Toburen *et al.*[12] derived a simple way to estimate the effective path length, which is longer than the physical length because of gas streaming from the apertures. Our increased path length was estimated to be less than 1%.

In determining the absolute value of the cross section, the slope of F^- vs p was determined. Special precautions were taken to ensure that the slope was linear and that conditions of a thin target existed. Shown in Fig. 2 are examples of the linear growth. The density of the H_2 gas target was increased to 3×10^{15} molecules/cm^2 for D^- formation of 100–600-keV HD_2^+. In this range the curves were linear within an estimated 3%.

To be sure that the reaction products were not being intercepted by the exit aperture of the gas cell, the aperture diameter was increased from 1.5 to 2.5 mm. The D^- fraction increased 20% with 100-keV HD_2^+, 10% at 200 keV, 1–2% at 300 keV, and at higher energies no changes were detected. Increasing the exit-aperture diameter to 3 mm produced the same D^- fraction as was obtained with a 2.5-mm aperture.

If the detectors were incorrectly aligned, some of the reaction products would be lost. The alignment was checked continuously, especially at each change of energy. Since the energy stability of the accelerator was better than 10^{-5}, we estimate this source of error to be negligible.

Random uncertainties in the cross sections due to beam intensity and gas target densities can best be evaluated by the uncertainties encountered in obtaining the slope of the straight line through the data points of an F^- vs p plot. The slope can be repeated with uncertainties of less than 10%.

Errors encountered in the manometer calibration are less than 3%. The combined uncertainties of both random and systematic errors are estimated to be less than ± 12%.

V. RESULTS AND DISCUSSION

Figure 3 shows H^- and D^- production cross sections from collisions of H_2^+, H_3^+, and HD_2^+ in H_2 gas. The cross sections are plotted as a function of the H^- or D^- velocity. The cross sections for negative-ion production are the same for diatomic and triatomic molecular ions within the accuracy of the experiment. For negative-ion production from H_2^+ ions, there is a different velocity dependence than that from H_3^+ or HD_2^+ for velocities

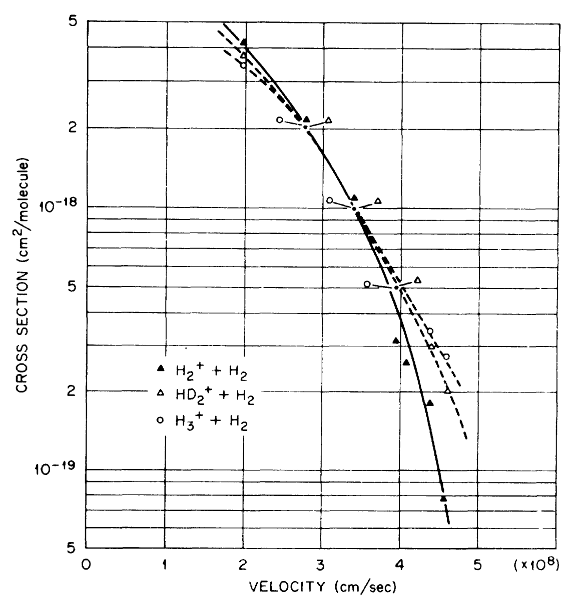

FIG. 3. Cross section for the production of D^- and H^- for HD_2^+, H_3^+, and H_2^+ beams dissociated in H_2.

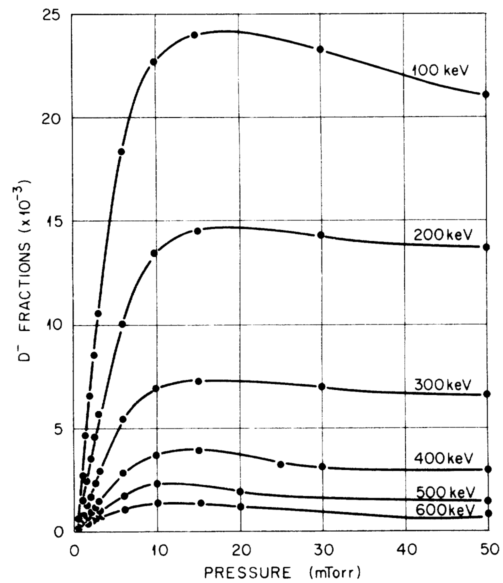

FIG. 4. Production of D^- by the dissociation of HD_2^+ in H_2 from 100 to 600 keV.

greater than 3.3×10^8 cm/sec. This same behavior has been observed for proton production.[5] H^- background prevented measurement of the H^- cross sections and equilibrium fractions from HD_2^+ dissociation. The upper limits of the H^- equilibrium fractions were 0.0032, 0.0024, 0.0018, and 0.0010 for 100-, 200-, 300-, and 400-keV HD_2^+, respectively. An explanation of the lower H^- yield is not

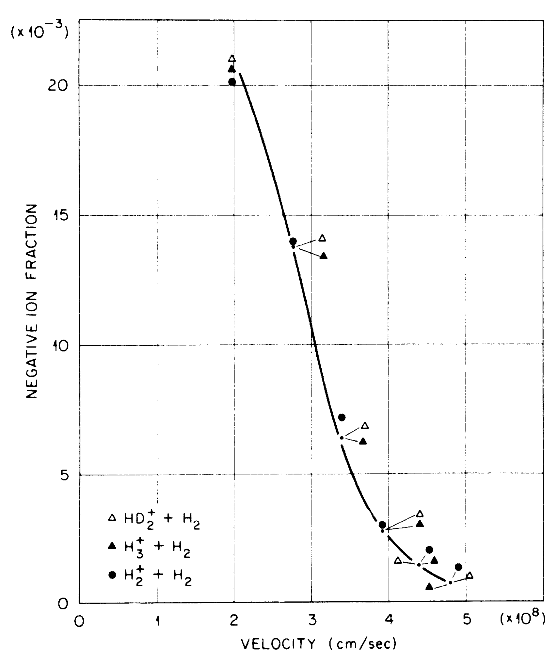

FIG. 5. Equilibrium fractions of D^- and H^- in H_2 as a function of the negative-ion velocity.

available at the present time. The previous measurements of H^- cross sections by Williams for H_2^+ and H_3^+ were at a lower energy or velocity than were the present measurements. Extrapolation of the present values to lower velocities indicates values differing by less than 50% from those reported by Williams,[13] who stated that negative-ion cross sections fluctuated up to 20% with time, whereas the proton cross sections were repeatable with small variations. Williams did a careful investigation of the cross sections as the ion-source parameters were changed, and found the fluctuations to be due to these changes. We find a similar dependence for H^- or D^- cross sections with fluctuations of 15%, depending on ion-source conditions. The negative-ion cross sections reported here are for source conditions adjusted to give maximum cross sections. This illustrates again the necessity of knowing vibrational level populations for molecular ions used in dissociative experiments.

Shown in Fig. 4 are the D^- fractions formed from HD_2^+ at 100–600 keV as the gas target density was increased to 4.8×10^{16} molecules/cm^2. At low nl the fraction is linear, reaches a maximum, and then decreases to an equilibrium value. The peak fraction occurs for a target density between 1×10^{16} and 2×10^{16} molecules/cm^2. The reduction in D^- fraction occurs as the D^- undergoes detachment collisions and the subsequent attachment to form D^- again. Similar curves were obtained of H_2^+ and H_3^+ producing H^-. Graphically, it appears that the D^- fraction for 100-keV incident HD_2^+ is decreasing at target pressures of 50 mTorr. Increasing the target gas pressure to 150 mTorr produced a negligible effect on the equilibrium. The 0.021 D^- equilibrium fraction for 100-keV incident HD_2^+ can

TABLE I. Peak fractions and equilibrium fractions of H^- or D^- produced in collisions of H_2^+, H_3^+, and HD_2^+ in hydrogen gas. [a]

HD_2^+ energy (keV)	100	200	300	400	500	600
D^- peak fraction	2.4	1.5	0.7	0.4	0.2	0.14
D^- equilibrium fraction	2.1	1.4	0.6	0.3	0.1	0.07
H_3^+ energy (keV)	60	120	180	240	300	360
H^- peak fraction	2.3	1.6	0.7	0.3	0.2	0.14
H^- equilibrium fraction	2.0	1.4	0.6	0.3	0.1	0.07
H_2^+ energy (keV)	40	80	120	160	200	240
H^- peak fraction	2.4	1.6	1.1	0.4	0.2	0.10
H^- equilibrium fraction	2.0	1.4	0.7	0.3	0.1	0.07

[a] All fractions are in percent and are expressed in terms of H^- or D^- per incident H or D atom.

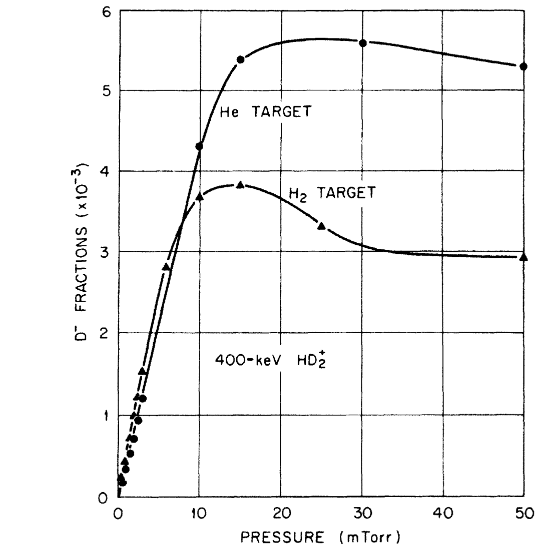

FIG. 6. Comparison of the production of D^- by the dissociation of HD_2^+ in H_2 and in He at 400 keV.

be compared with a 0.019 H^- fraction at the same velocity for 20-keV H^+ gas.[14] The equilibrium fractions of negative ions formed are shown in Fig. 5 for HD_2^+, H_2, and H_3^+ in H_2 gas. The values of all equilibrium and peak fractions are expressed in terms of H^- or D^- per incident H or D atom. For absolute yields the quoted fractions should be multiplied by 2 for H_2^+ and HD_2^+, and by 3 for H_3^+. For equivalent velocities the fraction was independent of the incident species. Both the maximum or peak and equilibrium negative fractions are given in Table I.

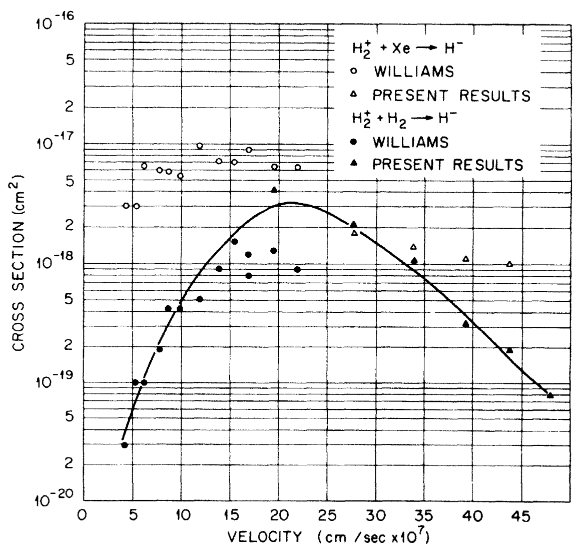

FIG. 7. Comparison of the present results for H^- formation for the reactions $H_2^+ + H_2$ and $H_2^+ + Xe$ with the results obtained by Williams.

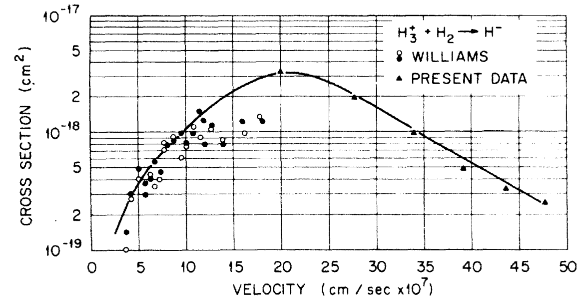

FIG. 8. Present results compared with those of Williams for H^- formation from the $H_3^+ + H_2$ reaction.

In Fig. 6 the D^- fractions obtained from 400-keV HD_2^+ are compared with those obtained with He as a target gas. For helium the cross sections were lower; however, the peak fractions and equilibrium values are approximately 50% greater.

During the course of the measurements we were informed of some results obtained at Grenoble[15] in which it was reported that H^- production was large for H_2^+ collisions in Xe gas at low energies. Williams[13] has also reported cross sections for Xe gas in the velocity range 0.5×10^9 to 2.2×10^9 cm/sec. With Xe as a target gas, no changes were made in the 2.5-mm gas cell exit aperture. If the scattering for negative-ion formation was greater in Xe than in H_2, the exit aperture could have intercepted a fraction of the negative ions. Thus, the cross sections for H^- formation in Xe as shown in Fig. 7 may be low. The xenon results are a factor of 50 higher than for H_2 at 5×10^7 cm/sec and approximately equal at 2×10^8 cm/sec. Also shown in Fig. 7 are the low-energy results of Williams compared with the present results. The solid line is a best fit to the two measurements, which are in good agreement.

Shown in Fig. 8 are the present H^--formation cross sections for H_3^+ colliding with H_2, compared with those obtained by Williams. At velocities of 2×10^8 cm/sec the present results are a factor of 2 larger than those of Williams.

These measured cross sections and equilibrium values indicate that the formation of negative ions from 400–600-keV molecular hydrogen ion dissociation in gases is inefficient, and is not a feasible method to obtain large fluxes of negative ions that will be needed to fuel and heat thermonuclear-type plasmas.

ACKNOWLEDGMENTS

We are grateful for the many stimulating discussions with Professor A. Russek, University of Connecticut, and for his helpful comments.

*Operated by the Union Carbide Corporation for the U. S. Energy Research and Development Administration.

[1]K. H. Purser, IEEE Trans. Nucl. Sci. NS-20, 136 (1973).

[2]G. W. McClure and J. M. Peek, *Dissociation in Heavy Particle Collisions* (Wiley, New York, 1972).

[3]B. Bederson and W. L. Fite, in *Methods of Experimental Physics* (Academic, New York, 1968), Vol. 7.

[4]R. Caudano, thesis (Universite Catholique de Louvain, 1969) (unpublished).

[5]N. V. Fedorenko, V. V. Afrosimov, R. N. Il'In, and D. M. Kaminker, Zh. Eksp. Teor. Fiz. 36, 385 (1959) [Sov. Phys.-JETP 9, 267 (1959)].

[6]J. F. Williams and D. N. F. Dunbar, Phys. Rev. 149, 62 (1966).

[7]F. W. Meyer and L. W. Anderson, Phys. Rev. A 11, 589 (1975).

[8]C. F. Barnett, M. Rankin, and J. A. Ray, in *Proceedings of the Sixth International Conference on Ionization Phenomena in Gases, Paris, 1963*, edited by P. Hubert (SERMA, Paris, 1964), p. 63.

[9]D. R. Sweetman, Proc. R. Soc. A 256, 416 (1960).

[10]J. A. Ray and C. F. Barnett, Report No. ORNL-TM-2818, 1969 (unpublished).

[11]C. F. Barnett and H. B. Gilbody, in *Methods of Experimental Physics*, edited by B. Bederson and W. L. Fite (Academic, New York, 1968), p. 390.

[12]L. H. Toburen, N. Y. Nakai, and R. A. Langley, Phys. Rev. 171, 114 (1968).

[13]J. F. Williams, thesis (Australian National University, 1965) (unpublished).

[14]P. M. Stier and C. F. Barnett, Phys. Rev. 103, 896 (1956).

[15]P. N. Fournier, Université Paris-Sud (private communication).