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ABSTRACT

Experimental electron-capture and loss scross sections of 5- to 70-keV hydrogen atoms and ions in magnesium vapor are reported for the processes

$$H^+ \rightarrow H^0,\ H^0 \rightarrow H^+,\ H^0 \rightarrow H^-, \text{ and } H^- \rightarrow H^0.$$

The cross sections for these processes are compared with measurements by other groups. Results for the $H^+ \rightarrow H^0$ capture process in magnesium are compared with the semiclassical formulation by Bates and Mapleton and with Born-approximation (Brinkman-Kramers) calculations by Hiskes adjusted according to prescriptions by Mapleton and by Nikolaev.

I. INTRODUCTION

At proton energies between about 5 and 30 keV, cross sections for electron capture from metal vapors are much larger than from common gases. This is true of total capture cross sections and for capture into highly excited levels. Capture from magnesium vapor into the level with principal quantum number n = 6 is the subject of a separate paper.[1] Here we report measurements of total cross sections for electron capture and loss by 5- to 70-keV protons in Mg for the following processes:

$$\sigma_{10}: \quad H^+ + Mg \rightarrow H^0 + \cdots \tag{1}$$

$$\sigma_{01}: \quad H^0 + Mg \rightarrow H^+ + \cdots \tag{2}$$

$$\sigma_{0\bar{1}}: \quad H^0 + Mg \rightarrow H^- + \cdots \tag{3}$$

$$\sigma_{\bar{1}0}: \quad H^- + Mg \rightarrow H^0 + \cdots . \tag{4}$$

At present, cross sections for electron capture from heavy atoms are essentially impossible to calculate in the energy range considered here. As a result, there is considerable interest in classical approximations[2] and in semiempirical methods of adjusting results of the relatively easily evaluated Brinkman-Kramers (B-K) approximation.[3,4] Both approaches have given good results for the common gases; we shall see that the adjusted B-K results give reasonably good agreement with magnesium experiments.

II. APPARATUS AND PROCEDURE

The experimental arrangement is shown schematically in Fig. 1. A collimated, momentum-analyzed beam of hydrogen atoms or ions, chopped at a frequency of 10.5 Hz, passed through an oven containing magnesium vapor. The various emerging charge components were separated electrostatically and

the charged fractions were collected in a Faraday cup with magnetic electron suppression. The neutral component was detected with a pyroelectric, phase-sensitive detection system[5] whose calibration was checked with a proton beam at frequent intervals during the experiment. Both signals were amplified and integrated.

The oven is shown schematically in Fig. 2. A commercial 50-watt heating element was press-fit into a hole in a stainless steel cylinder in which a reservoir for the granular magnesium was machined. The oven and thermocouples were surrounded by a three-layer heat shield of dimpled 0.25-mm-thick stainless steel.

A gas-inlet line was provided so that the oven chamber could be used as a conventional gas cell. In this case a capacitance manometer was used to determine the gas pressure. The spacing between the entrance (0.75 mm diam) and exit (1.25 mm diam) collimators was 4.45 cm.

Collimators ahead of the oven constrained the incident proton beam to a maximum possible angular divergence of ±3 mrad. This geometry, together with angular-distribution measurements by Wittkower et al.[6] for protons traversing various gases, indicates that the 1.25-mm-diam exit aperture should transmit essentially all of the reaction products as well as the noninteracting fraction of the incident beam. Similarly, all emerging particles fell within the collectors. This was demonstrated for the charged components and inferred from geometry for the neutral beam.

Chromel-alumel thermocouples were fastened at the top and bottom of the oven. At equilibrium, these thermocouples agreed to within 0.3% in our operating range. Measurements with the bottom thermocouple were used in the magnesium vapor-pressure determinations. The thermocouple

was calibrated in situ in the following way:[7] The oven was loaded with pure zinc, which melts within the temperature range used in our experiments. The oven temperature was slowly raised and lowered past the melting point of zinc (419.4°C) with constant power in the oven heater. A plateau in an oven temperature vs time plot allowed a calibration at 419.4°C with an uncertainty of approximately ±2°C. A similar calibration was made with metallic lead at 327.5°C.

After the oven was loaded with magnesium it was outgassed at a high temperature for 12 or more hours before data were taken. After the oven had been heated for approximately an hour, cross sections did not vary until the magnesium was almost exhausted.

For the σ_{01} and $\sigma_{0\bar{1}}$ measurements, a neutral hydrogen beam could be produced by allowing the protons to capture electrons from helium gas introduced upbeam of the first collimator; in this case the ions remaining in the beam were swept out by a magnet ahead of the oven. By introducing helium in the region between the accelerator and the momentum-analyzing magnet, a small current of H^- ions could be produced by double electron capture; the momentum-analyzing magnet could then be adjusted to transmit only this charge state.

Measurements at each energy were made for at least five different target thicknesses. The maximum target thickness for each process was determined by the competition of secondary processes, as discussed in the appendix, and never exceeded 3×10^{14} cm^{-2}. Most of the measurements reported here were taken during a period of 1 month, but σ_{10} measurements have been repeated at frequent intervals during an 8-month period; they agree among themselves to within 10%.

III. ANALYSIS AND RESULTS

Although the cross sections, σ_{if}, for collisions in which a hydrogen ion (or atom) of charge i either captures or loses an electron and is left with charge f were determined by passing a beam initially in charge state i through fairly thin targets, the analysis requires enough comment that we defer it to the appendix.

The measured values of σ_{10}, σ_{01}, $\sigma_{\bar{1}0}$, and $\sigma_{0\bar{1}}$ are presented in Table I. The values of magnesium vapor pressure as a function of oven temperature that were used to obtain these cross sections were taken from the supplement[8] to the book by Hultgren et al.[9] The standard errors shown in Table I are based on estimates of uncertainties in the effective length of the target cell, the temperatures used in calculating vapor pressures, approximations and constants used in the data analysis, and on internal consistency. They do not include the uncertainty in the magnesium vapor pressure, which is apparently quite difficult to determine. The evaluated data in Ref. 8 have an assigned uncertainty (95% confidence level) equivalent to about ±20% in the vapor pressure.[10]

Our values of σ_{10} are plotted in Fig. 3, together with the other data of which we are aware.[11-13] The values of Barnett et al.[13] in the energy range 15 to 50 keV are based on the same vapor-pressure data that we used. The Futch and Moses 4- to 45-keV values[12] were based on Ref. 9, and in Fig. 3 have been multiplied by 0.81 to take account of the new thermodynamic evaluation given in Ref. 8. According to Il'in et al.,[14] their vapor pressures were not accurately determined, and consequently only the shape of the curve should be considered. Nevertheless, their data are in fairly good quantitative agreement with the other experiments.

Table I. Experimental cross sections in units of 10^{-16} cm^2/atom.

Energy (keV)	σ_{10} ±15%	σ_{01} ±15%	$\sigma_{\bar{1}0}$ ±25%	$\sigma_{0\bar{1}}$ ±15%
5	15.2	0.313		1.06
7.5	22.5			
10	15.6	0.609	24	0.659
15	10.7	1.05		0.375
20	6.16	1.68		0.212
25	3.94	2.07		0.112
30	2.22	2.34		0.0749
35	1.42	2.61	12	0.0500
40	0.83	2.54		0.0346
45		2.71		0.0258
50	0.408	2.75		0.0187
60	0.278	2.92		0.0141
70	0.213	3.23	13	0.0117

Excluding uncertainties in the tabulated vapor pressures, we estimated probable errors of about ±15%. Our data are consistently about 30% below the values of Barnett et al.,[13] who also assign errors of ±15% to their results. Il'in et al. and Futch and Moses did not report estimated uncertainties.

Also shown in Fig. 3 are the results of various theoretical models for the capture cross section. The curve labeled B&M(Cl) obtained[15] from the classical expression of Bates and Mapleton[2] lies well above the experimental points at the higher energies, where the theory should be most valid. The other curves are based on Brinkman-Kramers calculations by Hiskes.[16] Although the Brinkman-Kramers approach is known to overestimate cross sections at low energies, Hiskes has shown that ratios of various quantities calculated in this way quite accurately agree with reality.[17] He has consequently calculated cross sections for capture into individual quantum states (n = 1 to 11) for many of the elements. His calculated total cross sections for capture of the $3s^2$, $2p^6$, and $2s^2$ electrons of magnesium, using the best available wave functions[18] in the prior and the post approximations, are given by curves H(Pr) and H(Po).

Nikolaev has shown[19] that an empirical expression can be obtained that quite accurately adjusts Brinkman-Kramers calculations (using the post interaction and hydrogen-like wave functions with $Z = Z_{eff}/n_{eff}$ determined by Slater's method) to agree with experiment in the case of common gases. To allow comparison with the present experimental results Hiskes has evaluated this form of the B-K cross section [curve N(H)][16] and we have applied Nikolaev's expression to curve N(H) to obtain curve N.

Mapleton has suggested another approach for adjusting Brinkman-Kramers calculations: The Jackson-Schiff (J-S) form of the Born approxi-

mation is known to give approximately the correct results for hydrogen. Mapleton has shown,[20,3] that quite good agreement with experiment is obtained in the cases of nitrogen, oxygen, and argon if the (J-S)/(B-K) ratio, evaluated for capture into H(1s) from hydrogen, is used to adjust the B-K result for the target of interest. In this spirit, we used these ratios[15,21] to multiply the average of Hiskes' prior and post B-K calculations and obtained curve M of Fig. 3.

The other measured cross sections are shown in Fig. 4. As in Fig. 3, the original data of Futch and Moses have been multiplied by 0.81 before plotting, to adjust them to the magnesium vapor-pressure data of Ref. 8. The lines through our points are drawn in to guide the eye, and have no other significance.

IV. DISCUSSION

As can be seen from Figs. 3 and 4, measurements at various laboratories are in fair agreement. For those cases where error estimates are quoted, i.e., this experiment and Ref. 13, the σ_{01} data agree within the uncertainties, but the values of σ_{10} and $\sigma_{0\bar{1}}$ are separated by roughly twice the uncertainty of an individual measurement. There is a suggestion of a systematic error in one or both of these measurements, and the magnesium vapor-pressure determination might seem most suspect. In our operating range, approximately 320 to 420°C, the vapor pressure of magnesium changes about 2% per degree centigrade. If the thermocouple were not located at the point of lowest temperature within the oven, the vapor pressure would be overestimated and the calculated cross sections would be too low. We have no reason to believe that we overestimated the controlling temperature, since heat is introduced at the top of our oven,

the thermocouple is at the bottom, and no magnesium condenses on the entrance and exit apertures during normal operation. It is also easy to show that escape of vapor through the orifices cannot affect the density appreciably. In spite of the difficulties in determining the magnesium vapor pressure, the disagreement among the different laboratories is not much worse than it is for ordinary gas targets.

The classical and Brinkman-Kramers formulations for σ_{10} are both most applicable at high energies. Unfortunately, the agreement between the classical theory and experiment at the upper end of our energy range is apparently worse for magnesium than it is for gases like neon and argon. The Brinkman-Kramers curves increasingly overestimate the cross sections as the energy decreases, but show a maximum at about the right energy. The prescription of Mapleton adjusts the B-K results for magnesium in magnitude and shape so that they are in quite good agreement with experiment. (The agreement is better than it is for low-energy protons in N_2, O_2, and Ar.) The agreement between experiment and the curve (N) based on B-K calculations with hydrogen-like wave functions and Nikolaev's empirical expression is not as good as that obtained by Nikolaev for the common gases. (Nikolaev got 20 to 25% agreement in H_2, He, Li, N_2, Ne, Ar, and Kr for 20-keV to 13-MeV protons.)

In conclusion, although it is not yet possible to predict total cross sections to the accuracy with which experiments can be performed, the prescriptions of either Nikolaev or of Mapleton improve the Brinkman-Kramers results significantly. Using either of them, it would appear to be possible to predict σ_{10} for protons in many gaseous materials to within a factor of two or three for energies from perhaps 5 keV up to the relativistic region.

ACKNOWLEDGMENTS

We take pleasure in thanking Dr. C. M. Van Atta for his support of this research, and Drs. J. R. Hiskes and R. A. Mapleton for theoretical advice and for performing the Brinkman-Kramers and classical calculations, respectively. Professors L. Brewer, R. R. Hultgren, and D. Olander made valuable comments on evaluating magnesium vapor-pressure data.

APPENDIX

The population of a charge state k, expressed as a fraction F_k of the total beam, is related to the target thickness $\pi \equiv$ (target density) x (path length through target), by the coupled equations

$$\frac{dF_k}{d\pi} = \sum_{j \neq k} (F_j \sigma_{jk} - F_k \sigma_{kj}), \qquad j,k = 1, 0, -1. \tag{A1}$$

The complete solutions to these equations for various initial conditions have been tabulated by Allison and Garcia-Munoz,[22] but approximate solutions are satisfactory for our purposes. To determine the magnitude of the various cross sections, we have used the solutions to first order in π, which for a beam initially in charge state i are

$$F_f = \pi \sigma_{if}. \tag{A2}$$

From these we determined which secondary processes were important, even at small values of π, and made appropriate corrections to the first-order solutions.

A. Determination of σ_{10} and σ_{01}

For energies greater than 10 keV the production of H^- by two-electron capture $(\sigma_{1\bar{1}})$[12] or one-electron capture $(\sigma_{0\bar{1}})$ is small compared to the

processes described in Eqs. (1) and (2); consequently, we can determine σ_{10} and σ_{01} by considering only a two-level system consisting of H^+ and H. For a two-level system the exact solution to Eqs. (A1) for F_1, when the incident beam consists only of H atoms, is

$$F_1 = \frac{\sigma_{01}}{\sigma_{01} + \sigma_{10}} \left\{ 1 - \exp\left[-\pi(\sigma_{01} + \sigma_{10})\right] \right\}. \tag{A3}$$

If we expand the exponential in powers of $\pi\sigma_{01}$ and solve Eq. (A3) for $\pi\sigma_{01}$, we get, to first order in $\pi\sigma_{01}$,

$$\pi\sigma_{01} = \frac{\left[F_1 - 1 + \exp(-\pi\sigma_{10})\right] + \left\{\left[F_1 - 1 + \exp(-\pi\sigma_{10})\right]^2 + 4\pi\sigma_{10}F_1 \exp(-\pi\sigma_{10})\right\}^{\frac{1}{2}}}{2 \exp(-\pi\sigma_{10})}. \tag{A4}$$

By symmetry it is clear that the solution for σ_{10}, when the incident beam consists only of H^+, is obtained by permutation of the indices 0 and 1.

The cross sections σ_{10} and σ_{01} were obtained by an iteration procedure; for example, in the case of electron capture our first estimate of σ_{10} was obtained from a linear fit to $F_0(\pi)$ vs π. This estimate of σ_{10} and the measured $F_1(\pi)$ were used in Eq. (A4), and σ_{01} was obtained from a least-squares fit to the various $\pi\sigma_{01}$ results. This value of σ_{01} and the measured $F_0(\pi)$ were then used in the permuted form of Eq. (A4) to obtain a least-squares weighted value of σ_{10}. Our criterion for convergence was that the results of successive iterations should differ by less than 5%. This was achieved in all cases after the second interation cycle.

At our two lowest energies, $\sigma_{0\bar{1}}$ and $\sigma_{\bar{1}0}$ are comparable to or exceed

σ_{01}; hence the production of H^- is no longer negligible, and we must determine whether or not this invalidates our two-level calculational model. A large H^- fraction might affect the determination of σ_{01} because of proton production by the two-step process $H^0 \rightarrow H^- \rightarrow H^+$. For small enough values of π, the proton fraction from this process is equal to $\frac{1}{2}\pi^2[(\sigma_{0\bar{1}}\sigma_{\bar{1}1}^2)/(\sigma_{\bar{1}1} + \sigma_{\bar{1}0})]$. Although the cross section for two-electron loss ($\sigma_{\bar{1}1}$) is not known, it must be smaller than that for one-electron loss ($\sigma_{\bar{1}0}$). From Allison's compilation for gas targets we find that the ratio $\sigma_{\bar{1}0}/\sigma_{\bar{1}1}$ is always greater than five in the 5- to 10-keV range; we assume that five is also a minimum value for this ratio in Mg. From our measured $\sigma_{\bar{1}0}$ (see below) we determined the contribution to the two-step process as a function of π. Our measurements were restricted to target thicknesses for which this contribution was less than ~ 2%, so we used the two-level system for our analysis.

B. Determination of $\sigma_{\bar{1}0}$

The cross section $\sigma_{\bar{1}0}$ is larger than any of the others, and no corrections to Eq. (A2) for secondary processes were necessary. However, our method of producing H^- was very inefficient. As a result the measurement of $F_0(\pi)$ was complicated by detector noise, and we limited ourselves to establishing the magnitude of this cross section at three points of our energy range.

C. Determination of $\sigma_{0\bar{1}}$

For the determination of $\sigma_{0\bar{1}}$ we again argue that $\sigma_{\bar{1}1}$ must be less than $\sigma_{\bar{1}0}$. Thus the main competition is between $H^0 \rightarrow H^-$ and $H^- \rightarrow H^0$ [Eqs. (3) and (4)], and Eq. (A4) (with the index 1 replaced by -1) can be used in the analysis. Since Eq. (A4) does not take into account the attenua-

tion of the H^0 beam due to electron loss, $H^0 \rightarrow H^+$ [Eq. (2)], we limited our target thicknesses so that the error introduced by this process was less than 5%. The $\sigma_{\bar{1}0}$ used in the appropriate form of Eq. (A4) was interpolated from our three measured values, and $\sigma_{0\bar{1}}$ was obtained from a least-squares fit to the various $\pi\sigma_{0\bar{1}}$. These corrections for $\sigma_{\bar{1}0}$ changed our first estimate based on Eq. (A2), typically by 30% but by as much as 40% for the worst case (5 keV). Since our estimated error in $\sigma_{\bar{1}0}$ is ±25%, this correction introduces, at worst, an uncertainty of ±10%.

FOOTNOTES AND REFERENCES

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about 20% lower for temperatures in our operating range. Approximate values quoted in the Handbook of Chemistry and Physics, 48th Edition (The Chemical Rubber Company, Cleveland, Ohio, 1967-68), p. D-112, are about 40% below those of Ref. 8.

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FIGURE LEGENDS

Fig. 1. The experimental arrangement. The Faraday cup was used alternatively for H^+ and H^- measurements.

Fig. 2. The magnesium vapor cell.

Fig. 3. The total electron-capture cross section, σ_{10}, for the process $H^+ + Mg \rightarrow H^0 + \cdots$ vs proton energy. Experimental: ■, present work; o, Ref. 11; Δ, Ref. 12; ◇, Ref. 13. Theoretical: H(Pr) and H(Po) are Brinkman-Kramers prior and post calculations by Hiskes; N(H) is a B-K calculation by Hiskes using hydrogen-like wave functions (see text). B&M(Cl) is a classical calculation, Refs. 2 and 15; curve N was obtained by adjusting N(H) with Nikolaev's semiempirical prescription of Ref. 4; curve M was obtained by multiplying the average of H(Pr) and H(Po) by ratios suggested by Mapleton, Refs. 3 and 15.

Fig. 4. Experimental total charge-exchange cross sections of protons in magnesium vapor for the processes $H^- + Mg \rightarrow H^0 + \cdots$ $(\sigma_{\bar{1}0})$; $H^0 + Mg \rightarrow H^+ + \cdots$ (σ_{01}); $H^0 + Mg \rightarrow H^- + \cdots$ $(\sigma_{0\bar{1}})$. ■, present work; o, Ref. 11; Δ, Ref. 12; ◇, Ref. 13.

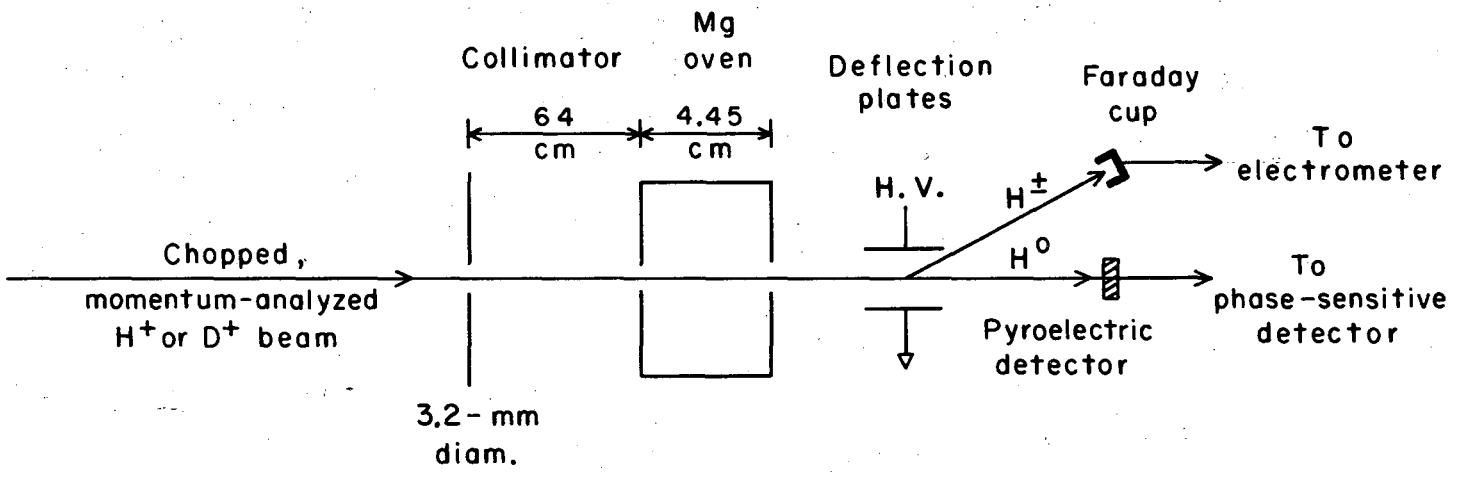
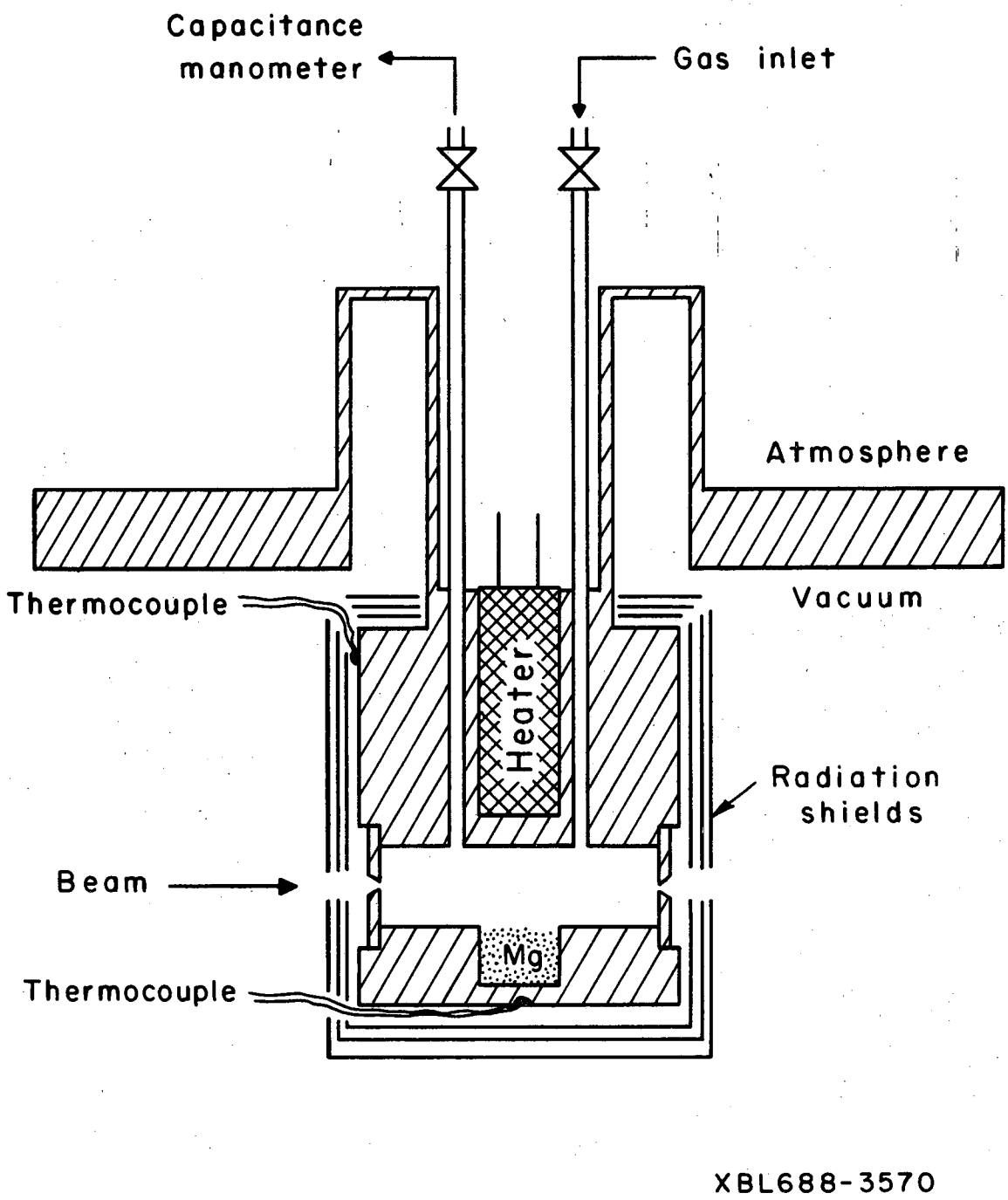

Collimator
Mg oven
Deflection plates
Faraday cup
64 cm
4.45 cm
To electrometer
H.V.
$H^{\pm}$
Chopped, momentum-analyzed H^+ or D^+ beam
H^0
To phase-sensitive detector
Pyroelectric detector
3.2-mm diam.
XBL688-3569

Fig. 1.


Capacitance manometer
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Atmosphere
Vacuum
Thermocouple
Heater
Radiation shields
Beam
Mg
Thermocouple
XBL688-3570

Fig. 2.

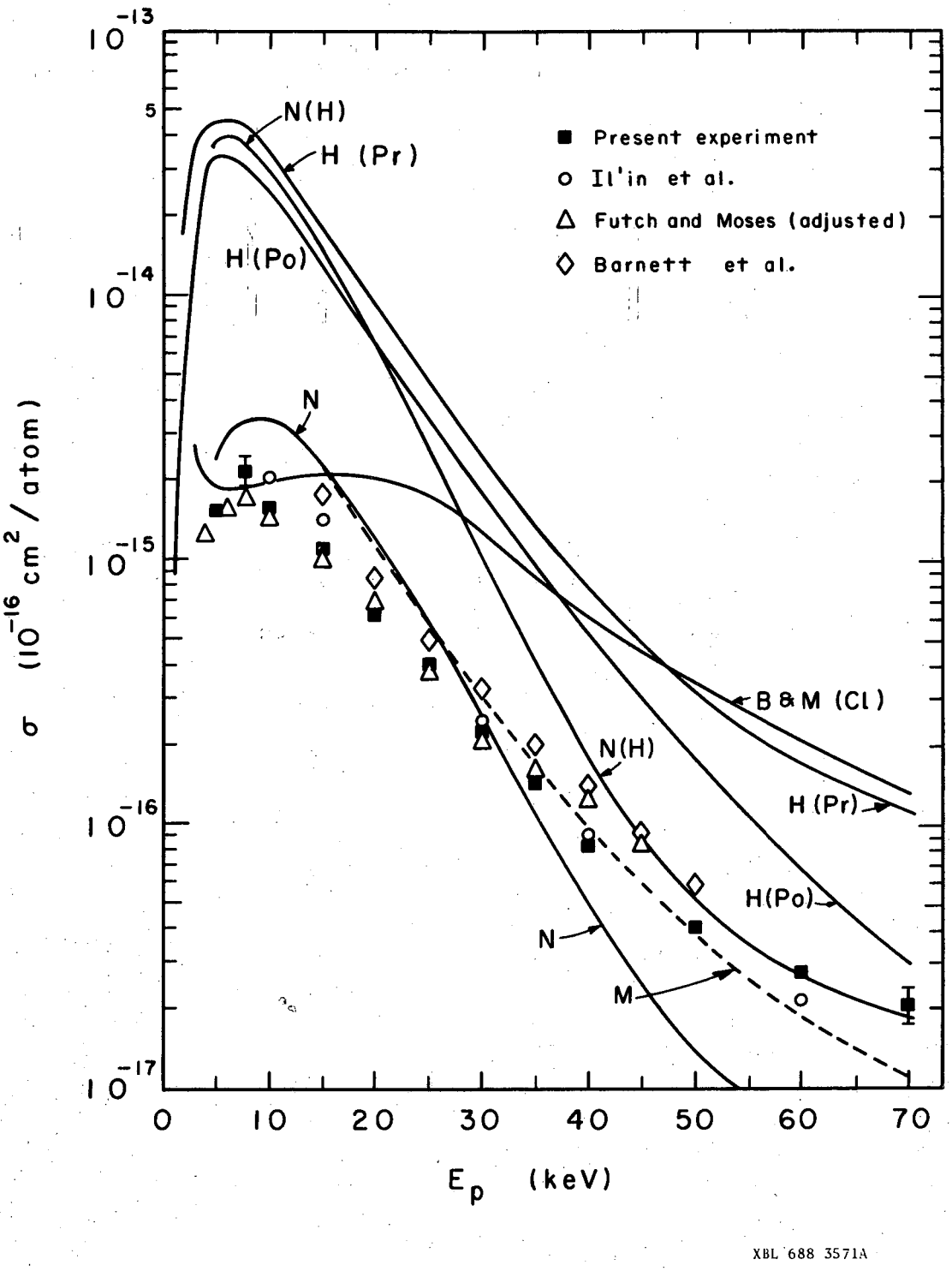
Present experiment
Il'in et al.
Futch and Moses (adjusted)
Barnett et al.
N(H)
H (Pr)
H(Po)
N
B & M (Cl)
M
σ (10⁻¹⁶ cm² / atom)
E_p (keV)
XBL 688 3571A

Fig. 3.

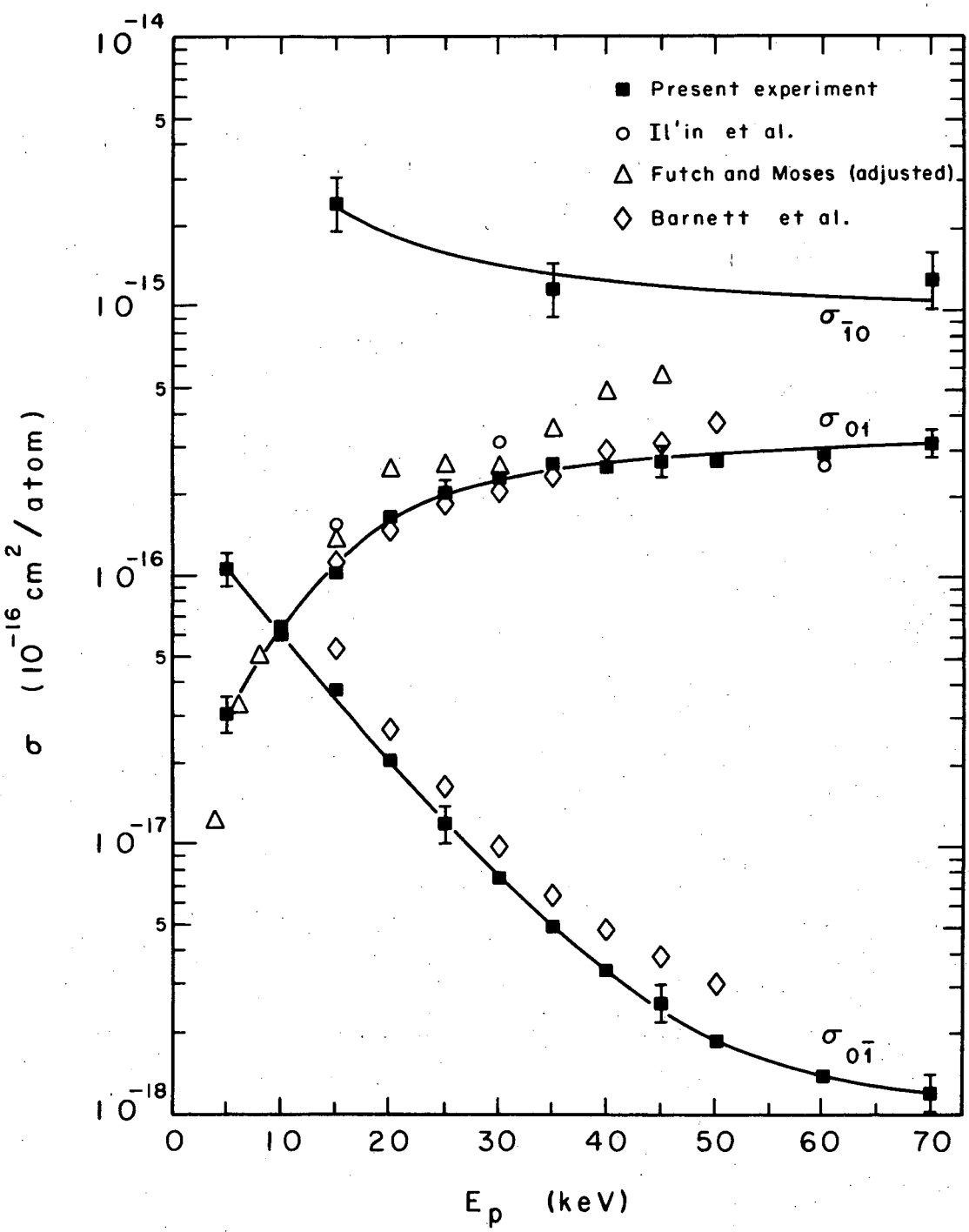

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Fig. 4.