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ABSTRACT

The cross sections, σ_{21} and σ_{20}, for single and double electron capture of $^3He^{++}$ ions with energies between 7.7 and 166 keV have been measured in thin targets of N_2. Our results join quite smoothly to higher-energy measurements by others.

INTRODUCTION

There is a considerable attempt at present to try to understand electron capture by ions as they traverse various gases. For the simplest case, the capture of one electron by a proton, cross-section measurements are available for energies from a few hundred eV to tens of MeV for most of the common gases. However, the problem of electron capture by helium nuclei has not been studied so extensively. Published measurements of total cross sections are available mainly for energies greater than 38 keV per nucleon,[1-5] and we know only of one low-energy experiment: single electron capture by He^{++} in He by Hertel and Koski for the range 0.25 to 2 keV per nucleon.[6] We report here cross-section measurements for two-electron capture, σ_{20}, and one-electron capture, σ_{21}, by doubly charged helium ions passing through N_2 for the energy range 2.6 to 55 keV per nucleon.

APPARATUS AND PROCEDURE

The apparatus is shown schematically in Fig. 1. To avoid problems due to contamination of the primary beam by hydrogen isotopes, we used $^3He^{++}$ ions.[7] A momentum-analyzed beam of $^3He^{++}$ ions passed through a 9.5-cm-long gas cell with entrance and exit apertures of 1.6 and 3.2 mm diam respectively. The incident He^{++} beam was collimated ahead of the gas cell so that the maximum possible angular divergence was ±3.5 mrad. Angular distribution measurements by Wittkower et al.[8] for He^{+} in various gases indicate that the 3.2-mm-diam exit aperture should transmit essentially all of the reaction products, as well as the noninteracting fraction of the incident beam. As an experimental check we reduced the exit aperture to 2.4 mm for a set of measurements at a low energy, 15.4

keV. and within the experimental uncertainties obtained the same values of σ_{21} and σ_{20} as were measured with the larger aperture.

Since the negative-ion fraction of the emerging beam was negligible, we measured only the He^{++}, He^{+}, and He^{0} components. The charged components were analyzed electrostatically and simultaneously stopped in Faraday cups that had small transverse magnetic fields for secondary-electron suppression. The He^{0} atoms were detected by a CsI(Tl) scintillation crystal attached to a photomultiplier. The crystal was covered with a thin layer of gold to prevent accumulation of surface charge and was periodically calibrated by directing an ion beam of the same energy and known intensity onto it. All three signals were amplified and integrated simultaneously.

The pressure in the gas cell was monitored with a capacitance manometer whose calibration was checked during the experiment in the range 100 to 500 mtorr with an oil manometer to an accuracy of 1%. Linearity of the capacitance manometer was assumed[9] for the lower target pressures used in the experiment, where a continual cross-check with a VG-1A ionization gauge was made. Pressures in the drift sections were less than 2×10^{-6} torr.

At each energy approximately ten different measurements were made at various pressures, from background ($< 10^{-5}$ torr) to about 2×10^{-3} torr.

DATA REDUCTION

The cross sections were determined by observing the growth of the fractions of the total beam that were in the charge states 0 and +1, as gas was introduced into the target cell. For our analysis we assume that the incident beam is entirely in the charge state +2.

Let σ_{if} be the cross section per target particle for the reaction that changes a beam particle from charge state i to charge state f; let F_i be the measured fraction of beam in charge state i; and let $\pi \equiv nl$ be the target "thickness," where n is the number density and l is the length of the target. Then the three charge components are described by

$$\frac{dF_0}{d\pi} = - F_0(\sigma_{01} + \sigma_{02}) + F_1\sigma_{10} + F_2\sigma_{20}, \tag{1a}$$

$$\frac{dF_1}{d\pi} = - F_1(\sigma_{10} + \sigma_{12}) + F_0\sigma_{01} + F_2\sigma_{21}, \tag{1b}$$

and

$$F_0 + F_1 + F_2 = 1. \tag{1c}$$

The complete solutions to this set of equations have been published by Allison.[1,10] We used the Taylor expansions of these solutions to second order:[11]

$$F_0/F_2 = \pi\sigma_{20} + \frac{1}{2}\pi^2(\sigma_{21}\sigma_{10} + \sigma_{20}\sigma_{21} + \sigma_{20}{}^2 - \sigma_{20}\sigma_{01} - \sigma_{20}\sigma_{02}). \tag{2}$$

The symmetry between indices 0 and 1 is evident from Eqs. (1a) and (1b), hence we show only one of the solutions. The cross sections σ_{02} and σ_{12} are much smaller than the other relevant cross sections in the energy range we are considering,[10] and we neglect terms involving these cross sections. We can further simplify Eq. (2) by noting that, to first order, we have $\pi\sigma_{2i} = F_i/F_2$. Substituting this expression in each of the second-order terms of Eq. (2), using Eq. (1c), and rearranging terms we get

$$\pi\sigma_{20} = \frac{F_0 - (F_1/2)\pi\sigma_{10} + (F_0/2)\pi\sigma_{01}}{1 - \frac{1}{2}(F_1 + F_0)} \tag{3a}$$

and similarly,

$$\pi\sigma_{21} = \frac{F_1 - (F_0/2)\pi\sigma_{01} + (F_1/2)\pi\sigma_{10}}{1 - \frac{1}{2}(F_1 + F_0)} . \qquad (3b)$$

These equations yield the desired cross sections σ_{21} and σ_{20} in terms of the measured fractions F_i and the known cross sections σ_{10} and σ_{01}.[12] (The pressure-dependent terms in the numerator correct for the two-step processes in which $He^{++} \rightarrow He^{+} \rightarrow He^{0}$ or $He^{++} \rightarrow H^{0} \rightarrow He^{+}$; the denominator corrects for the attenuation of the primary beam.) A sample plot of the uncorrected fractions F_0 and F_1, as well as $\pi\sigma_{20}$ and $\pi\sigma_{21}$ obtained from Eq. (3) is shown in Fig. 2. We see that the calculated values of $\pi\sigma_{20}$ and $\pi\sigma_{21}$ vary linearly with pressure, as they should. A least-squares fit to the $\pi\sigma$ points was used to determine the cross sections listed in Table I.

The second-order Taylor expansion used in this derivation is accurate to 5% if the He^{++} attenuation is less than 40%. The 40% attenuation was the criterion for determining an upper limit for the pressures used for measurements at a given energy.

The σ_{10} and σ_{01} corrections were quite large at the highest pressures used in the analyses. The capture correction amounted to 15% in F_1 and 25% in F_0 in the worst case (7.7-keV $^3He^{++}$). Propagation of a 10% uncertainty[12] in the σ_{10} cross section through our analysis turns out to give a 1% uncertainty in σ_{21} and a 2% uncertainty in σ_{20}.

The σ_{01} corrections are largest at high energies; however, they never exceeded 2% in F_1 and 11% in F_0 at the highest pressure for the worst case (166-keV $^3He^{++}$). Allison,[1] Barnett and Stier,[12] and more recently Wittkower et al.[13] have pointed out that σ_{01} measurements are ambiguous because the content of metastable helium atoms in any beam is strongly dependent on the type and thickness of the neutralizer. We find that an uncertainty in σ_{01} of 100% would, at worst, change our value of σ_{20} by 9% and σ_{21} by 1.5%.

The uncertainties in our results are estimated to be about ±10% for σ_{21} and ±15% for σ_{20}. These uncertainties are compounded from a 5% uncertainty for the measurements of the charged components, 10% for the neutral component, 5% for the target thickness, 5% for the σ_{10}, σ_{01}, and background corrections, and 5% for the approximations made in solving Eq. (1).

RESULTS AND DISCUSSION

The results for σ_{21} and σ_{20} are listed in Table. I. For comparison with other measurements our results are also shown in Fig. 3, where we have chosen the abscissa to be the energy of ^4He ions of the same velocity as the ^3He ions. The measurement of σ_{21} at 7.2 keV (triangle) was for a $^4He^{++}$ primary beam; the corresponding double-capture measurement was considered unreliable due to the possible H_2^+ contamination mentioned previously. Our results join quite smoothly to those of Allison,[1] Pivovar et al.,[2] and Nikolaev et al.[3,4] Allison's results (in air) were deduced from thick-target, equilibrium-fraction measurements. If the problem of metastable atoms is serious, one might expect to see a difference between the cross sections derived from a thick target and those derived from thin-target measurements. Therefore it is encouraging to see such good agreement between the two methods.

Both cross sections have maxima somewhat below 100 keV. The σ_{21} curve is apparently going toward zero at low energies, as would be expected for nonresonant single electron capture. It is not clear from our data what the low-energy trend of the double electron-capture cross section may be.

We know of no theoretical calculations for double electron capture by He^{++} in nitrogen, but Janev recently has reported a method for calculating this type of quantity,[14] and has shown good agreement with experiment (over a limited energy range) for reactions such as $H^+ \rightarrow H^-$ in He and Ne, and $Kr^{++} \rightarrow Kr$ in Ar. Preliminary evaluation of his expression for the present N_2 case indicates that σ_{20} increases with increasing energy much more rapidly than our experimental results. There is no indication of a maximum at energies below 200 keV, the upper limit of validity of the model for this reaction.

Calculations of one-electron capture are shown in Fig. 4 together with the experimental results. The assumptions on which the σ_{21} calculations are based imply interactions at higher energies than were used in this experiment. Nevertheless (as is customary in problems of this type) we present calculations below the obvious range of validity in the hope that they may be semiquantitatively correct. The solid line summarizes the various measurements shown in Fig. 3. The results of the classical model of Bates and Mapleton are indicated by the dashed line labeled B-M.[15] The validity of this model is restricted to $^4He^{++}$ energies >> 400 keV,[16] and the agreement with the measurements of Pivovar et al. and Nikolaev et al. at high energies is quite good. In the low-energy regime of our experiment the cross section is overestimated by an order of magnitude and continues to increase at very low energies, but it does have a secondary maximum near the energy of the experimentally observed maximum. A refinement of the classical model, in which a Hartree-Fock-Slater description of the target replaces the Thomas-Fermi description of Ref. 15, has also been developed by Bates and Mapleton.[17] Mapleton informs us that this method when applied to nitrogen gives results in somewhat better agreement with our experiment.[16]

We have also evaluated σ_{21} for the Brinkman-Kramers model (first Born approximation considering only the interaction of the incoming nucleus and the electrons of the target). Brinkman and Kramers[18] derived a formula for the capture of s electrons from a hydrogenic target, and suggested that a value for the effective nuclear charge of $Z_{eff} = 2$ or 2.4 (based on consideration of the ionization potential of N) be used to evaluate capture of the 2s electrons of nitrogen. An alternative is to follow the prescription of Slater[19] and use $Z_{eff} = 3.9$. We show results for both $Z_{eff} = 2.4$ and 3.9 to illustrate the sensitivity of σ_{21} to the choice of Z_{eff}. To calculate the capture of 2p electrons, we have evaluated the Brinkman-Kramers formula derived by Bates and Dalgarno;[20] again we show results for both $Z_{eff} = 2.4$ and 3.9 for the capture of 2p electrons. The results, which were multiplied by 2 and 3 respectively to account for the contribution of each electron, are shown in Fig. 4; the dashed lines are for capture of 2s electrons, the broken lines for 2p electrons. It is clear that in this energy range the Brinkman-Kramers results are extremely sensitive to the choice of Z_{eff}.

It should be noted that the calculations are made for atomic nitrogen and have been multiplied by two for comparison with the results of the experiment, for which molecular nitrogen was used. The measurements and the Bates-Mapleton results include capture into all excited states, whereas the Brinkman-Kramers formulas were evaluated only for capture into the 1s state of He^+.

ACKNOWLEDGMENTS

We wish to thank Dr. C. M. Van Atta for his support of this research, Dr. R. A. Mapleton for providing us with numerical solutions for the classical model described in Ref. 15, and Dr. M. H. Mittleman for helpful discussions regarding the theoretical aspects of this problem.

Table I. Measured cross sections for one-electron (σ_{21}) and two-electron (σ_{20}) capture by doubly ionized helium ions in N_2 (in units of 10^{-16} cm^2 per molecule).

Energy (keV) $^4He^{++}$	$^3He^{++}$	σ_{21} (±10%)	σ_{20} (±15%)
7.2		4.1	---
	7.7	5.6	2.6
	15	9.2	2.6
	24	12.	3.0
	42	14.	3.8
	64	12.	3.1
	118	11.	2.1
	166	8.3	1.4

FOOTNOTES AND REFERENCES

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

1. S. K. Allison, Phys. Rev. 109, 76 (1958). These measurements were actually made in air, but the measurements of Barnett and Stier (Ref. 12) show that equilibrium fractions in nitrogen and oxygen are quite similar. Therefore, it seems reasonable to compare σ_{21} and σ_{20} for air and nitrogen.

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FIGURE LEGENDS

Fig. 1. Experimental arrangement.

Fig. 2. The results for 15-keV $^3He^{++}$ in N_2 as a function of target pressure. The circles are the measured charge fractions F_1 and F_0. The triangles are the values of $\pi\sigma_{21}$ and $\pi\sigma_{20}$ obtained from Eq. (3) with these charge fractions. The cross sections listed in Table I were obtained from a least-squares fit of the $\pi\sigma$ points (solid lines).

Fig. 3. Results of cross-section measurements for capture of one (σ_{21}) and two (σ_{20}) electrons by helium nuclei in N_2. ▲, ●, o this paper; ◆ Rutherford for air (Ref. 5); □ Nikolaev et al. (Ref. 4); ▣ Nikolaev et al. (Ref. 3); the line marked A presents the results of Allison for air (Ref. 1); the line marked P, Pivovar et al. (Ref. 2).

Fig. 4. Comparison of theory and experiment for σ_{21} in N_2. The solid line summarizes the various experimental results shown in Fig. 3 and the points are the results of this paper. The theoretical predictions are: — — — the Bates and Mapleton classical model (Ref. 15); - - - the Brinkman-Kramers model for capture of the 2s electrons of nitrogen into the 1s state of He^+ (Ref. 18); and — · — the Brinkman-Kramers model for capture of 2p electrons into the 1s state of He^+ (Ref. 20). The Brinkman-Kramers curves are labeled with the values of Z_{eff} used in the calculation.

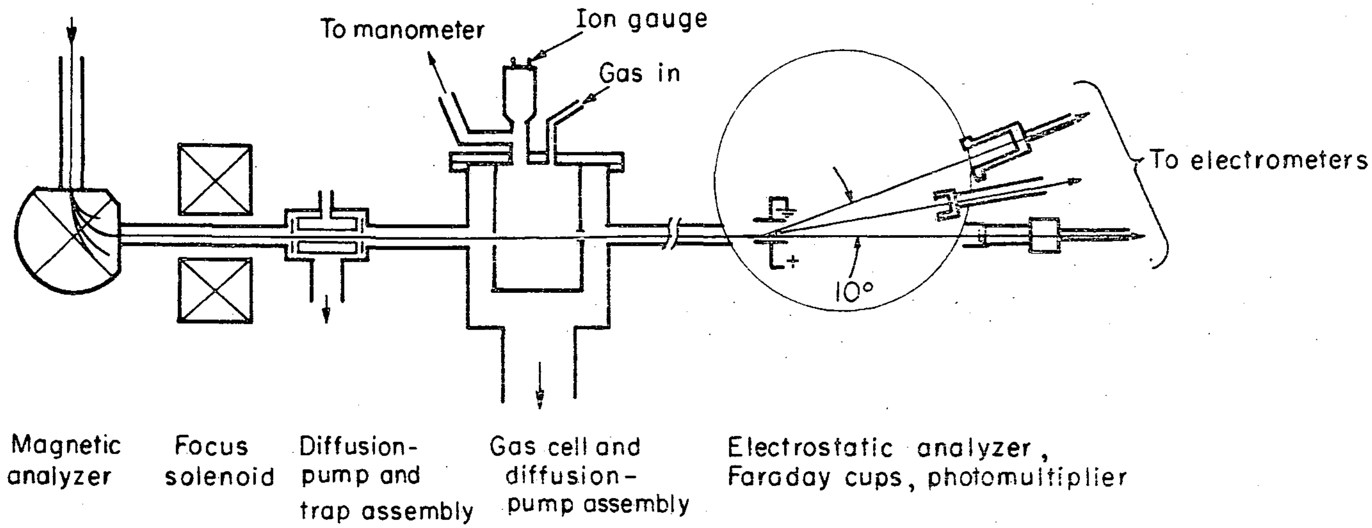

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Fig. 1

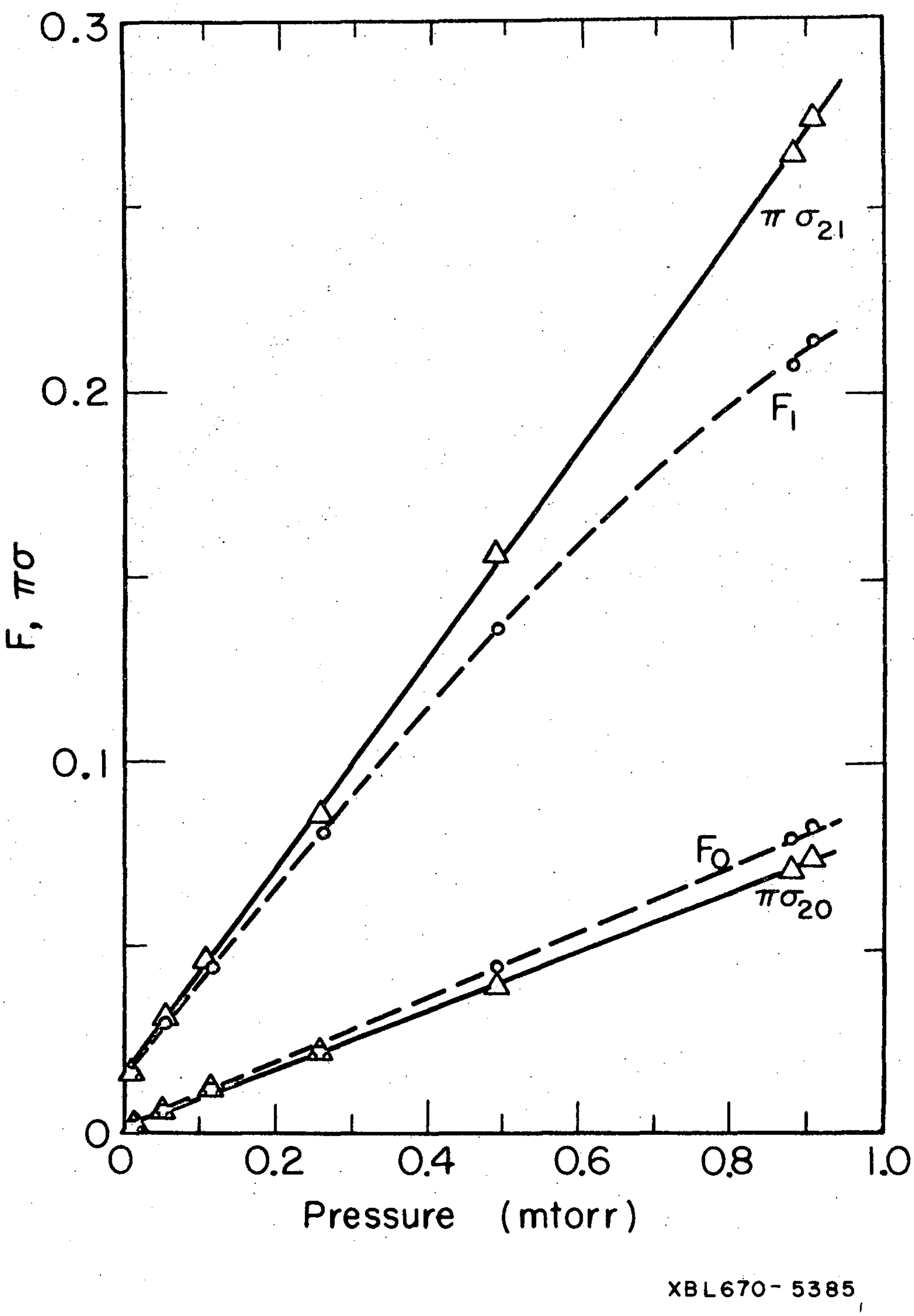
0.3
0.2
0.1
0
F, $\pi\sigma$
$\pi\,\sigma_{21}$
F_1
F_0
$\pi\sigma_{20}$
0
0.2
0.4
0.6
0.8
1.0
Pressure (mtorr)
XBL670-5385

Fig. 2

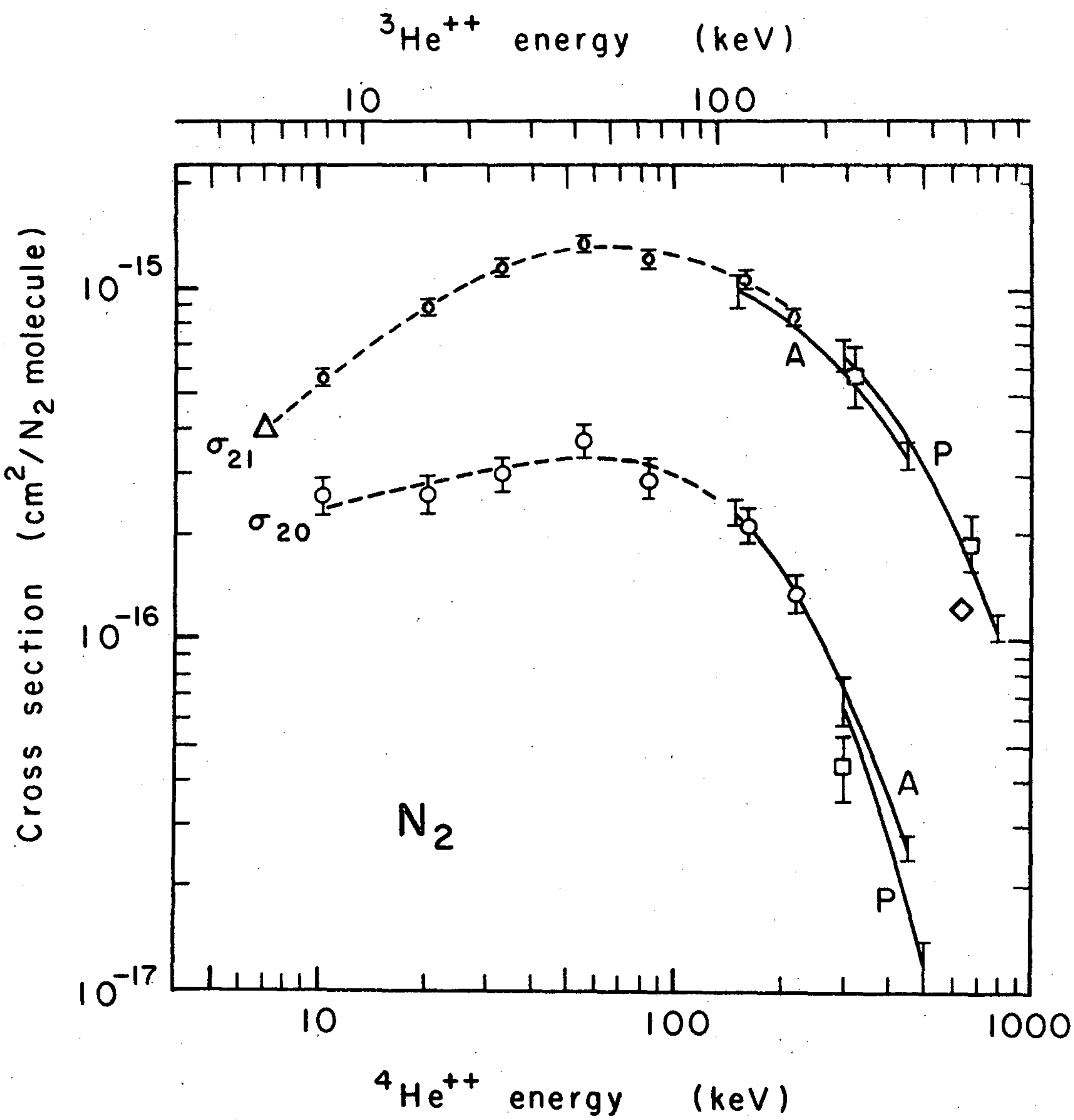

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Fig. 3

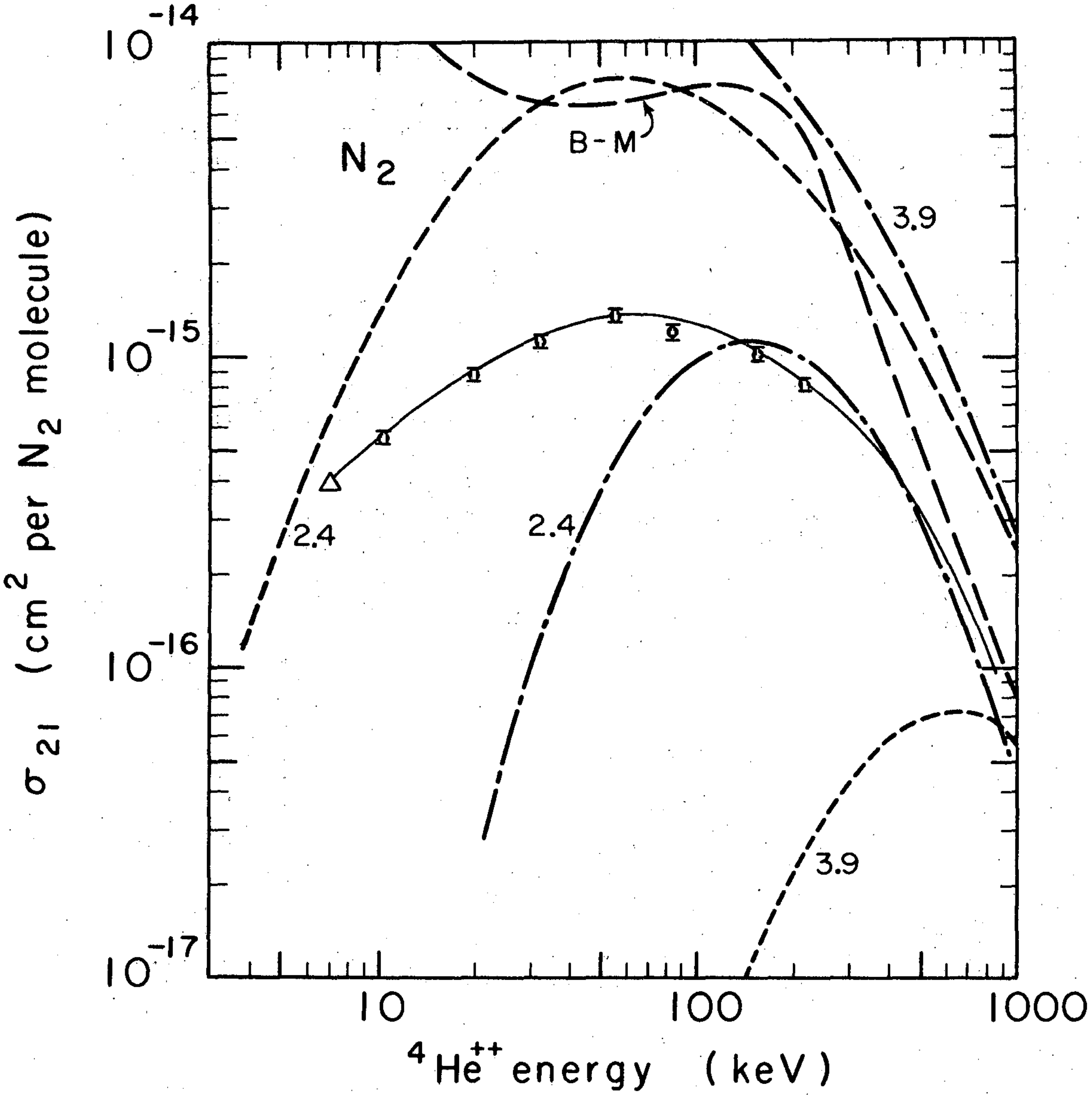

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Fig. 4