

## Spectroscopic Study of Controlled Proton Impact on Molecular Nitrogen\*

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Protons of energy 5–130 keV were passed into molecular nitrogen gas at low pressure and the resulting radiation spectroscopically analyzed. Prominent emissions studied were: (1) two  $N_{II}$  lines located at  $\lambda 5005 \text{ \AA}$  and  $\lambda 5680 \text{ \AA}$ ; (2) bands of the  $N_2^+$  first negative band system ( $B^2\Sigma - X^2\Sigma$ ); and (3) two Balmer lines,  $H_\alpha$  and  $H_\beta$ , formed by charge changing collisions with nitrogen molecules. Absolute cross sections for the production of these emissions were measured as a function of the energy of the incident protons. Population cross sections for the  $v=0$  and  $v=1$  levels of the  $B^2\Sigma$  state of  $N_2^+$  were obtained as well as an estimate of the level population cross section of the  $n=3$  and  $n=4$  levels of atomic hydrogen formed by charge exchange.

### I. INTRODUCTION

STUDIES of spectra resulting from bombardment of atmospheric gases by ions are usually undertaken for two fundamental reasons: the first, inherent in any physical measurement is a basic objective—to obtain quantitative information about the fundamental processes involved; the second is the need for laboratory-experiment results before a complete understanding of upper atmospheric phenomena such as the aurora can be acquired. Meinel's<sup>1</sup> positive identification of Doppler-shifted hydrogen radiation from auroral displays leaves little doubt that protons are among the primary particles entering the upper atmosphere and producing the aurora. The 140-kV positive-ion accelerator at the University of Arkansas provides a source of energetic protons for use in investigating proton-atmospheric gas collisions.

The accelerator employs an Ortec rf ion source and has a useful range of about 5 to 130 kV for studies of this type. A calibrated JaCo 500-mm Ebert spectrometer provides spectral analysis. The experimental apparatus, as well as the calibration procedure, has been described elsewhere.<sup>2</sup>

Beam currents from 0.15 to 5.0  $\mu\text{A}$  were used with the nitrogen pressure kept in the range from 1 to 5  $\mu\text{Hg}$ . Beam current at the 5–10 keV region was substantially less than that attainable at other energies and somewhat limits our confidence in data taken at these low energies. All data were taken with a spectral slit width of 25  $\text{\AA}$  with the exception of two members of the  $v'=1$  progression of the  $N_2^+$  first negative band system. For these two bands, located at  $\lambda 4236 \text{ \AA}$  and  $\lambda 3884 \text{ \AA}$ , a somewhat smaller slit width was found to be necessary in order to affect the necessary resolution.

### II. RESULTS AND DISCUSSION

When a fast proton enters a region occupied by gas molecules, the ensuing collisions may result in excita-

tion of the molecules by one or more mechanisms. The dependence of the intensity of emitted radiation on beam current and gas pressure provides evidence as to the particular mechanism responsible. All spectral features reported in this paper were found to be linear with both current and pressure in the pressure range used, indicating that the emissions were the result of a primary collision process.

Since the emissions observed were caused by a primary collision process, the excitation cross section can be calculated from the following equation:

$$n = \sigma \rho F,$$

where  $n$  represents the number of photons emitted from a cubic centimeter,  $\rho$  is the number density of nitrogen molecules in the target chamber,  $F$  is the proton flux, and  $\sigma$  is the cross section for excitation.

#### A. $N_{II}$ Lines at $\lambda 5005 \text{ \AA}$ and $\lambda 5680 \text{ \AA}$

The cross sections for these  $N^+$  emissions resulting from simultaneous dissociation, ionization, and excitation were the same within experimental error and are displayed in Fig. 1. The cross section at 200 keV, available from previous work,<sup>3</sup> is included for com-

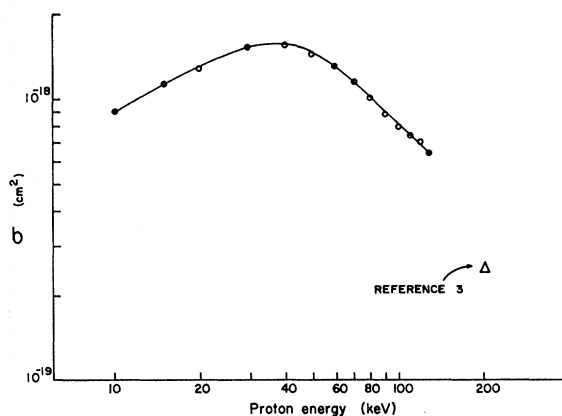


FIG. 1. Cross sections for  $\lambda 5005\text{-\AA}$  and  $\lambda 5680\text{-\AA}$  emission from  $N^+$  induced by  $H^+$  impact on  $N_2$ .

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<sup>1</sup> A. B. Meinel, *Astrophys. J.* **113**, 50 (1950).

<sup>2</sup> R. H. Hughes, Sabrina Lin, and L. L. Hatfield, *Phys. Rev.* **130**, 2318 (1963).

<sup>3</sup> R. H. Hughes, J. L. Philpot, and C. Y. Fan, *Phys. Rev.* **123**, 2084 (1961).

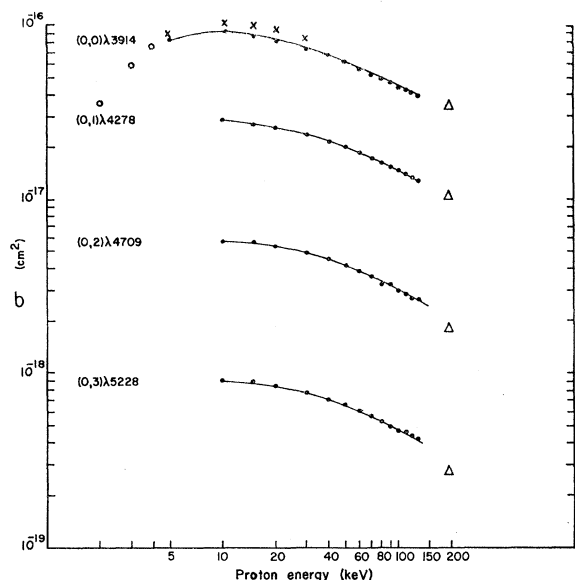
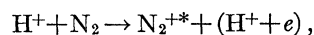


FIG. 2. Cross sections for  $v'=0$  progression emissions in the  $N_2^+$  first negative system.  $\Delta$ ...Ref. 3,  $\times$ ...Ref. 4,  $\circ$ ...Ref. 5.

parison. The maximum cross section appears at the relatively high energy of 40 keV which is indicative of the large energy defect in the excitation process.

### B. $N_2^+$ First Negative Band System ( $B^2\Sigma \rightarrow X^2\Sigma$ )

The process here is



followed by

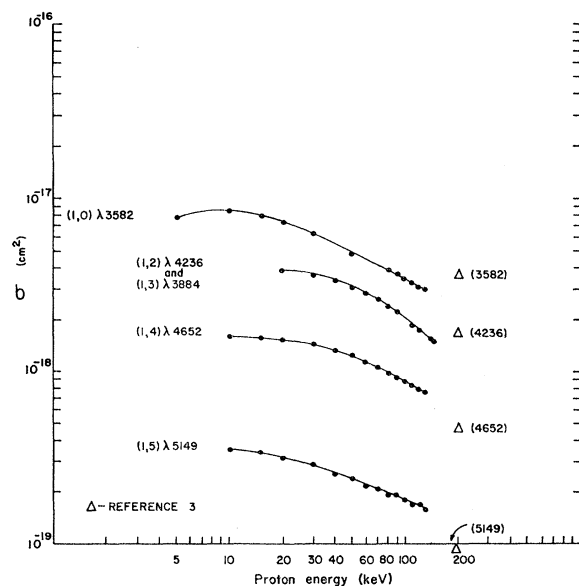
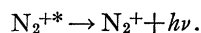


FIG. 3. Cross sections for the  $v'=1$  progression emissions in the  $N_2^+$  first negative system.

The electron resulting from the ionization of the  $N_2$  molecule may be free or captured by the proton in a bound state.

Graphs of the excitation cross section versus energy are shown in Figs. 2 and 3. Results of previous measurements made at 200 keV<sup>3</sup> are also shown. Curves were extrapolated out to 200 keV and it seems that reasonable agreement with the previous work may be justly claimed. The excitation cross section for the (0-0) band, located at  $\lambda 3914 \text{ \AA}$ , has been reported up to 30 keV by Sheridan *et al.*<sup>4</sup> Our measurements agree reasonably well with values read from their curves, showing a peak at approximately the same energy and differing in magnitude by only about 10%. Their results are indicated in Fig. 2. The values obtained by Carleton and Lawrence<sup>5</sup> are also shown in Fig. 2, giving some indication of the behavior of the cross section for energies less than 5 keV. It should be noted

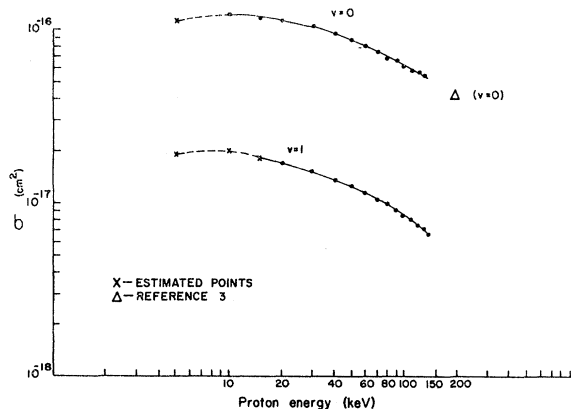


FIG. 4. Population cross sections for the  $v'=0$  and  $v'=1$  levels of the  $B^2\Sigma$  state of  $N_2^+$ .

that the band located at  $\lambda 3577 \text{ \AA}$  belonging to the  $N_2$  second positive system, which should be pressure-dependent, was not resolved from the  $\lambda 3582\text{-\AA}$  band of the  $N_2^+$  first negative system. The  $\lambda 3577\text{-\AA}$  band can be excited by a neutral component of the beam<sup>6</sup> or by secondary electrons.<sup>3</sup> The excitation of the  $N_2$  second positive system by direct proton impact is ruled out by the Wigner spin conservation rule.<sup>7</sup> Values listed in this paper would seem to be nearly correct since measurements made on the  $\lambda 3582\text{-\AA}$  band were made at such low pressures that the contribution from the  $\lambda 3577\text{-\AA}$  band is expected to be negligible.

Summing the cross sections for the bands of the  $v'=0$  progression up to and including  $v''=3$  and the

<sup>4</sup> W. F. Sheridan, O. Oldenberg, and N. P. Carleton (abstracts) *Second International Conference on the Physics of Electronic and Atomic Collisions, University of Colorado, 1961* (W. A. Benjamin, Inc., New York, 1961), p. 159.

<sup>5</sup> N. P. Carleton and T. R. Lawrence, *Phys. Rev.* **109**, 1159 (1958).

<sup>6</sup> N. P. Carleton, *Phys. Rev.* **107**, 110 (1957).

<sup>7</sup> E. Wigner, *Nachr. Akad. Wiss. Goettingen, Math.-Physik. Kl. IIa. Math.-Physik.-Chem. Abt.* **1927**, 375.

TABLE I. Transition probabilities associated with the  $N_2^+$  first negative band system.

Transition	A <sup>a</sup>	B <sup>b</sup>	C <sup>c</sup>
0-0	0.715	0.69	0.67
0-1	0.229	0.26	0.23
0-2	0.048	0.04	0.08
0-3	0.007 <sub>5</sub>		
1-0	0.40	0.23	0.28
1-1	0.24 <sub>5</sub>	0.27	0.25
1-2	0.23 <sub>5</sub>	0.38	0.26
1-3	0.10	0.11	0.14
1-4	0.02	0.01	0.04

<sup>a</sup> A—measured by this experiment (fast  $H^+$  impact).

<sup>b</sup> B—measured by Herzberg, Ref. 8.

<sup>c</sup> C—calculated by Pillow, Ref. 9.

$v'=1$  progression up to and including  $v''=4$  yields the population cross sections for the  $v'=0$  and the  $v'=1$  levels of the  $B^2\Sigma$  state of  $N_2^+$  (Fig. 4). The contribution from bands having larger  $v''$  values in the progressions has been neglected. Our measurements of the relative

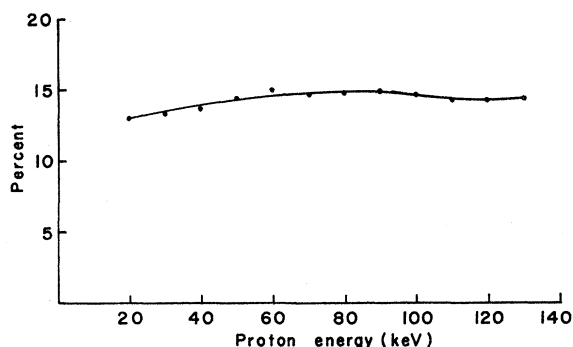


FIG. 5. Percent of  $N_2^+$  excited to the sum of the  $v'=0$  and the  $v'=1$  levels of the  $B^2\Sigma$  electronic level.

transition probabilities for each band of the system are listed in Table I, where they are compared with the measurements of Herzberg,<sup>8</sup> and the calculated values of Pillow.<sup>9</sup> Our values for members of the  $v'=0$  progression are very nearly constant over the entire energy range, giving us considerable confidence in the listed values. Our values for members of the  $v'=1$  progression were not so consistent over the energy range, varying as much as 7%. Most of this variation may be attributed to the difficulty in resolving the two bands located at  $\lambda 4236 \text{ \AA}$  and  $\lambda 3884 \text{ \AA}$ . The possibility cannot be overlooked that  $\lambda 3582\text{-\AA}$  measurements may be in slight error because of the previously mentioned background problem. This still leaves considerable disagreement between our transition probabilities as measured by proton impact and Herzberg's values for this progression.

Our relative band intensities for the (0-0), (0-1),

<sup>8</sup> G. Herzberg, Ann. Phys. Lpz., 86, 191 (1928).

<sup>9</sup> M. E. Pillow, Proc. Phys. Soc. (London) A64, 772 (1951).

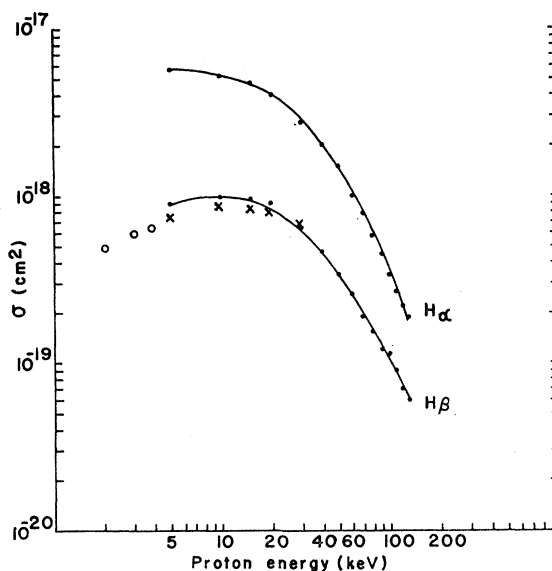


FIG. 6. Cross sections for  $H_\alpha$  and  $H_\beta$  emission from  $H^+$  impact on  $N_2$ .  $\times$ ...Ref. 4,  $\circ$ ...Ref. 5.

and (0-2) members of the band system are 1.00:0.32:0.067 which compare quite favorably with Bates<sup>10</sup> calculated values of 1.0:0.31:0.072 as quoted by Stewart<sup>11</sup> who obtained relative band intensities of 1.00:0.39:0.10 by electron impact.

Experimental evidence indicates that the excitation of  $v'$  progressions for  $v' \geq 2$  is small. The sum of the cross sections for the  $v'=0$  and  $v'=1$  progressions may then be assumed to closely represent the total excitation cross section for the  $B^2\Sigma$  state. Using the recent

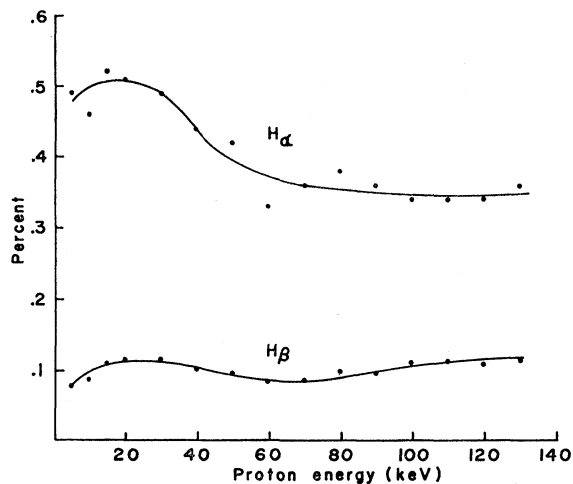


FIG. 7. Percent of total charge transfer resulting in  $H_\alpha$  and  $H_\beta$  emission by  $H^+$  impact on  $N_2$ .

<sup>10</sup> D. R. Bates, Monthly Notices Roy. Astron. Soc. 112, 614 (1952).

<sup>11</sup> D. T. Stewart, Proc. Phys. Soc. (London) A69, 437 (1956).

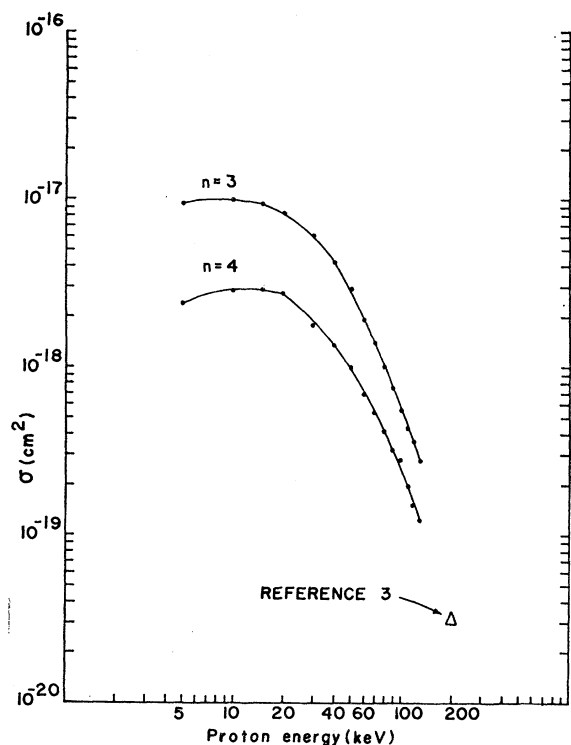


FIG. 8. Estimate of the population of the  $n=3$  and  $n=4$  levels of hydrogen through charge transfer in  $N_2$ .

measurements of Solov'ev *et al.*<sup>12</sup> on the cross section for formation of the  $N_2^+$  ion under proton bombardment, we can calculate the fraction of  $N_2^+$  formed in the excited  $B^2\Sigma$  state. This procedure, displayed in Fig. 5 shows the rather interesting result that the fraction of  $N_2^+$  excited to the  $B^2\Sigma$  state by proton collision remains at the nearly constant level of  $\sim 15\%$  over the entire energy range studied. Unfortunately, their data cut off on the low-energy end at 20 keV, and we could not plot this curve down to our low-energy limit of 5 keV.

### C. Hydrogen Lines, $H_\alpha$ and $H_\beta$

The reaction for the first-order process resulting in these emissions is electron capture from the nitrogen molecule.

<sup>12</sup> E. S. Solov'ev, R. N. Il'in, V. A. Oparin, and N. V. Fedorenko, *Zh. Eksperim. i Teor. Fiz.* **42**, 659 (1962) [translation: *Soviet Phys.—JETP* **15**, 459 (1962)].

The excitation cross sections for  $H_\alpha$  and  $H_\beta$  emissions are displayed in Fig. 6. The values obtained by Sheridan *et al.*<sup>4</sup> and Carleton and Lawrence<sup>5</sup> are also shown on this figure. Comparison of our results for  $H_\beta$  with these workers up to 30 keV shows that our results agree quite well with their work.

Using the values for the total charge exchange cross section from Allison's review article,<sup>13</sup> and our measured values for the excitation cross section, we plotted the percentage of charge changing collisions resulting in  $H_\alpha$  and  $H_\beta$  emission. The results of this procedure are displayed in Fig. 7.

By using our data, we may estimate the level population cross sections of the  $n=3$  and the  $n=4$  states of hydrogen formed by charge exchange. The procedure outlined in a previous paper<sup>2</sup> was used. To obtain these figures, it is necessary to assume that the cross sections for capture into the  $np$  and  $nd$  levels relative to the capture cross section into the  $ns$  level (where  $n$  is 3 or 4) do not differ appreciably from the ratios calculated from the work of Bates and Dalgarno<sup>14</sup> on  $H^+$  on  $H$ . The results are plotted in Fig. 8.

*Note added in proof.* The cross-section measurements involving radiation from fast hydrogen atoms shown here are based on the assumption of a constant excited hydrogen-atom density along the beam path in our observation region. This is likely a very poor assumption in our experimental set up. Our observation region should be at a distance from the beam-entrance aperture which is great compared with the product of the velocity times the lifetimes of the excited states in order that treatment given here to be entirely valid. The center of our 3-cm-long observation region is perhaps a little better than 2 cm from the beam-entrance aperture. If this effect is taken into account, it is possible that our hydrogen cross-section values may be too small by as much as 25% and 300% at 5 and 100 keV, respectively. This effect is being checked experimentally. These remarks also apply to prior publications.

### ACKNOWLEDGMENTS

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<sup>13</sup> S. K. Allison, *Rev. Mod. Phys.* **36**, 1137 (1958).

<sup>14</sup> D. R. Bates and A. Dalgarno, *Proc. Phys. Soc. (London)* **A66**, 972 (1953).