

Absolute Cross Sections for Excitation of Nitrogen by Protons of a Few keV Energy*

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Cross sections for excitation by proton impact are necessary for quantitative interpretation of auroral spectra. These have been measured for excitation resulting when protons of a few keV energy shoot into low-pressure nitrogen. Interference filters select various spectral features and a photomultiplier detects the light emitted. The light-detection apparatus is calibrated against a standard tungsten filament lamp. The measured cross sections are not for the excitation of a given molecular level, but for a process resulting in the emission of one photon in a given transition. Cross sections measured include: the 0,0 first negative band of N_2^+ , $\lambda 3912$; the 2,0 Meinel band of N_2^+ , $\lambda 7850$; a group of N I lines around $\lambda 8216$; the Balmer line $H\beta$; and the (4,2) and (3,1) first positive bands of N_2 , excited by impact of fast atoms. Results are presented graphically, covering the range 1.5 to 4.5 keV. Also included are measurements of the total charge-exchange cross section for protons in nitrogen and an estimate of the ionization cross section.

I. INTRODUCTION

MEASUREMENTS of cross sections for excitation by proton impact serve two purposes. The first is that inherent in any such measurement: to give quantitative information about a fundamental process by which fast ions suffer an energy loss and by which they affect the matter through which they pass. Excitation by ion impact has received some experimental attention¹⁻⁶ but mostly under ill-defined conditions, so that cross sections for particular reactions could not be determined. Recently, Bates and collaborators have made many theoretical calculations on collisions; they are limited mostly to those between hydrogen and helium atoms and ions.⁷⁻¹¹ In general, collisions of ions with molecules must be investigated by experiment.

The second reason for these present measurements is the need for data on which to base a quantitative interpretation of auroral spectra. It seems certain that at least part of the primary excitation of the aurora is due to fast protons entering the atmosphere from outside.¹² Hence it is of interest to find out the mechanisms by which such protons can excite molecules

of atmospheric gases. A previous paper¹³ reported some semiquantitative work on this problem which established excitation mechanisms for several spectral features. The present paper is a report of further measurements giving absolute cross sections for excitation of nitrogen by impact of fast protons and H atoms, and for production of excited H atoms in collisions with nitrogen. Measurements of absolute cross sections for charge exchange also accompanied the optical measurements, giving a link between optical and electrical phenomena.

II. APPARATUS

To measure these cross sections we must shoot a proton beam of well-defined energy into gas at a low enough pressure so that the number of protons in the beam is not reduced by more than a few percent through charge exchange and scattering in the observing region. We then must measure the beam current, the gas pressure, and the light emitted from a measured length of the beam path. For charge-exchange measurements we must in addition measure the number of slow gas ions formed in a given length of the beam.

The vacuum and gas-handling system and the arrangement for producing the proton beam are just those described in I. Briefly, ions are drawn from an rf discharge, accelerated and run through a mass analyzer, and the resulting proton beam is focused with about 20 μ a current on a $\frac{1}{16}$ -in. diameter hole leading to an observation chamber. The beam collector used in the present experiments, shown in Fig. 1, is designed to detect slow electrons and N_2^+ ions created through charge exchange and ionization by the passage of the beam.¹⁴ These slow particles are swept out by a transverse field, maintained by batteries between the plates shown in Fig. 1. The pair of plates *B* spans the length of the beam that is actually under optical observation; the pairs *A* and *C* serve as guard rings, and pair *C* also

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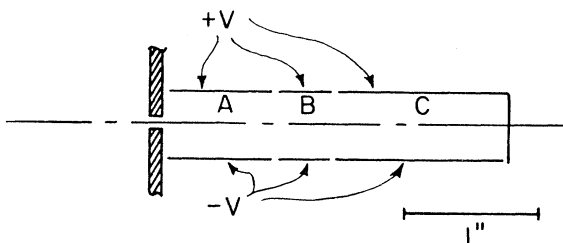


FIG. 1. Electrodes for measuring total beam current and currents due to charge exchange and ionization.

serves as a Faraday cage. It is possible to measure the current to ground from any one of the plates separately, or the algebraic sum of all the currents to ground from any of the three pairs of plates. The current reaching the positive *B* plate is a measure of the number of free electrons produced by the beam, while the net positive current to the pair of plates *B* is a measure of the number of slow N_2^+ ions formed by charge exchange. An electron-ion pair formed between the plates *B* does not contribute to this net current collected by the pair. A given potential difference applied to the plates *A* and *B* sweeps out only those ions formed between them which have less than a certain amount of forward momentum. Thus, the potentials required to reach a saturation value of the currents to the electrodes will give a measure of the forward momentum of the ions and electrons.

The gas pressure in the region of observations is measured by a Pirani gauge, separately calibrated against a McLeod gauge. This gauge is attached to the bottom of the observation chamber directly below the part of the beam which is under optical observation. Thus, though there is a flow of gas through the chamber, the gauge is not upstream or downstream from the point where we wish to know the pressure. At the pressures used ($0.5\text{--}5\ \mu\text{ Hg}$), with the mean free path in the range $1\text{--}10\text{ cm}$, it seems reasonable that the gauge reading approximates the true pressure at the beam well enough so that other errors (discussed below) limit the accuracy of the measured cross sections.

The optical system is shown in Fig. 2. It is fortunately possible to isolate spectrally, with interference filters of $75\text{--}100\ \text{\AA}$ band width, most of the features of interest in the spectrum of the proton beam in nitrogen. The beam lies at the focal point of lens L_1 so that light enters the interference filter as nearly parallel as possible. The lens L_2 focuses the light, through additional glass filters, making an image in the plane *S*, where a mask defines the portion of the beam from which light is accepted. The lens L_3 is a field lens, collecting light onto the cathode of a photomultiplier. The photomultipliers used are RCA type C7160 for the infrared, and RCA type 6199 for the visible and near ultraviolet. These can be used interchangeably, having the same envelope and socket. The shield can shown in Fig. 2 is in turn enclosed in a box of

polystyrene foam (not shown) so that the photomultipliers can be cooled with dry ice. This is especially necessary for the infrared tube. Dishes of P_2O_5 inside the shield can and the preamplifier chassis remove water vapor, preventing condensation of moisture on the cold parts.

This optical system was calibrated against a tungsten filament standard lamp. The first step in the calibration was to measure the transmission as a function of wavelength of the various interference filters used, each together with its appropriate colored glass blocking filters, by means of a prism monochromator. Then, with the whole optical system removed from the rest of the apparatus, the standard lamp replaced the beam as source of light, being set up about 25 cm further from the mirror than was the beam. This different position required us to shift the lens L_2 and the interference filter from their previous positions, but all elements of the optical system, including a dummy piece of quartz window, remained in the path of the light. To limit and define the intensity, the area utilized of L_1 was from 0.1 to 0.3 cm^2 , depending on the wavelength being observed, and the mask at *S* selected radiation from only 1.6 mm^2 of the filament. Under these conditions, with a given interference filter in place, we could determine the number of microwatts of radiation incident on the photomultiplier, except for a factor depending on the transmission of the optical system. Since this factor was arranged to be the same for both measuring and calibrating exposures, we thus had an absolute calibration of our detector in the wavelength range of each interference filter, assuming that the sensitivity of the photomultipliers did not vary much over the pass band of a filter. The manufacturer's data show this latter assumption to be good.

In order to keep a continual check on the sensitivity of the detector, we provided as a secondary standard a piece of white paper illuminated by a small incandescent bulb run at low voltage from a battery. This arrangement could be moved into a well-defined position so as to illuminate the photomultiplier, as shown in Fig. 2. We measured the detector response to this light for a given current through the bulb at the time the detector was calibrated, and checked the sensitivity of the detector against this value before and after every measurement. Since the light from this

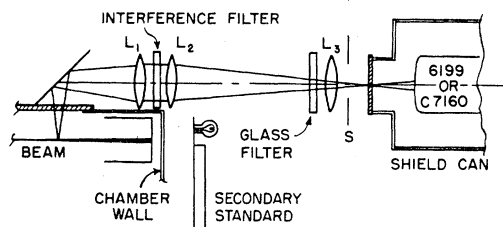


FIG. 2. Optical system for absolute measurement of intensities of light.

secondary standard was filtered only by the glass filters, this method of checking assumes that the shape of the spectral sensitivity curve of the photomultiplier does not change with time over the range covered by the glass filters.

III. METHOD AND RESULTS

Electrical Measurements

In order to find the true value of the beam current and to calculate the charge-exchange cross section we studied how the currents to the plates *B* and the collector *C* (see Fig. 1) varied with the potential *V* and with the gas pressure. With the best possible vacuum (5×10^{-5} mm Hg) in the observation chamber, at low sweeping voltages ($V < 5$ v) a negative current amounting to a few percent of the positive collector current reached the pair of plates *B*. At higher voltages this negative current disappeared. It was presumably due to secondary electrons ejected either from the back of the collector or from the sides of the entrance canal.

With nitrogen of $1-5 \mu$ Hg pressure in the observation chamber the net positive current to the pair of plates *B* increased with increasing *V*, reaching a saturation value at about $V = 30$ v. At the same time the collector current decreased, approaching a steady value. Figure 3 shows a typical variation of these currents with the potential *V*; the voltage dependence of these curves did not change much with pressure in the above range, nor with the proton energy in the range 1.5-4.5 kev. The large voltage required to reach saturation of the net positive current seems to indicate that the N_2^+ ions formed by charge exchange have considerable forward momentum. The saturation voltage is presumably that at which the fastest ions formed just inside the entrance to the chamber are all captured by the *A* plates. Above this voltage it should be correct to say that the plates *B* are collecting a number per second of positive ions equal to that formed in a length *L* of the beam. (See Fig. 4.) As the voltage is further increased, plates *A*

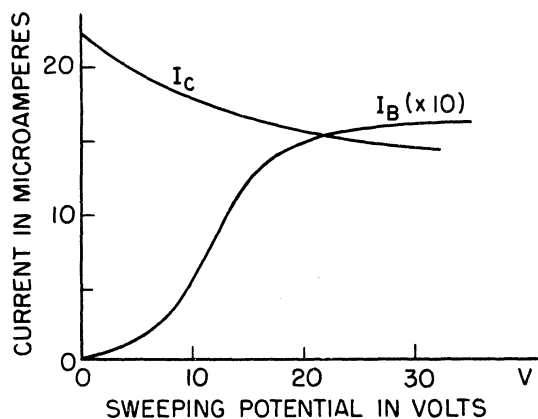


FIG. 3. Beam current, I_C , and swept-out current of slow N_2^+ ions, I_B , as functions of sweeping potential. N_2 pressure 6 microns Hg, the highest value used.

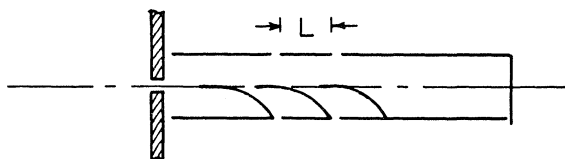


FIG. 4. Trajectories of slow N_2^+ ions in sweeping field.

collect more ions, plates *C* collect fewer, but plates *B* collect the same. We assume that the pressure is low enough so that the rate of ion formation along the track of the beam is nearly constant.

The shape of the I_B vs *V* curve for the *B* plates is determined by the momentum distribution of secondary ions and electrons formed in the gas. Unfortunately, lack of knowledge of the angular distribution of these particles makes it impossible to extract any detailed information from the measured curves. We can conclude, however, from the observed saturation voltages that there are some N_2^+ ions formed by charge exchange which have as much as 50 ev energy. These saturation voltages are considerably higher than those observed by Keene,¹⁴ who measured charge exchange for higher energy protons in various gases. This suggests that slower charge-exchange collisions result in a greater transfer of momentum, but a direct comparison is not possible since Keene did not work with nitrogen. There is no appreciable variation of the saturation voltage with energy in the range of this experiment. Since there must exist, in the range 0-100 kev bombarding energy, an energy for which there is maximum probability for large momentum transfer, it may be that this maximum is just in the range 1-5 kev.

The program for measuring cross sections for charge exchange was to plot $\ln(1 + I_B/I_C)$ as a function of N_2 pressure, where I_B and I_C are the net positive currents at saturation to plates *B* and plates *C* (the collector). This plot should be a straight line if the slow ions collected at *B* are produced by a simple charge-exchange process; the cross section for this process can be determined from the slope of the line. The experimental data do give straight lines, when so plotted, up to a pressure of about 5μ Hg. Above this pressure there is a departure from linearity in the direction explained by a failure to sweep out all the slow ions formed between plates *A* and *B*. The fraction of these which reaches plates *C* becomes an increasingly important addition to I_C at higher pressures, where I_B is as much as 10% of I_C . The measured cross sections, derived from the slopes of the logarithmic plots just described, in the pressure region 0-5 μ Hg, are probably correct to about 5%, relative to each other. They may, however, be systematically in error because of incorrect measurement of the N_2 pressure and because of possible losses of ions by scattering out of the collection region or losses of secondary electrons out of the beam collector. Both these latter errors would tend to make the meas-

ured cross sections too small. A comparison with previous work of Stier and Barnett¹⁵ is possible, since the energy range of the present experiment just overlaps theirs. Our plot of cross section *vs* energy joins nicely onto theirs if our values are all increased by 30%. Since their experiment was designed specifically for electrical measurements, whereas ours was designed for optical measurements, they had more opportunity to determine systematic errors, so that we believe that they are more likely than we to have the right absolute value. Hence we have plotted our cross sections increased by 30% in Fig. 6.

From the negative current to the positive *B* plate at saturation it should be possible to determine the number of free electrons produced by the proton beam and hence the cross section for ionization, in addition to that for charge exchange. Unfortunately, at the saturation voltage of 30 v or so, electrons produced in the beam gain enough energy while being swept out to produce secondary ionization. The ion-electron pairs produced by this secondary mechanism do not affect the charge-exchange measurements, which depend upon the *net* positive current to the *pair* of plates *B*. Indeed, we have observed that the individual currents to the *B* plates are increasing rapidly with increasing voltage, while the net current is leveling off to a constant value. Thus we cannot make an accurate measurement of the free-electron current at sweeping potentials higher than the ionization potential of N_2 . On the other hand, this current does not show a complete saturation for lower sweeping potentials. Therefore, on the matter of ionization of N_2 by proton impact, we shall restrict ourselves to saying that the cross section for this is between 1% and 1.5% of the cross section for charge exchange in the energy range covered.

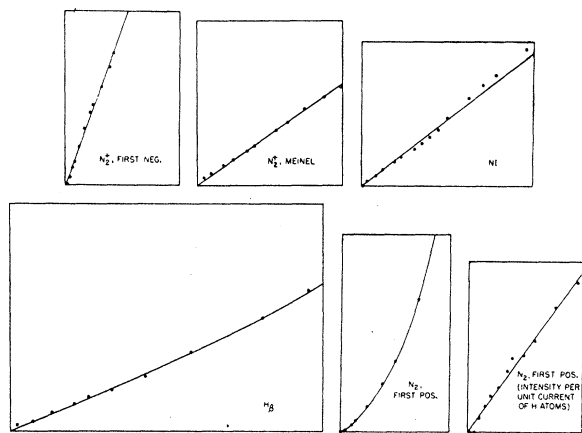


FIG. 5. Plots showing dependence of light intensity per unit current of bombarding particles (ordinates) on N_2 pressure (abscissas). The scales are all linear, with arbitrary units.

¹⁵ P. M. Stier and C. F. Barnett, Phys. Rev. **103**, 896 (1956).

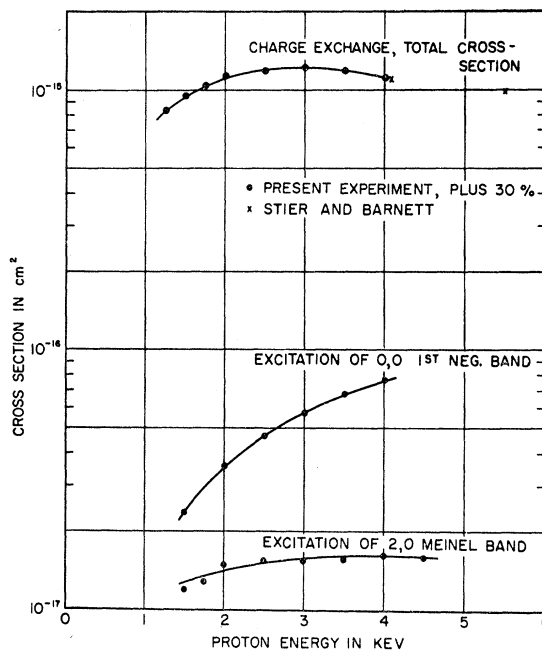


FIG. 6. Cross sections for total charge exchange and for excitation through charge exchange, protons in N_2 .

Optical Measurements

With this information on the electrical events taking place in the beam, it is possible to interpret the optical measurements. To measure cross sections for excitation by proton impact we used the arrangement described in Sec. II to measure as a function of N_2 pressure the light output per unit beam current for a given spectral feature, isolated by an interference filter. During these measurements we had to keep the sweeping voltage lower than the excitation potential of the N_2 emissions we were observing. Since this meant in all cases keeping it below the saturation voltage, the collector current included some N_2^+ ions which were not swept out. We made a correction for these, using the previously measured I_B *vs* V curves (see Fig. 3). Also, from these curves we could compute the average current of protons passing through the observed region. Now, for the corrected value of beam current, a plot of light intensity per unit of current against N_2 pressure should be a straight line if the excited state is produced in a single collision between a proton and an N_2 molecule. Sample plots are shown in Fig. 5 for the following emissions: the (0,0) band of the first negative system of N_2^+ , $B^2\Sigma_u^+ - X^2\Sigma_g^+$, $\lambda 3914$ Å; the (2,0) band of the Meinel system of N_2^+ , $A^2\Pi_u - X^2\Sigma_g^+$, $\lambda 7850$ Å; certain N I lines, $3p^4P^0 - 3s^4P$, $\lambda 8188-8216$ Å; the (2,0) and (3,1) bands of the first positive system of N_2 , $B^3\Pi_g - A^3\Sigma_u^+$, $\lambda 7500$ Å; and the line H_β of atomic hydrogen, $\lambda 4861$ Å. This figure presents a summary of evidence already cited and discussed in paper I.

The N_2^+ bands have plots which are linear up to a

pressure of 5 or 6 μ Hg. Above this pressure there are again departures from linearity, presumably because we could no longer calculate the proton current accurately. To obtain absolute values of light output, we used the calibration of the photomultiplier described in Sec. II. Then, by comparing the profile of a band, determined spectrographically, with the transmission curve of the interference filter, we could find what fraction of the total number of photons in the band was transmitted by the filter. From the slopes of the linear portions of the plots we have calculated cross sections for the production of one photon in each band. These are slightly different from cross sections for exciting a given level, since effects of cascading are included. The cross sections for the N_2^+ bands are shown in Fig. 6 as functions of the proton energy. As discussed in paper I, these bands are presumably excited by charge-exchange collisions leaving the resulting N_2^+ ions in the excited state. The relative scarcity of free electrons suggests that for these energies there are not many collisions which produce a free electron and an excited ion. Therefore, the lower curves in Fig. 6 are presumably to be compared directly with the one giving the total charge-exchange cross section. This comparison gives the fraction of all charge-exchange collisions which result in emission of a 3914 A photon or a 7850 A photon, as shown in Fig. 7.

It would be interesting, in addition, to know the relative numbers of ions produced in the $A^2\Pi_u$, $B^2\Sigma_u^+$, and $X^2\Sigma_g^+$ states, summing over all vibrational levels. This requires a photometric study of all bands of the first negative and Meinel systems, which is difficult to make, because of overlapping of these systems with the first and second positive systems, and

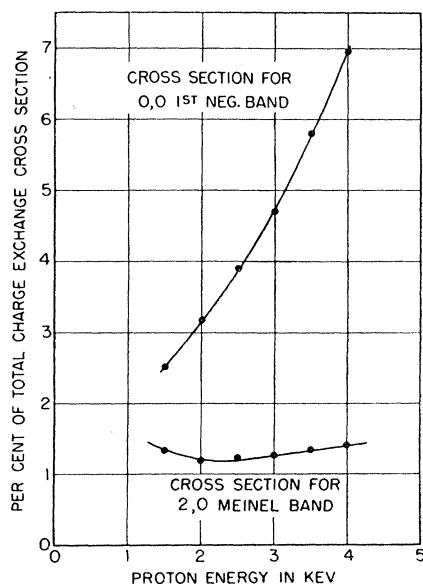


FIG. 7. Relation of cross sections for excitation through charge exchange to total charge-exchange cross section, protons in N_2 .

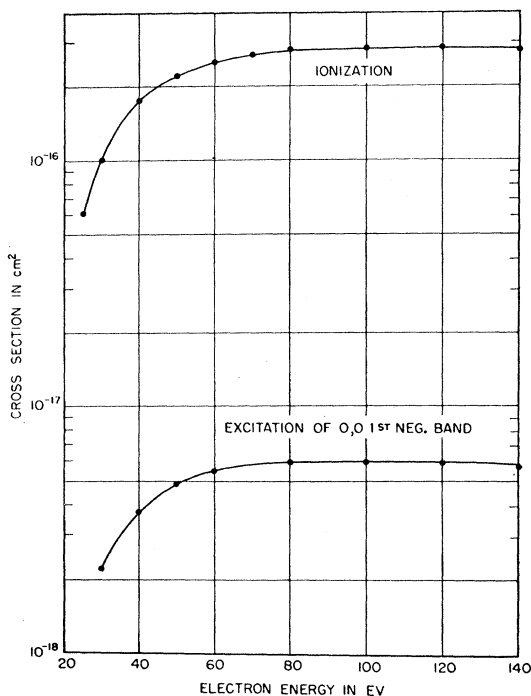


FIG. 8. Cross sections for ionization and excitation of N_2 by electron impact, for comparison with Fig. 6.

because the Meinel system extends as far into the infrared as 1.1 μ . For these reasons we were not able to make this measurement in these experiments.

Figure 8 shows the cross section for ionization of N_2 by electron impact, as measured by Tate,¹⁶ and the cross section for exciting a 3914 A photon by electron impact, as measured by Stewart.¹⁷ Comparison of Figs. 6 and 7 indicates that the charge-exchange process is apparently more efficient at producing light than the electron-excitation process, by a factor of about two. High-energy protons ($E > 40\,000$ kev) must behave about as electrons in exciting and ionizing N_2 , but low-energy charge-exchange collisions, having no counterpart in electron excitation, are not expected to follow the same rules. Massey and others have discussed how inelastic collision cross sections for ion impact should vary with energy. Paper I makes reference to this work and shows that for protons colliding with N_2 , the cross section for a collision involving a net internal energy change ΔE of both partners should fall off rapidly with decreasing energy below a value $E_0 \cong 300(\Delta E)^2$, if both energies are measured in ev. The energy E_0 is about 4000 ev for the Meinel band, and about 8000 ev for the first negative band. For the total charge-exchange cross section, involving the production of either of the two above excited states or the ground state of N_2^+ , E_0

¹⁶ J. T. Tate and P. T. Smith, as quoted in H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Clarendon Press, Oxford, 1952).

¹⁷ A. L. Stewart, Proc. Phys. Soc. (London) A69, 437 (1956).

should be lower than 4000 ev. These predictions are very well fulfilled by the data shown in Fig. 6.

The N I lines, $\lambda 8188-8216$ A, are also apparently excited by a direct collision between H^+ and N_2 . Figure 9 gives the cross section for their excitation as a function of energy. Stewart¹⁸ has excited these lines by controlled electron impact, finding an appearance potential of about 23 v, which is about the minimum one could expect, since it is necessary to dissociate the molecule (9.8 ev) and excite one of the atoms to the $3p^4P$ level (11.8 ev). Presumably the same process is operating in the proton excitation. The only puzzling thing is that according to Massey's criterion, cited above, the cross section for this process should fall off rapidly below proton energies of about 130 kev. If this applies, the cross section for exciting these lines by high-energy protons must be huge. Exciting these lines through charge exchange, producing an atomic ion and the excited atom, seems no more likely. The lines do appear in the auroral spectrum as a reasonably bright feature, but are not outstanding.

From Fig. 5 it is clear that the first positive bands are not excited by a single collision. The last panel of Fig. 5 shows the light per unit current of neutral H atoms as a function of pressure. We could estimate this neutral "current" from our electrical measurements, though not with great accuracy, since the beam may have had some small unknown neutral component as it entered the observation chamber. The linearity of this plot, together with other evidence cited in paper I, leads us to decide that the first positive bands in these experiments were primarily excited by impact of fast H atoms (the excitation by direct proton impact being forbidden by conservation of spin requirements). Figure 10 shows the cross section for this process as a function of energy. The scatter in the data demonstrates our uncertainty in calculating the neutral atom current.

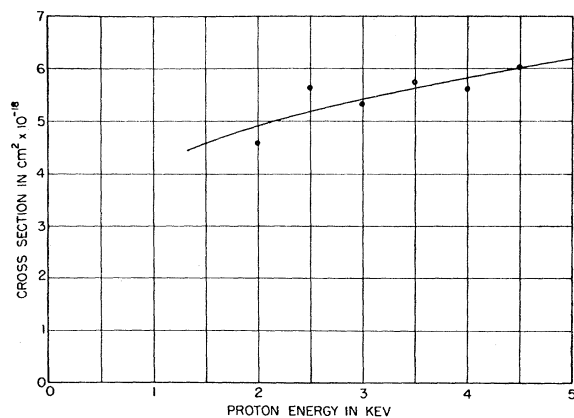


FIG. 9. Cross section for excitation of N I lines, $\lambda 8188-8216$ A by proton impact on N_2 .

¹⁸ A. L. Stewart, Proc. Phys. Soc. (London) A68, 404 (1955).

If the excitation of these bands really does not occur by direct proton impact, then the plot of intensity per unit proton current against pressure should approach the origin with zero slope. By inspection of these experimental curves we can set an upper limit on the slope at the origin which corresponds to a limit on the cross section of 8×10^{-20} cm² for excitation by proton impact.

We now come to the Balmer line H_β , which looks from Fig. 5 to be excited both by a direct process and also by a secondary process, since the curve rises notably faster than linearly at the higher pressures. We expect this (see paper I), since there is the possibility of direct capture of an electron into an excited state and also that of excitation by subsequent collision of a fast H atom formed in its ground state. We attempted to fit our data for the intensity of H_β as a function of N_2 pressure by an expression having a term proportional to the proton current and a term proportional to the

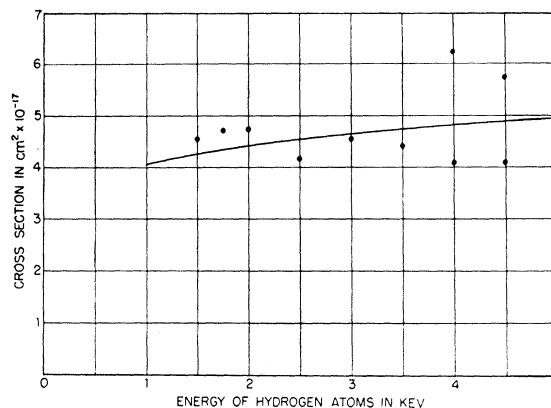
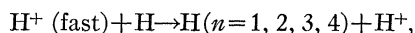


FIG. 10. Cross section for excitation of the 2,0 and 3,1 first positive bands by impact of fast H atoms on N_2 .

neutral beam current. For the cross section for direct capture into the excited state (proportional to proton current) we found reasonably accurate values which are shown in Fig. 11. For the process of excitation of an H atom our calculations are inaccurate, since the departure of the curves like that of Fig. 5 from linearity is slight in the region of pressure which we trust. This, combined with our uncertainties about the current of neutral atoms, makes the cross sections very inaccurate indeed. We obtain a cross section decreasing linearly from 12×10^{-19} cm² at 1500 ev to 5×10^{-19} cm² at 4000 ev. We do not entirely believe these figures, since it does not seem reasonable for the cross section to be decreasing rapidly with energy in this region. Hence, we shall restrict ourselves to saying that the process of exciting a fast H atom by collision with an N_2 molecule (with H_β resulting) has a cross section of about 8×10^{-19} cm² in this energy range.

Bates and Griffing⁷ and Bates and Dalgarno¹⁰ have calculated cross sections for the reactions



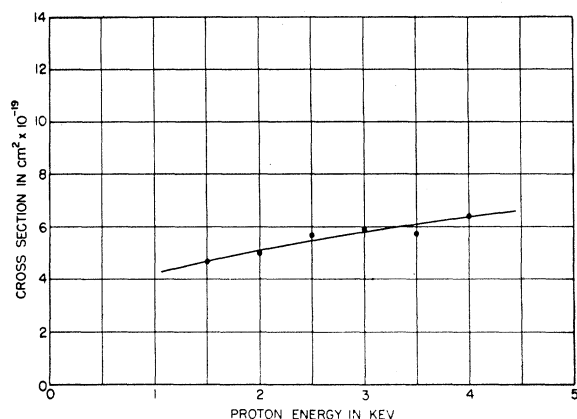
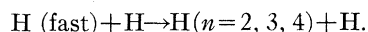


FIG. 11. Cross section for producing the Balmer line H_β through electron capture into excited states by protons passing through N_2 .

and



They find that the cross sections for producing excited states with $n=3$ or higher by either of these reactions are about equal at the energies of our experiments. We estimate from their results that the cross section for producing H_β by either reaction would be of the order of $2 \times 10^{-19} \text{ cm}^2$. The close agreement between these calculations and our measurements is certainly fortuitous, especially since charge exchange of ions in a gas of like atoms has unusual properties. Since the results of Bates and collaborators have been used in approximate calculations on the aurora,¹⁹ it is helpful that this fortuitous agreement exists, giving us more confidence in these calculations.

In conclusion, we must estimate the reliability of the optical measurements. The chief error here is in the calibration and use of the photomultiplier, since the errors in calibrating the filters, the multiplier itself, the standard lamp, and the secondary standard could all

add together. In our estimation, the cross sections for direct processes are probably correct to within 15%. The cross section for excitation of the first positive bands could be in error by 50% or so, for reasons mentioned above, and that for excitation of fast H atoms to produce H_β could be in error by a factor of two.

One undetected systematic error could lie in the pressure measurement, and indeed our failure to agree with Stier and Barnett on the electrical measurements could be attributed to such an error. As stated above, we believe that the lack of agreement is due to other factors, but if we are wrong here, our figures for the optical cross sections may be too low.

Another source of systematic error is light transmitted in the wings of the pass band of the interference filter. This is probably not important in the measurements on the beam, but may be more important in the calibration, where the filter is supposed to select a narrow band from a continuous spectrum. We measured the transmission curve for each filter out to wavelengths where the transmission was less than 1% of the peak value. This is a good rejection ratio for unwanted light; supplementing it are the effect of the glass blocking filters used, and the effect of our wavelengths of interest fortunately being near the maximum of the photomultiplier sensitivity curves in all cases. Therefore we estimate that any error caused by extra light coming through the filters is at most a few percent. Such an error would again tend to make our figures for the cross sections too low. A reassuring fact in this connection is that the optical cross sections are reasonable fractions of the total charge-exchange cross section. These last-mentioned systematic errors cannot be very large, else the optical cross sections would be an unreasonably large fraction of the total.

IV. ACKNOWLEDGMENTS

We are indebted to Dr. T. L. Collins for the essentials of the amplifier circuit used with the photomultiplier, and to Professor O. Oldenberg for his helpful interest throughout the project.

¹⁹ J. W. Chamberlain, *Astrophys. J.* **120**, 360 (1954).