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Excitation of Nitrogen by Protons of a Few kev Energy*†

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To aid in interpreting auroral spectra, we have investigated the excitation of nitrogen by protons of a few kev energy. The dependence of the intensity of the various spectral features on the proton current and gas pressure gives evidence about the excitation mechanisms. The conclusions are that the first negative and Meinel systems of N_2^+ are excited through charge exchange, that the first positive system of N_2 is excited, not by proton impact, but by the impact of fast neutral atoms, that allowed N I lines are excited by a single process involving dissociation and excitation, and that the Balmer lines result from charge capture directly into an excited state. As compared with excitation by electron impact, vibration is enhanced in the upper level of the first positive system, but rotational temperatures derived from the first negative system of N_2 agree with the gas temperature.

I. INTRODUCTION

SINCE Meinel's positive identification[1] in 1951 of Doppler-shifted Balmer lines in the spectrum of the aurora, it has been of special interest to investigate in the laboratory excitation of atmospheric gases by proton impact. For quantitative interpretation of auroral spectra we wish to identify the reaction or reactions which excite each spectral feature and to measure the cross sections for these reactions. Meinel and Fan have begun work on this problem[2,3]; this paper presents information on excitation of nitrogen by protons in the energy range 2–3 kev. Though the auroral protons initially have much greater energy than this, they eventually slow down and stop in the atmosphere, so that low-energy protons are also contributing to the excitation. Dieterich began work on a similar problem at this laboratory, observing the excitation of He and H_2 by H^+ and H_2^+ ions.[4]

When a beam of ions shoots into a gas, the following reactions may cause excitation: (a) direct excitation by ion impact, (b) charge exchange, which may leave either of the two resulting particles excited, (c) direct excitation by a fast atom formed by charge exchange (the atom may also be excited or reionized by collision), and (d) excitation by secondary electrons, which may have sufficient energy as formed, or may be accelerated if there are electric fields present.

General considerations put forward by Massey and others[5] show that if there is a collision reaction in which the colliding systems suffer a net energy change ΔE, then the cross section for this reaction should have its maximum value when the effective duration of the collision, T, is about the same as τ where $h/\tau = \Delta E$. The classical equivalent of this condition is that a bombarding particle will most easily excite an oscillator when the duration of the collision is about one period. The cross section should fall off slowly at higher bombarding energies, and should fall off sharply at lower energies, where the collision becomes adiabatic. Hasted and Stedeford[6] have found that this theory describes well the variation with energy of cross sections for charge exchange between various ions and atoms if one takes the time of collision to be one-half the sum of the gas kinetic radii of the colliding partners divided by their mutual velocity. Applying this information to the collisions of protons with nitrogen molecules, we find that the bombarding energy for maximum cross section for a reaction involving an energy change ΔE will be given by $E \cong 300(\Delta E)^2$ if both energies are measured in electron volts. As a consequence of this principle the character of the auroral spectrum excited by the primary stream of protons may be expected to vary with altitude, since as the incident ions slow down, they excite preferably states with lower excitation potentials. Also, as the composition of the primary stream becomes predominantly neutral, excitation by atom impact becomes important. An established example of this effect is the increase in relative intensity of the Balmer spectrum with decreasing altitude.[7]

II. APPARATUS

Figure 1 shows the arrangement of apparatus used in these experiments, which is substantially the same as that of Dieterich. Protons are generated in an rf-discharge ion source,[8] are accelerated toward a $\frac{1}{16}$-in. diam exit canal by a dc bias of 1–5 kv in the source, and are collected into a parallel beam, $\frac{1}{4}$ in. in diameter, by a lens of special design.[9] This beam travels through a combination of electric and magnetic fields serving as a mass separator; a second lens focuses it with a current of about 20 μa, onto a $\frac{1}{16}$-in. diam aperture, through

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[1] A. B. Meinel, Astrophys. J. **113**, 50 (1951).

[2] A. B. Meinel and C. Y. Fan, Astrophys. J. **120**, 360 (1954).

[3] C. Y. Fan, Phys. Rev. **103**, 1740 (1956).

[4] E. J. Dieterich, Phys. Rev. **103**, 632 (1956).

[5] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Clarendon Press, Oxford, 1952), p. 513.

[6] J. B. Hasted and J. B. H. Stedeford, Proc. Roy. Soc. (London) **A227**, 466 (1955).

[7] A. B. Meinel, Mém. soc. roy. sci. Liège **12**, 203 (1952).

[8] Moak, Reese, and Good, Nucleonics **9**, No. 3, 18 (1951).

[9] N. P. Carleton, Rev. Sci. Instr. **28**, 9 (1957).

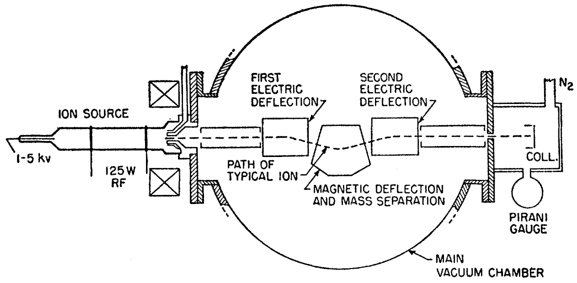


FIG. 1. Schematic diagram of apparatus.

which it passes into an observation chamber. The pressure of target gas in the observation chamber can be varied from 0.5–50 μ Hg; the ion-source pressure is 10–20 μ Hg; the vacuum maintained between these by differential pumping is about 2×10^{-5} mm Hg. Target gas enters the chamber through an all-metal valve[10] used as a controlled leak, and a Pirani gauge measures its pressure.

Light is emitted from the track of the beam itself, with very little spreading. It is analyzed with a Kipp liquid prism spectrograph, which has an $f/2$ camera and a dispersion of 100–900 A/mm, and also with an auroral grating spectrograph, which has an $f/1$ Schmidt camera, and a dispersion of 140 A/mm in the first-order infrared, and of 40 A/mm in the third-order violet. Spectra are recorded on Eastman I-N, 103a-F and Process plates. The Process plates have very fine grain and are only a factor of 3 slower than I-O plates at 4000 A. Exposure times run from a few hours in the infrared to a few minutes in the violet.

III. RESULTS

Figure 2(a) is a spectrogram of a 2400-ev proton beam in nitrogen taken on an I-N plate with the Kipp spectrograph, showing all the spectral features excited. Figure 2(b) shows the infrared portion of the spectrum with greater dispersion, obtained with the auroral grating spectrograph. The variation of the intensity of each feature with beam current and target gas pressure gives evidence as to the excitation mechanism responsible. If the excitation involves a single collision of a proton and an N_2 molecule, then the intensity should depend linearly on both the current and pressure; if the excitation is due to one of the products of a primary collision striking an N_2 molecule, then the intensity should depend quadratically on the N_2 pressure. Hence, such measurements were made, both with the spectrograph and later with a photomultiplier and interference filters. Information on specific features is as follows:

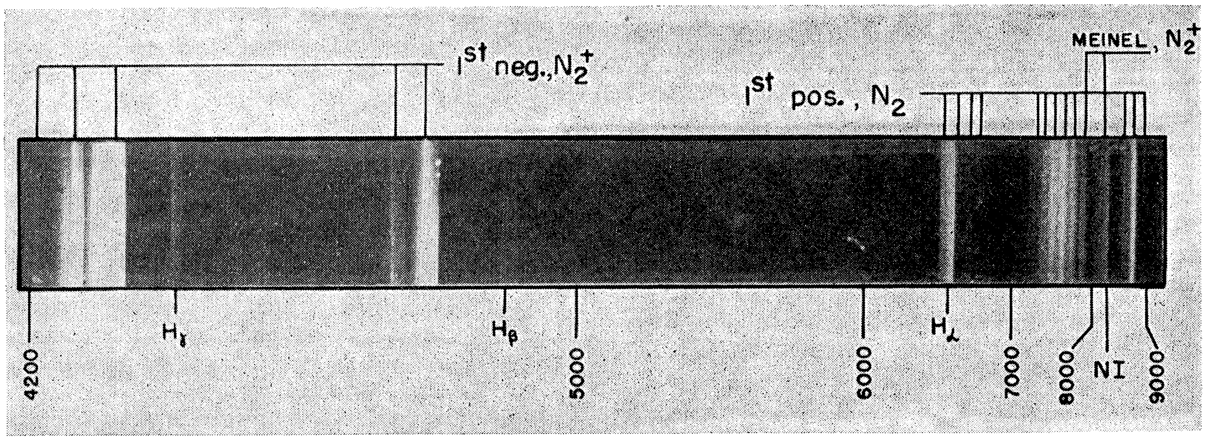
N_2^+, First Negative and Meinel Systems

The current-pressure variation shows these to be excited by a single collision. Therefore the reaction is presumably

$$H^+ + N_2 \rightarrow N_2^{+\prime} + (H^+ + e),$$

with the electron either free or captured by the proton in a bound state. The prime indicates an excited state. Frost and McDowell[11] have summarized experimental and theoretical information on the electronic structure of N_2 and N_2^+ which shows that it should be possible to remove an electron from any of three different orbitals of N_2, leaving N_2^+ in the ground state $(X\,^2\Sigma_g^+)$, the

(a)



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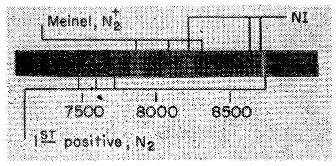

FIG. 2. (a) Spectrogram of 2.4-kev proton beam in N_2, Kipp spectrograph, exposure 2 hr, N_2 pressure 20 microns Hg. (b) Infrared region of same spectrum, auroral spectrograph, exposure 12 hr, N_2 pressure 1 micron Hg.

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[10] D. G. Bills and F. G. Allen, Rev. Sci. Instr. 26, 654 (1955).

[11] D. C. Frost and C. A. McDowell, Proc. Roy. Soc. (London) A232, 227 (1955).

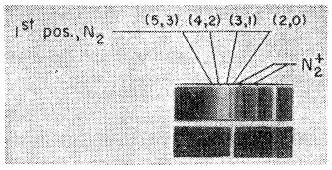

FIG. 3. Top, first positive system of N_2, excited by 2.4-kev protons. Bottom, the same, excited by electrons in the positive column of a discharge.

upper state of the Meinel bands ($A\ ^2\Pi_u$), or the upper state of the first negative bands ($B\ ^2\Sigma_u^+$). For the two processes, ionization with excitation and charge capture with excitation, the net energy changes are $\Delta E = 18.5$ ev or $\Delta E = 5$ ev, for the first negative system, and $\Delta E = 17.5$ ev or $\Delta E = 4$ ev, for the Meinel system. Massey's criterion, cited above, suggests that this bombarding energy should favor the charge-capture processes. Fan[3] reports that the Meinel system is excited only very weakly by 20-kev protons, but strongly by 300-kev protons. Since this system appears readily in the experiments here reported, it would seem (pending confirmation by comparison of absolute cross sections) that its excitation cross section, as a function of bombarding energy, has two peaks, one at low energy due to charge capture, and one at high energy due to ionization. This would be a good support of Massey's description of the collision process.

N_2, First Positive System

The intensity of this system depends approximately on the square of the N_2 pressure, indicating that a product of a primary collision must interact with a second N_2 molecule. This is to be expected, since the excitation of this system from the ground state of N_2 is an intercombination transition, only to be excited by electron exchange, which cannot take place under proton impact. Fan[3] discusses this problem, coming to the conclusion that low-energy secondary electrons, produced in primary collisions or by beam particles scattered to the walls, are responsible for the excitation. There are several reasons against applying this conclusion to these experiments, done at a lower energy than Fan's. First, there should be few secondary electrons produced at this energy (see above). Second, most of those which are produced will not have sufficient energy to excite N_2.[12] Third, and more conclusive, the emission is confined to the immediate track of the beam. Therefore, a more likely reaction is the collision of a fast H atom, formed by charge exchange, with an N_2 molecule. In this case, electron exchange can take place. As a check on this hypothesis, current and pressure variation shows that the first positive system is directly excited by H_2^+ ions (which have an electron to exchange); there is no apparent contribution from secondary processes. If the proton beam were producing secondary electrons which excited the 1st positive system, then the H_2^+ beam should do likewise. The absence of secondary electrons in the second case therefore implies their absence in the first case. Excitation by impact of fast neutral atoms is presumably of importance in the aurora, especially at lower altitudes, where the incoming particles are slowing down and spending more time in the neutral state.

Balmer Lines

The current-pressure dependence indicates that these are excited predominantly by a simple collision, with a slight contribution from a secondary process. The two reactions are presumably charge capture directly into an excited state, and excitation by a subsequent collision of an H atom, formed by charge capture in its ground state. Both these processes are presumably operating in the aurora; the continuation of the experiments reported here should give at least an estimate of the relative cross sections for the two processes.

N Atomic Lines

These are also excited by a simple collision reaction, which is most likely to be

$$H^+ + N_2 \rightarrow N + N' + H^+.$$

For this reaction, however, $\Delta E = 21.5$ ev, so that it should not be favored at these low bombarding energies. Massey and Burhop[13] state that there is no known process through which a nitrogen molecule may be dissociated into two neutral atoms by electron impact, but their evidence is not conclusive. Indeed, some recent work by Bills and Carleton[14] on adsorption of nitrogen activated by electron bombardment indicates that such a process may exist. Another possible reaction would be

$$H^+ + N_2 \rightarrow N^+ + N' + H,$$

which has $\Delta E = 22.5$ ev. There does not seem to be much reason to choose one of these processes over the other.

Distribution of Excited Molecules Among Vibrational and Rotational States

The first negative bands of N_2^+ show about the same vibrational distribution as when excited by electron impact. This is according to expectations for charge-exchange collisions at low energies in which little exchange of momentum generally takes place.[3,15] The first positive bands of N_2, however, show considerable enhancement of higher vibrational levels, as illustrated by Fig. 3. This gives an opportunity of judging from auroral spectra how much of the excitation of the first positive system may be due to fast H atoms. Unfortunately, even in good published auroral spectra[16] the first positive system in the infrared is confused by the presence of O_2 bands, both in emission and absorption

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[12] D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **A66**, 961 (1953).

[13] Reference 5, p. 263.

[14] D. G. Bills and N. P. Carleton (to be published).

[15] H. D. Smyth and E. G. F. Arnott, Phys. Rev. **36**, 1023 (1930).

[16] E.g., A. B. Meinel, Astrophys. J. **113**, 583 (1951).

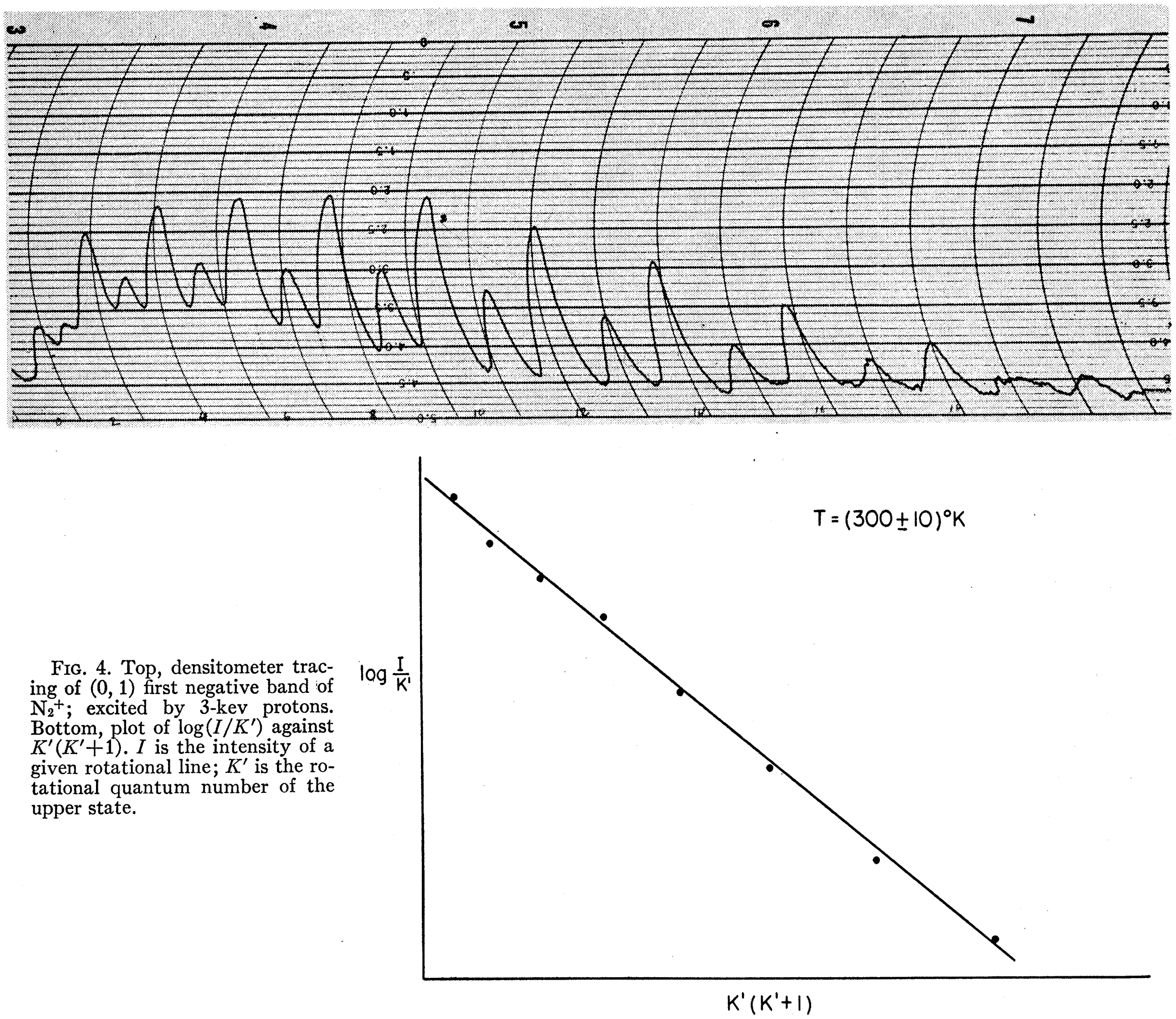

FIG. 4. Top, densitometer tracing of (0, 1) first negative band of N_2^+; excited by 3-kev protons. Bottom, plot of $\log(I/K')$ against $K'(K'+1)$. I is the intensity of a given rotational line; K' is the rotational quantum number of the upper state.

(atmospheric system). The best that can be said is that it looks as though there were some contribution by heavy particles to the excitation of the first positive system in the aurora. A systematic study of intensity distribution in this system in various auroral forms would be of interest.

The distribution of excited molecules among rotational levels has a great importance for the determination of rotational temperatures in the auroral region. As several authors have mentioned,[17] when such a determination is made on an allowed system, the rotational temperature is equal to the gas temperature only if the excitation mechanism does not alter the rotational distribution. Hence, the validity of such measurements on the first negative system in auroral spectra, presumably excited by proton impact, has been in doubt because of lack of knowledge of the effect of such excitation on the rotational distribution. Figure 4 shows a densitometer tracing of the (0, 1) first negative band as excited by 3000-ev protons, and a curve plotted according to the analysis of Vallance Jones and Harrison. The linearity of the experimental curve shows that the rotational distribution follows Boltzmann's law, and the temperature obtained from the slope of the line is 300°±10°K, which is about 10° higher than the actual gas temperature during the exposure. Thus excitation by proton impact (presumably through charge exchange) at low energies, does not affect rotational distribution appreciably.

Further work in progress includes firmer establishment of the excitation mechanisms and absolute measurements of cross sections for the various reactions.

IV. ACKNOWLEDGMENT

It is a pleasure for me to acknowledge the guidance and interest of Professor O. Oldenberg, who initiated this work.

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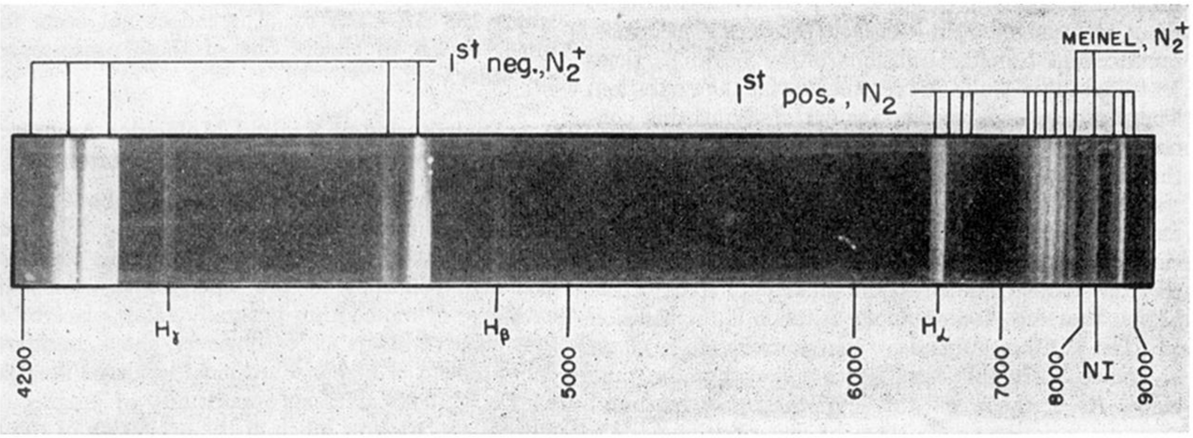
[17] See A. Vallance Jones and A. W. Harrison, J. Atmospheric and Terrest. Phys. 6, 336 (1955).

(a)



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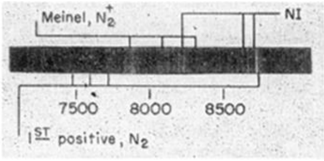

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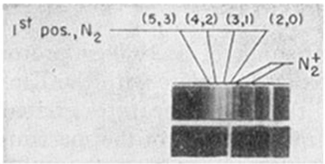

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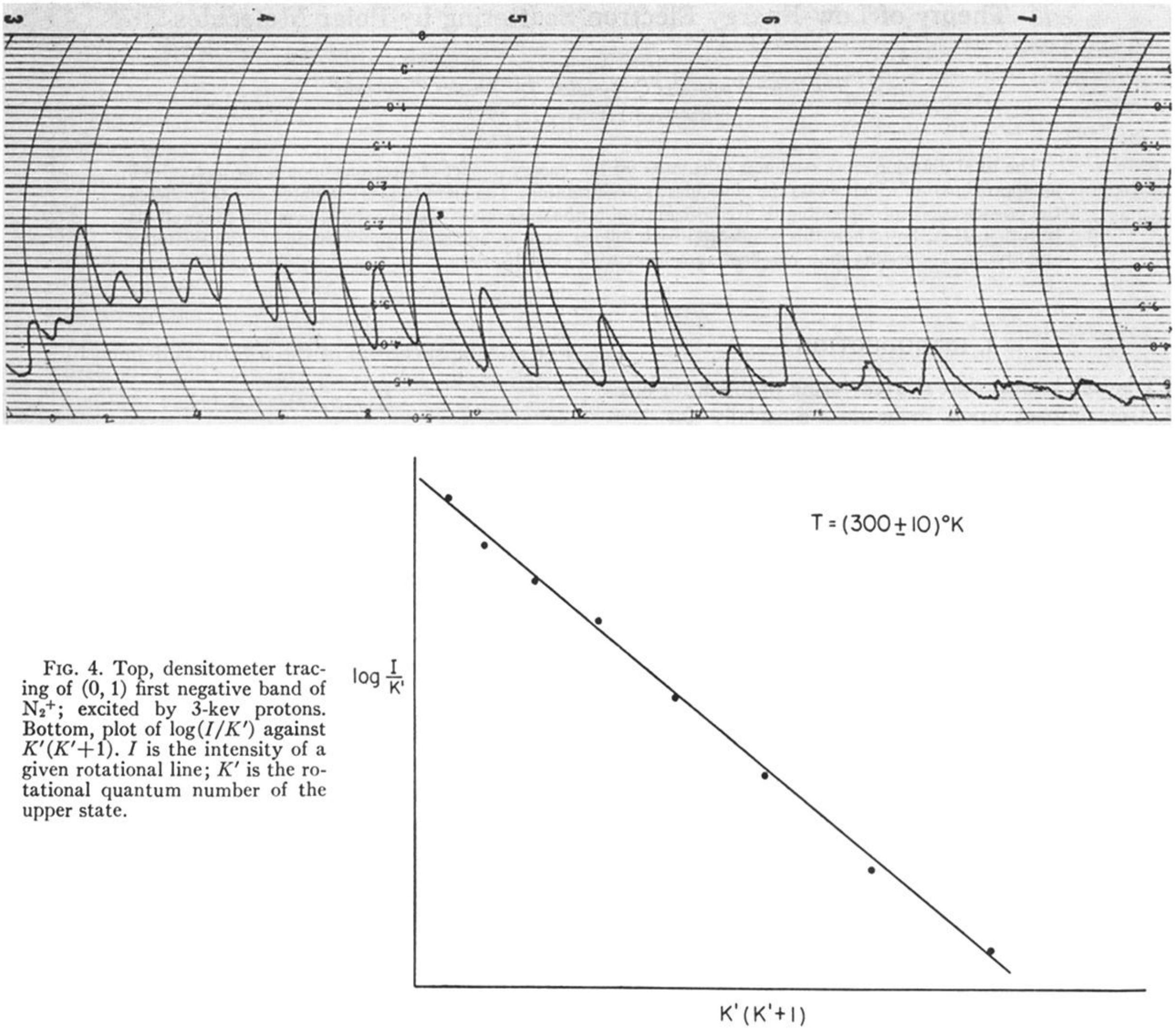

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