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Light emission from nitrogen molecules excited by the impact of 80–160 kev protons was studied over a wave length ranging from 3300 to 5400 Å. The lateral distributions of the light intensities were also measured. From the target gas pressure dependence of the light intensities and their lateral distributions, it was shown that the state $B^2\Sigma^+{}_u$ of $N^+{}_2$ was excited by a direct proton collision, and that the state $C^3\Pi_u$ of N_2, having a singlet-triplet mixing, was excited by secondary processes.

1. Introduction

The investigation of the excitation of gaseous target molecules by the impact of fast charged particles is very important for the fundamental understanding of the complex phenomena of atomic collisions. The analysis of photons, emitted from the excited target molecules at various projectile energies and target gas pressure, provides much detailed information concerning the excitation mechanism. By using this spectroscopic technique, many experiments on the excitation of nitrogen molecules by the impact of protons have been reported.[1–13] Most of these studies, however, were made below 100 kev of projectile energies and were concerned mainly with the direct excitation process in the single collision region at a low pressure.

It has been reported that the light intensity of the 3914 Å (0, 0) band of the N_2^+ first negative system is proportional to the target pressure up to 0.03 Torr,[2] and that of the 3371 Å (0, 0) band of the N_2 second positive system varies quadratically with pressure above 2×10^{-3} Torr.[12] The quadratic pressure dependence of 3371 Å is attributed to some secondary excitation mechanism. Two different mechanisms have been considered, that is, 1) the excitation of a nitrogen molecule by collision with the neutral hydrogen atom which has been formed by an electron capture of an incident

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proton, 2) the excitation by a secondary electron. The secondary electron mechanism is expected to be dominant for higher projectile energies, because the fraction of the neutral hydrogen atom is small. However, both of these two mechanisms must be taken into account for lower energies. The relative importance between these two mechanisms can not be determined by measuring only the pressure dependence of the light intensity. In this paper, we report a new method of measuring the light intensity as a function of the distance from the beam axis (lateral distribution). The principle of this method is based on the different spatial distributions of hydrogen atoms and secondary electrons.

2. Experimental Procedure

The experimental arrangement is shown in Fig. 1. The proton beam from a Cockcroft-Walton accelerator was collimated 2 mm $\times$ 2 mm in a cross section with two slits placed about 2 m apart from each other. The intensity of the beam which passed through the differentially pumped collision chamber was measured with a Faraday cup. Both the entrance and exit slits of the collision chamber were 2 mm in diameter, 10 mm in length and had a threaded inner surface for suppressing the edge scattering. The collision chamber was 200 mm in length and 56 mm in diameter, and had two quartz windows of a 5 mm thickness on both sides. (Appendix I). The pressure in the collision chamber was measured with either an ionization gauge or a Pirani gauge. The light from the excited gas molecules was measured through the quartz window in a direction perpendicular to the beam axis, and then it entered into the movable optical system. After being focused by the movable optical system, the light was spectrally analyzed with a monochromator in conjunction with a Hamamatsu R–585

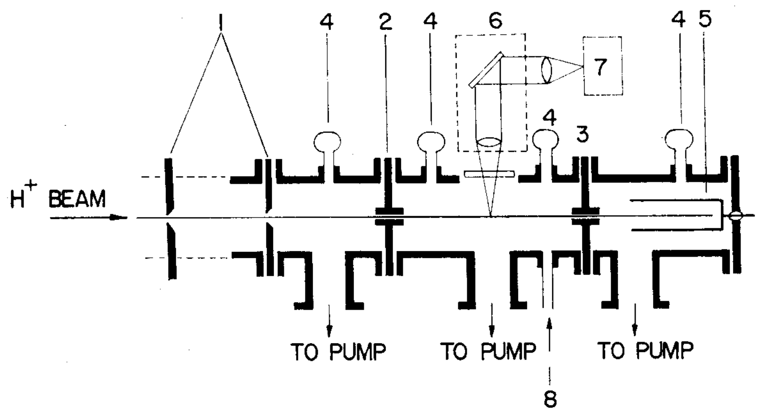

Fig. 1. Experimental arrangement: 1) collimating slit, 2) entrance slit, 3) exit slit, 4) vacuum gauge, 5) Faraday cup, 6) movable optical system, 7) monochrometor in conjunction with photomultiplier, 8) gas inlet.

photomultiplier. By using this movable optical system, scanning along both parallel and perpendicular to the beam axis could be made easily.

The spectral scans over a wave length ranging from 3300 to 5400 Å were made with a multiscaler. The analyzing wave length of the monochromator and the channel of the multiscaler were advanced by one unit simultaneously, after a definite count by the monitor photomultiplier which detected photons emitted through the quartz window on another side of collision chamber.

On the other hand, in the case of measuring the pressure dependence of the line intensities, the beam current on the Faraday cup was used for normalization. The effects of the charge exchange and the divergence of the proton beam in the course of

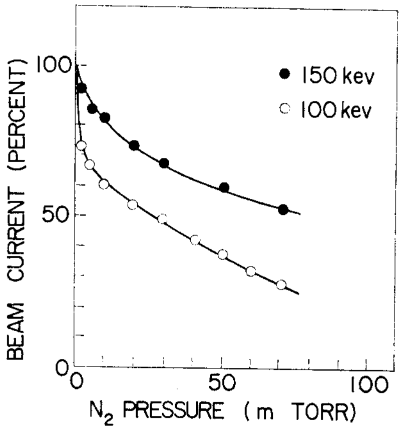

Fig. 2. Proton beam current as a function of N_2 gas pressure.

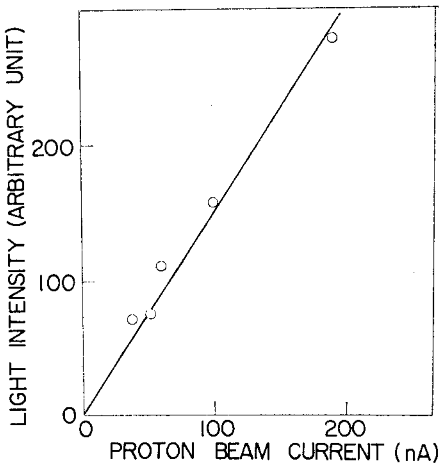

Fig. 3. Light intensity of 3914 Å produced by 150 kev proton impact on N_2 target gas at 0.06 Torr as a function of proton beam current.

passing through the gas chamber, and the contribution of secondary electrons produced in the gas chamber must be corrected carefully. The beam current on the Faraday cup will decrease as the gas pressure increases even at a constant incident beam flux. At a very low gas pressure, however, these effects could be ignored, and the Faraday cup current at our background pressure of 2×10^{-6} Torr was taken as the incident beam current itself. Above this pressure the correction factor was measured as follows. At a time when the incident proton flux was almost constant, the gas pressure was increased continuously, and corresponding variations of the Faraday cup current were measured. The beam current on the Faraday cup as a function of gas pressure is shown in Fig. 2. From these curves the correction factors were easily obtained.

In the present experiment, the energy of incident protons was 80 to 160 kev and the beam current on the Faraday cup was about 100 nA. The gas pressure in the collision chamber varied from 1×10^{-4} to 6×10^{-2} Torr.

The light intensity at a fixed gas pressure was measured as a function of the beam current. An example of the 3914 Å line measured at a pressure of 6×10^{-2} Torr is shown in Fig. 3. This proportionality means that, in the region of pressure and beam current in the present experiment, the effect of successive excitation* of the target molecules is negligibly small.

3. Results and Discussions

In Fig. 4 is shown the spectrum which was produced by the 150 kev proton incident on nitrogen gas at 6×10^{-2} Torr. In this spectral region, the vibrational bands of the N_2^+ first negative system ($B^2\Sigma_u^+ - X^2\Sigma_g^+$) and of the N_2 second positive system ($C^3\Pi_u - B^3\Pi_g$) are prominent lines just as reported by other investigators. A partial energy level diagram[14] of N_2 and N_2^+ is shown in Fig. 5. For the production of both states $B^2\Sigma_u^+$ and $C^3\Pi_u$, the contribution of a cascade transition from higher levels was reported to be negligible.[15] In the following, therefore, these states are considered to be excited directly from the ground state of N_2 ($X^1\Sigma_g^+$).

When the Frank-Condon principle is applicable to the excitation and radiation processes, the electric transition moments of both processes can be assumed to be constant for any combination of vibrational states. Therefore, the band intensity ratio between $I(j, i)$ and $I(k, l)$ is given by

$$I(j, i)/I(k, l) = (q_{0j}/q_{0k})\cdot(q_{ji}/q_{kl})\cdot(\lambda_{kl}/\lambda_{ji})^4\cdot(\tau_j/\tau_k),$$

where q's are the Frank-Condon factors, λ's the wave lengths of the radiation and τ's

* The excited molecule will make a direct transition to some other excited levels by collision with another incident projectile when the time between successive collisions is smaller than the life time of the excited state. We call these excitation processes as successive excitations.

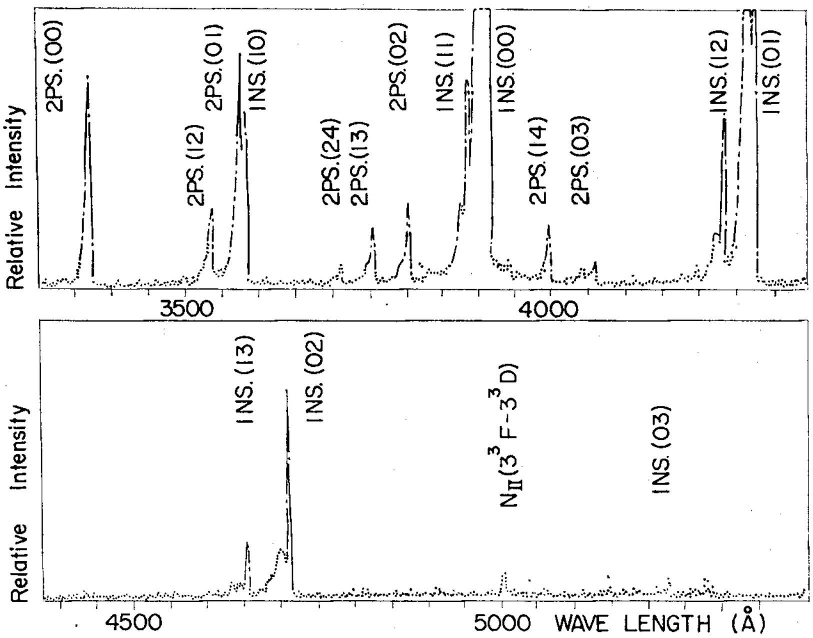

Fig. 4. Spectrum produced by 150 kev proton impact on N_2 at 0.06 Torr.

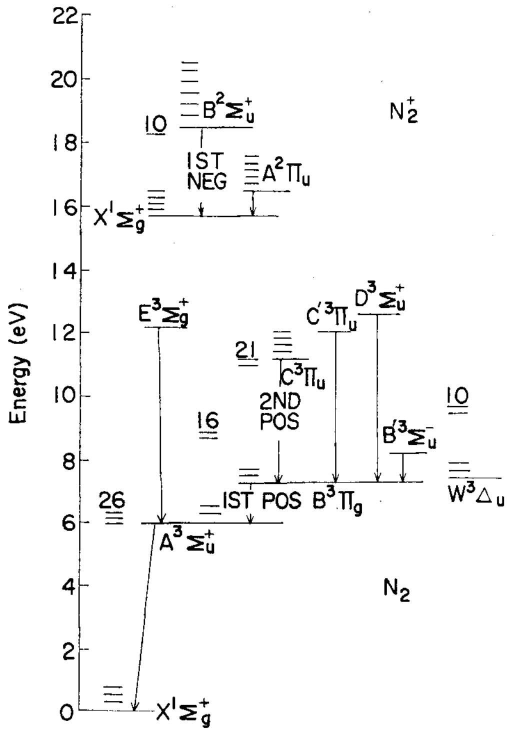

Fig. 5. A partial energy level diagram of N_2 and N_2^+ (reference 14).

Table 1 Comparison of the experimental and theoretical values of the relative band intensities in the 0–v'' and 1–v'' progressions of the N_2^+ first negative system

v' v''	00	01	02	03	10	11	12	13
wave length (Å)	3914	4278	4709	5228	3582	3885	4236	4652
PRESENT	1.0	0.31	0.045	0.0025	1.0	0.53	0.60	0.23
NICHOLLS[16]	1.0	0.28	0.051	0.0073	1.0	0.53	0.49	0.15
PILLOW[18]	1.0	0.31	0.099		1.0	0.82	0.79	0.38
PHILPOT[4]	1.0	0.29	0.056	0.007	1.0	0.55	0.49	0.19
SHERIDAN[3]	1.0	0.49	0.105					
THOMAS[7]	1.0	0.36	0.062	0.0082	1.0	0.65	0.64	0.27
DISCHARGE*	1.0	0.27	0.038	0.0058	1.0	1.15	0.423	0.346

* See Appendix II

the lifetimes of the states. In Table I, the present values of the relative band intensities of the radiations in the 0–v'' and 1-v'' progressions of the N_2^+ first negative system are compared with the theoretical values calculated by the above formula and other proton-incident experimental values.[3),4),7)] Also, the values obtained from a Geissler discharge tube filled with N_2 gas at 0.1 Torr are listed as a reference (see Appendix II).

The Frank-Condon factors, the lifetimes given by Nicholls[16),17)] and the transition probabilities given by Pillow[18)] were used in our calculation of theoretical values. In Table 1, the intensities of both 0–0 and 1–0 transitions are taken as unit intensity. The present values are in good agreement with those of Nicholls (theoretical) and of Philpot et al (experimental). In Table 2, the ratios I(1, 2)/I(0, 1) and I(1, 3)/I(0, 2) are compared with those of Nicholls. The present two values are in very good agreement with them.

The light emission divided by the product of the target gas molecule density (pressure) and the beam flux is called the emission cross section Q′. If the emitted light results from direct collisions with primary incident protons, Q' will be independent of the pressure. On the other hand, if it results from the collisions with secondary particles such as neutral hydrogen atoms or secondary electrons, Q' will be linearly dependent on the pressure. The emission cross sections as functions of pressure for the lines 3914 Å and 3371 Å are shown in Figs. 6 and 7, respectively. The cross section of 3914 Å is normalized to the absolute measurement of Hoffman et al.[11)] at 100 kev. This

Table 2 Intensity ratios of $I(1,2)/I(0,1)$ and $I(1,3)/I(0,2)$ of the N_2^+ first negative system

	$I(1,2)/I(0,1)$	$I(1,3)/I(0,2)$
PRESENT	0.13	0.25
NICHOLLS[16),17)]	0.15	0.25

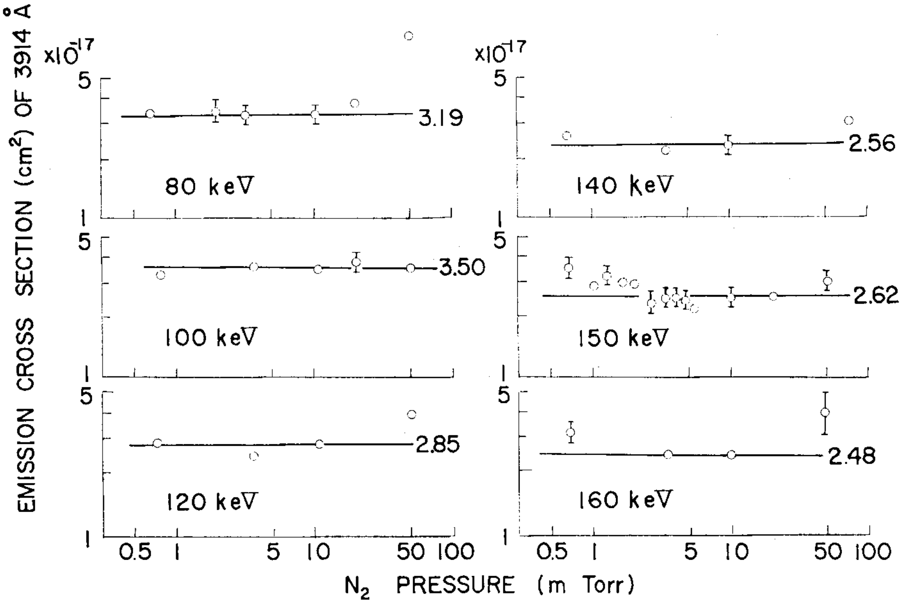

Fig. 6. Emission cross section of 3914 Å as a function of N_2 gas pressure.

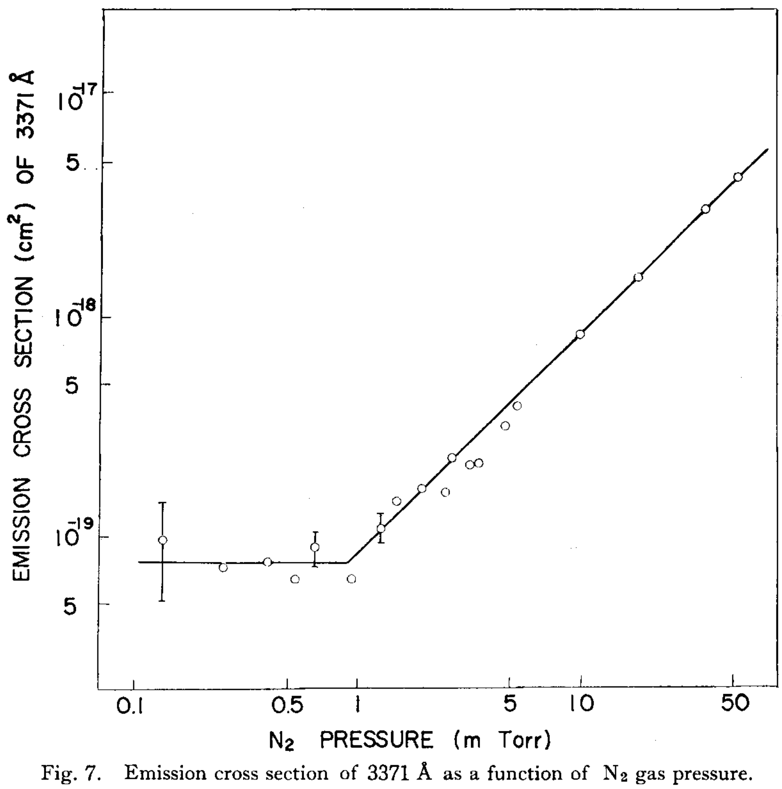

Fig. 7. Emission cross section of 3371 Å as a function of N_2 gas pressure.

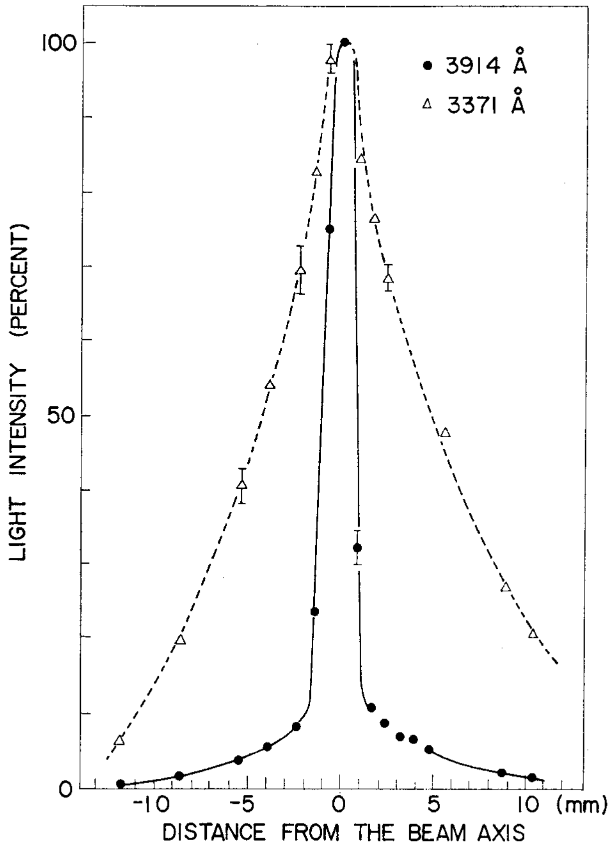

Fig. 8. Light intensities of 3914 Å and 3371 Å as a function of the distance from the beam axis.

normalization factor is also used for the cross sections of 3371 Å. The lateral distributions of these two lines are shown in Fig. 8 in which each of the maximum intensities is taken to be 100 percent.

For 3914 Å, the emission cross section is almost constant over this pressure region within experimental errors. The lateral distribution of this light source is strongly confined within the beam diameter of about 2 mm. From these results, it may be considered that only the direct excitation by protons can produce the excited $B^2\Sigma_u^+$ state.

As for the 3371 Å line, Q' is almost constant for pressure below 1×10^{-3} Torr, but above this pressure it increases linearly. This means that the excitation of state $C^3\Pi_u$ in the lower pressure region results from a direct collision with protons, but in the higher pressure region the dominant contribution to the excitation is from secondary collision processes.

Excitation of state $C^3\Pi_u$ from the ground state $X^1\Sigma_g^+$ must be accompanied by a change of the spin multiplet in the transition. No excitation of this state by a collision with H^+ will be expected if the collision process strictly follows the Wigner spin conservation rule.[19] Therefore, the direct proton excitation of the $C^3\Pi_u$ state in the lower pressure region must be attributed to a singlet-triplet mixing in this state due to the spin-orbit interaction. This interpretation is also suggested by other investigators.[12),20] In contrast to this, in the higher pressure region, both the hydrogen atom in the beam resulting from the charge exchange process and the secondary electrons produced in the ionization process of the target gas can excite the triplet state (by an electron exchange) without any violation of the spin conservation rule. It has been reported that the dominant secondary process is due to the neutral hydrogen atom at a low incident energy[1] and is due to the secondary electrons at a high incident energy.[2]

As pointed out by Thomas et al,[7] these experimental findings are consistent with the results of the energy dependence of the ionization cross section and of the charge exchange cross section of the H^+—N_2 collisions. The present experiment on the lateral distribution of light intensity supports the dominant secondary electron process in the high energy region. The lateral distribution of line 3371 Å shows a considerable amount of emission from a region far outside the proton beam. If the emission is due to the interaction with the hydrogen atom, the lateral distribution will be confined within the beam diameter like 3914 Å, because the hydrogen atom will not leave so far from the beam axis. On the contrary, secondary electrons are expected to be distri-

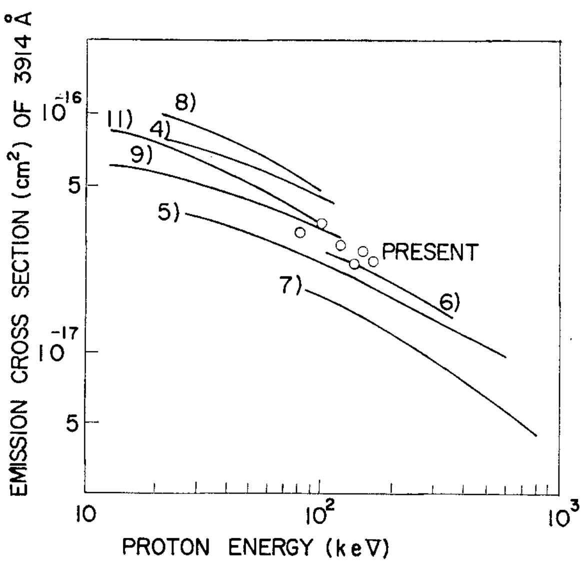

Fig. 9. Emission cross section of 3914 Å as a function of incident proton energy.

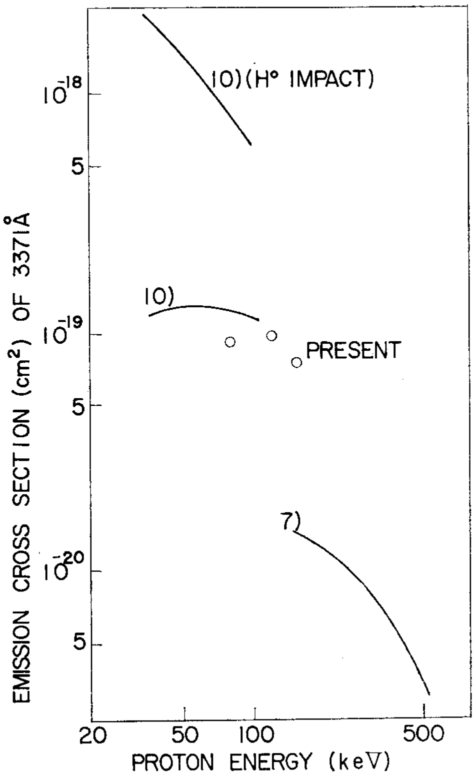

Fig. 10. Emission cross section of 3371 Å as a function of incident proton energy.

buted broadly around the beam; thus the molecules in the excited $C^3\Pi_u$ state are produced even in a region far from the beam. Therefore, we conclude that in the higher pressure region the excitation of $C^3\Pi_u$ is mostly due to the secondary electrons in this energy region.

The emission cross section as a function of energy for these two lines are shown in Fig. 9 and 10 in comparison with other experimental data.[4-11] The present energy dependence of 3914 Å seems to be in agreement with all other data within experimental errors. For an electric dipole transition, the Bethe-Born approximation predicts that the excitation cross section will have a $\ln E/E$ dependence on the incident particle energy E. The excitation cross section differs from the emission cross section only by the constant factor corresponding to the transition probability. Therefore, in this case the products of Q′ and E will have a linear relation with lnE. The products Q′E as a function of lnE for the 3914 Å line show this linearity as shown in Fig. 11. This confirms that the excitation of the state $B^2\Sigma_u{}^+$, $v=0$ takes place through a dipole transition, as has usually been recognized.

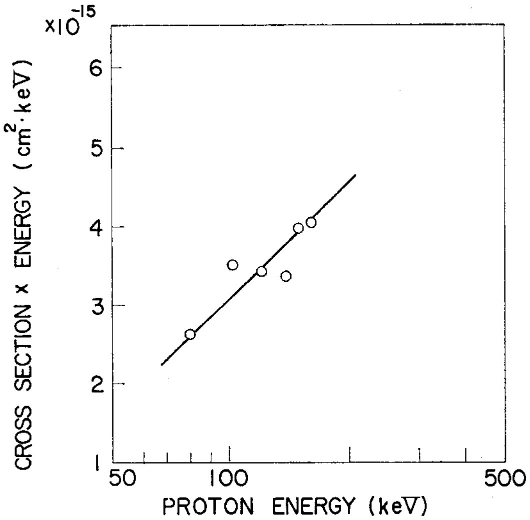

Fig. 11. Products of emission cross section of 3914 Å and incident proton energy E as a function of lnE.

As for the emission cross section of 3371 Å obtained at a low gas pressure (linear pressure dependence region), the energy dependence seems to be in agreement with that of Hoffman et al.[13] within our experimental errors (less than 10%). According to the experimental result of Dahlberg et al.,[12] the energy dependence of Q′ is lnE/E. Therefore, this state is considered to be excited by a proton collision through a dipole transition.

4. Conclusions

The light intensities and their lateral distributions of line spectra from a nitrogen gas target bombarded with 80–160 kev protons were measured in order to study the excitation mechanisms of the nitrogen molecules.

The band spectra from the N_2 first negative system and the N_2 second positive system were prominent. Relative band intensities of 0–v'' and 1–v'' progressions of the N_2^+ first negative system were in good agreement with the theoretical values.

The emission cross sections of 3914 Å of the N_2^+ first negative system (0, 0) band were almost independent of the target gas pressure up to about 6×10^{-2} Torr, and the light source of this line was confined within the beam diameter, indicating that the state $B^2\Sigma_u^+$ is excited by a direct proton collision. The light intensity of 3371 Å of the N_2 second positive system (0, 0) band varied quadratically above 1×10^{-3} Torr, and spread broadly around the beam. These results show that the state $C^3\Pi_u$ has a singlet-triplet mixing and is excited mainly by secondary processes.

There are very few experiments[21] on the excitation of molecules by the impact of projectile ions heavier than proton. The experimental studies on these lines are necessary to get a profound understanding of collision phenomena. For the investigation of such a complex collision between heavy ions and molecules, the method of measuring the spatial distribution of the light source described in this paper will be a very helpful method.

Acknowledgements

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Appendix I.

The light intensity must be corrected for the light absorption of the observation window. By using an incandescent lamp, the transmission rates of light were measured as a function of wave length for quartz, acryl and pyrex plate, each being 5 mm in thickness. The results are shown in Fig. 12. The light can transmit the quartz plate more than 95 percent over the whole range of the wave length investigated. Therefore, the quartz plate is quite suitable for the present experiment because correction for absorption is not necessary.

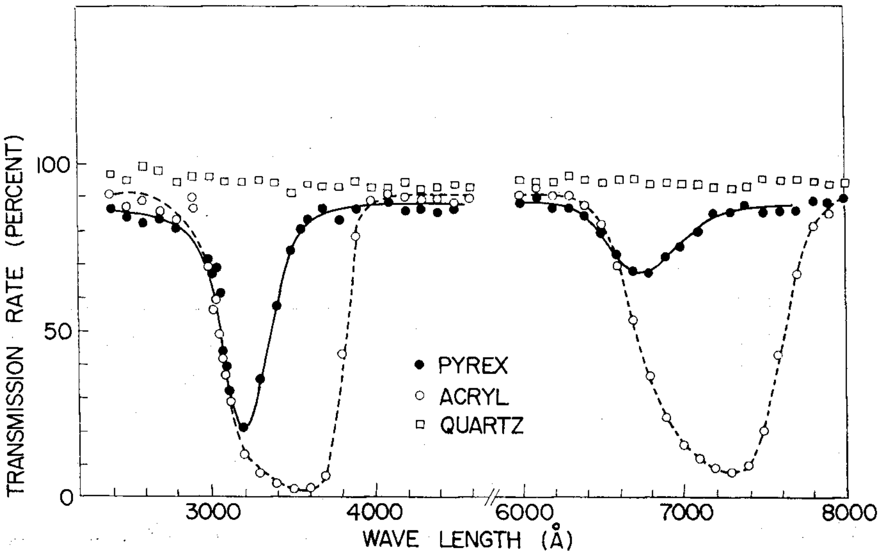

Fig. 12. Transmission rate of light in various materials (5 mm in thickness).

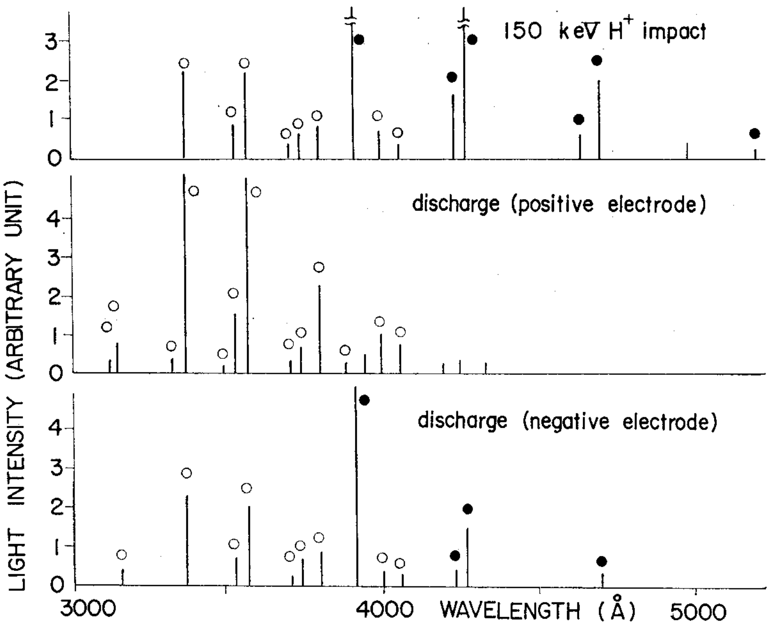

Fig. 13. Comparison of relative line intensity; 150 kev proton impact on N_2 at 0.06 Torr (upper), positive electrode (middle) and negative electrode (lower) in N_2 gas discharge at 0.1 Torr.

○ N_2 second positive system, ● N_2^+ first negative system.

Appendix II.

In Fig. 13, spectra of light from the Geissler discharge tube filled with N_2 gas at 0.1 Torr are compared with those from the target nitrogen gas excited by a proton impact. The relative light intensity at each band head is shown in this figure. The intensity ratio between 3914 Å and 3371 Å is about 18 for the case of the proton impact experiment (not shown explicitly) and about 2 for the case of the N_2 discharge. This difference may be due to the effect of an electron exchange in the collision which enhances the line of 3371 Å (see text).

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