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EXCITATION OF NITROGEN BY ENERGETIC H_3^+ IONS *

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Ions of H_3^+, accelerated to 1 MeV with a Van de Graaff accelerator, entered a differentially pumped target of nitrogen, the pressure of which could be varied up to 200 mTorr. Light generated in the collisions between the incident and target particles was viewed at 90° to the particle beam, and analyzed with a 1 m Czerny–Turner air spectrometer equipped with a 1200 l/mm grating blazed at 500 nm. We took spectra from 320 to 800 nm; spectral lines from N II and molecular bands from both N_2 and N_2^+ were identified. A high-resolution scan was made of the first negative band in N_2^+, with band head at 391.4 nm, from which we determined a rotational temperature of 350 ± 50 K, independent of pressure up to 200 mTorr. The IN bands from the ionic molecule and the monatomic lines vary linearly with pressure, while the IP and 2P bands from the neutral molecule show a squared pressure dependence.

Present experiments deal primarily with the pressure dependence of certain excitations and with the rotational temperature generated in a differentially pumped gas cell. Some of the important features of our work includes observations over a wide spectral range (320–820 nm) and the use of moderately high resolution (linewidth 0.1 nm) for most observations and better for some. H_3^+ ions were used, but it is intended to make comparisons with the effects of H_2^+ and H^+ at the same velocity.

Fig. 1 illustrates part of the observed spectrum, showing that numerous spectral features were detected. The prominent features were identified by comparison with tabulated values. What is significant for the identifications is that all expected features within our wavelength range, including sequences of band emission, were detected. Thus, in the structure of the neutral molecule three or four sequential vibrational transitions were seen.

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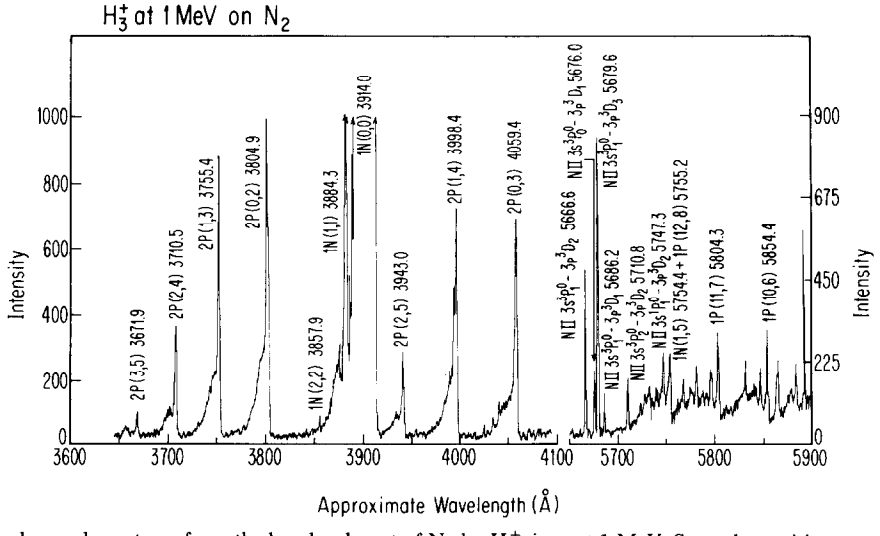

Fig. 1. Part of the observed spectrum from the bombardment of N_2 by H_3^+ ions at 1 MeV. Several transitions are identified. Note the scale change between the left and right sides of the figure.

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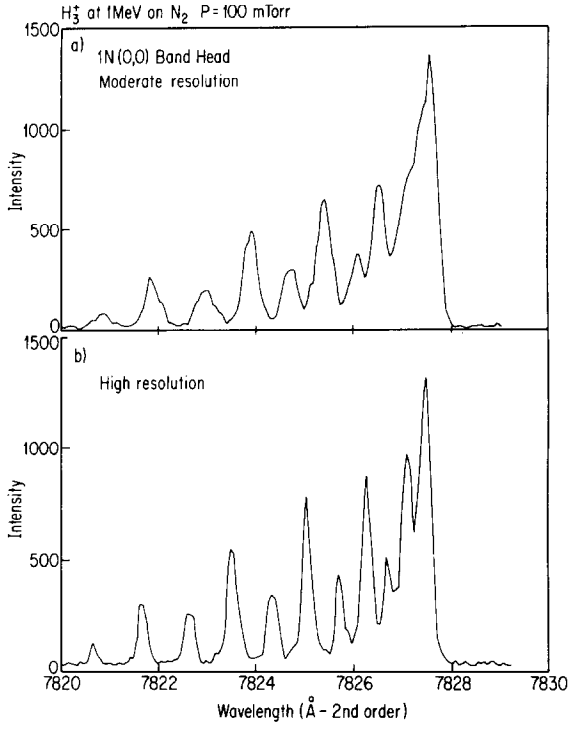

Fig. 2. Spectrum of the P-branch of the band head of the first negative band 1N(0,0) in N_2^+. (a) Moderate resolution. (b) High resolution.

The wavelength calibration of our apparatus is not well-known. As far as the raw data go, it is relatively easy to measure the relative yields of the monatomic emissions; the lines are simple in form. For the molecular features the situation is complicated. For one thing, many rotational transitions accompany a single vibrational transition. Consequently, the spectral shape of a branch is a sensitive function of the experimental resolution. This is readily seen from figs. 2a and b, which show the P-branch of the first negative 1N(0,0) band in the molecular ion. The difference between fig. 2a and fig. 2b is in the resolving power that was used. Furthermore, the width of the molecular patterns varies considerably from vibrational transition to vibrational transition. Because of these factors, it is not easy to deduce the relative populations of the molecular states. It is not always clear from the literature how other investigators measured their reported yields.

On the other hand, one can find the pressure dependence of the molecular parts of the spectrum by setting the spectrometer to a convenient wavelength, and measuring the yield as a function of the pressure. Here other effects enter.

One must, of course, know the number of incident particles at each measurement. That number is conventionally found from the charge which the incident particles deposit in a collector. With a gas target, three complications arise. First, there are charge-changing collisions in the target, so the collected charge is not simply related to the number of incident particles. This is especially important in an experiment such as ours, in which the incident particle (H_3^+) may be fragmented into several particles on passing through the target. These fragmentation and/or charge-changing events are functions of the pressure. Second, the presence of gas near the collector means that ionization may occur there, with drift of unwanted charges of either sign to the collector. Again, this effect is pressure-sensitive. Third, as the apparent current to the collector changes, the time it takes to obtain the aliquot of charge which is used to relate one measurement to another also changes, and this affects the background contribution, which is proportional to the time. This is of some importance for low target pressures, for then the yields are small.

To circumvent these problems, we measured the apparent beam current $i(p)$ as a function of target pressure and then made appropriate corrections. The results appear in fig. 3, where the relative yields from one of each of the monatomic, molecular and molecular ion transitions are displayed. A number of additional results with similar characteristics were also obtained.

The curves for the monatomic and molecular ion contributions are similar in shape, essentially linear with pressure at first, and then showing saturation.

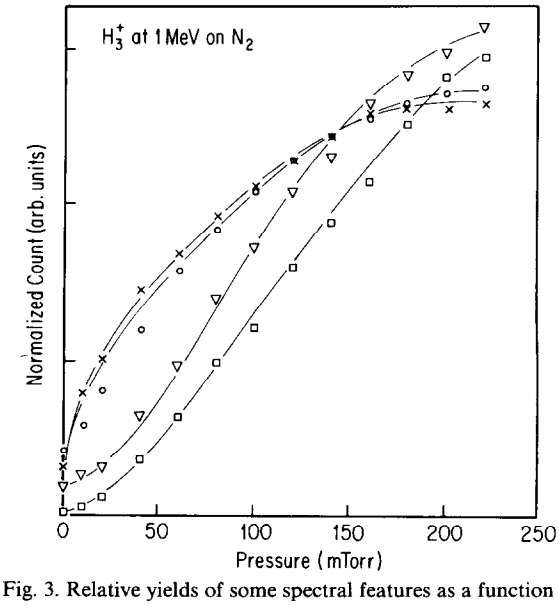

Fig. 3. Relative yields of some spectral features as a function of pressure. Note that the zeros have been displaced so as to clarify the behaviour of the several features near the origin, ×: Monatomic transition N II $3p^3D_2-3d^3P_3^0$. ○: transition 1N(0,0) in N_2^+. ▽: First positive band 1P(4,1) in N_2. □: Second positive band 2P(0,0) in N_2.

Curves from the neutral molecule show a different pressure dependence. This suggests that the monatomic and molecule-ion curves come from the same underlying physical process, whereas the excitation of the neutral molecule involves a separate kind of event. Indeed, the neutral curves, of which only one is shown in fig. 3, suggest that a second-order pressure dependence is present. Somewhat similar shapes for the 1N bands in N_2^+ and the first positive bands in N_2 have been reported before [1–13], although our resolution and method of analysis differ from those of the papers cited.

The emissions from the neutral molecule are interesting for another reason. The ground state of N_2 is a singlet, whereas the excited states whence the observed emissions emerge are triplets. Hence, as has been pointed out by others [3–5,8,10], one expects these radiations to follow from electron exchange processes.

To account for the pressure dependence of the various excitations, we propose the following as a possible source effect.

(1) The incident ions create excited N_2^+ in direct collisions. Those collisions also generate fast electrons.
(2) The fast electrons strike N_2 molecules, creating $N_2^{+\prime}$ plus fragments $N + N^{+}*$ and secondary electrons.
(3) The secondary electrons interact with N_2 to produce N_2^* (triplet state) and cold electrons.
(4) The cold electrons combine with N_2^+ to make more N.

A detailed analysis of this model shows that it leads to yields of excited N_2^+ and N^+ that are proportional to the pressure, while the yield of excited N_2 varies as the squared pressure. Somewhat similar suggestions have been made by refs. [13] and [14], except that they do not mention the multivalued electron-energy function. We believe that our data substantiate this hypothesis.

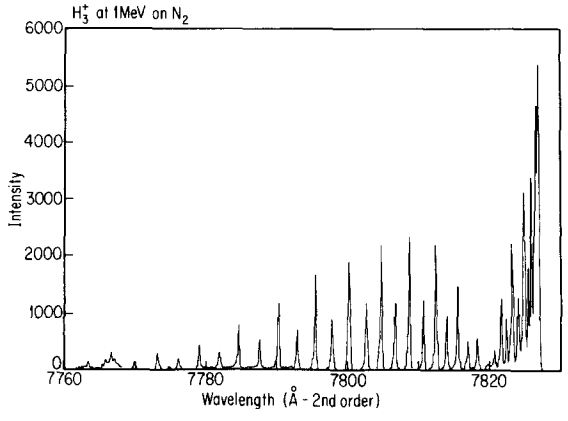

Fig. 4. High-resolution study of the 1N(0,0) band head in N_2^+.

In fig. 4 we show a high-resolution scan of the 1N(0,0) vibrational band in N_2^+. Analysis of these data led us to conclude that the rotational temperature was 350 ± 50 K, independent of pressure. This is close to values reported by others [4,14–16] who used proton bombardment. However, ref. [17] reported a temperature of 3500 K when the incident particles were low-energy Li ions, while refs. [9], [18] and [19] suggest that rotational equilibrium does not occur when low-energy protons excite the nitrogen. This might come about because the cross section for momentum transfer is large for small velocities.

A more complete report on this work is in preparation.

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