

Effective Rotational Temperatures of $N_2^+(X)$ Produced by Electron-Impact Ionization on N_2

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Synopsis. The laser-induced fluorescence of molecular ions, $N_2^+(X, {}^2\Sigma_g^+)$, was measured under the controlled electron-impact ionization of N_2 in the thermal gas jet. The effective rotational temperature of $N_2^+(X)$ increased from 320 ± 5 K to 367 ± 15 K for the incident electron energy from 750 eV to 17 eV.

The rotational distribution of ions should be a valuable probe of the dynamics of the ionization process. Nitrogen molecule is suitable for this purpose. The ionization process to the B state of N_2^+ has been studied upon the electron-impact ionization-excitation.^{1,2)} At high electron energies, $\Delta|J|=1$ is obeyed; at low electron energies (<100 eV), however, $\Delta|J| \geq 3$ transitions occur through multipole transitions.²⁾

The rotational distribution has been measured on the X state of N_2^+ produced by controlled electron impact in the thermal gas flow^{3,4)} and the supersonic beam^{5,6)} using laser-induced fluorescence (LIF) technique. The incident electron energies previously used, however, were only 60³⁾ and 100^{3,4)} eV in the former and 50,⁵⁾ 80,⁵⁾ and 300⁶⁾ eV in the latter.

In this study, we measured the rotational distribution of $N_2^+(X)$ in the thermal gas jet at the wide electron energy range of 17–750 eV, and determined the effective rotational temperature (T_R) of $N_2^+(X)$ in order to discuss the ionization process.

Experimental

The apparatus has been described in detail previously.⁴⁾ In brief, the sample gas was jetted into a collision chamber through a multicapillary array and was crossed perpendicularly with an electron beam. The excitation light was produced by a N_2 laser pumped tunable dye laser (Moletron UV24+DL14). The signal was accumulated using gated detection electronics with a boxcar integrator or a 100 MHz photoncounter. We calculated the number density of ions to be about 10^8 ions cm^{-3} from the electron-beam current (500 μA), the sample gas pressure (0.13 Pa), the ionization cross section (1.0×10^{-16} cm^2),⁷⁾ and the fly-out rate coefficient from the laser-irradiation region (1.4×10^6 s^{-1}). There are comparable number of ions in the ion trap techniques.^{8,9)} About 0.2 counts of the LIF signal per laser pulse were necessary to obtain a good spectrum with a signal to noise ratio of 15 in several hours of accumulation; the typical intensity of scattering light from the laser was 0.04 counts.

Results and Discussion

A typical excitation spectrum of the $B(v'=0)-X(v''=0)$ of $N_2^+(X)$ produced by controlled electron impact on N_2 at the incident electron energy of 28 eV is shown in Fig. 1. The relative rotational line intensities, $\ln\{I_{N',N''}/(N'+N''+1)\}$, were plotted versus

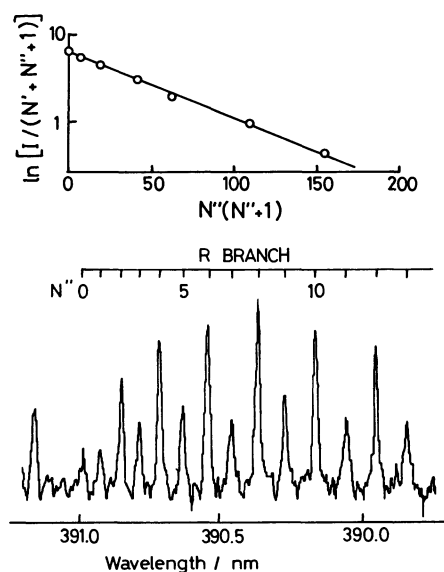


Fig. 1. Excitation spectrum of $N_2^+(X)$ (lower) and the rotational line intensity plot of even N'' lines of R branch (upper). Electron energy was 28 eV and N_2 pressure was 0.13 Pa.

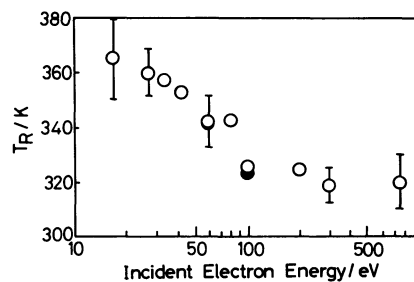


Fig. 2. The electron energy dependence of the effective rotational temperature (T_R) of $N_2^+(X)$ produced by electron-impact ionization on N_2 .

●: Allison et al., ○: this work.

$N''(N''+1)$, where N' and N'' are the upper and lower rotational quantum numbers of the transition, as also shown in Fig. 1. The plots were linear at all incident electron energies and gave the effective rotational temperature (T_R) of $N_2^+(X)$.

The electron energy dependence (17–750 eV) of T_R is shown in Fig. 2. The results obtained at 60 and 100 eV agree with the previous values³⁾ within experimental uncertainties. T_R was constant above 100 eV, and increased below 100 eV up to 367 ± 15 K at 17 eV; T_R at 17 eV was higher by about 50 K than T_R at 750 eV.

In order to estimate effects of collisional relaxation, fly-out rate of the ions from the laser-irradiation

region has been measured through the intensity changes of the LIF signal at various delay times (0–2 μ s) of the laser after the end of the pulsed electron beam. The LIF signal decayed exponentially. The first-order decay analysis leads to a fly-out rate coefficient of $1.4 \times 10^6 \text{ s}^{-1}$ and a characteristic fly-out time of 0.71 μ s. These values agree with the free diffusion rate estimated from the gas dynamics; a nitrogen molecular ion with the root-mean-square velocity of 511 m/s at 20 °C flies across the laser beam with the radius of 0.5 mm in about 1 μ s. The characteristic collisional time at a pressure of 0.13 Pa is about 100 μ s. These findings indicate that the observed spectrum was not disturbed by collisional relaxation. Cascade contributions are unimportant as the radiative lifetime of the A state (10 μ s) is longer than the characteristic fly-out time and the ionization-excitation cross section to the B state is small⁷⁾

In the ionization process for a homonuclear diatomic molecule such as H_2 and N_2 , the selection rule of a rotational transition indicates ΔN to be an even number, e.g., $N''=N$, $N''=N \pm 2$, and $N''=N \pm 4$, where N and N'' are the rotational quantum number of N_2 and N_2^+ (X).^{10,11)} Change of the rotational quantum numbers between neutral and ionic molecules balances the angular momentum of the ejected electron, since the total angular momentum is conserved. In the photoionization of H_2 , $\Delta N=0$ and $\Delta N=2$ transitions were observed with rotationally resolved photoelectron spectroscopy and $\Delta N=2$ transition was accounted for coupling between the p and f photoelectron waves.¹¹⁾ The electron-impact ionization may be accompanied by an ejected electron with the large angular momentum through the multipole transition.¹²⁾ Therefore, the increase of T_R at lower electron energies is due to the increase of larger N transitions. At higher electron energies, $\Delta N=0$ transition is predominant and T_R of N_2^+ (X) do not change from that of N_2 .

The electron energy dependence of T_R of N_2^+ (X) in this paper is considerably different from that of N_2^+ (B) previously reported.¹³⁾ This may be caused by the fact that the molecular orbital of the ejected electron for N_2^+ (X) is $\sigma_g 2p$, while that for N_2^+ (B) is $\sigma_u 2s$. The detailed analysis requires an experimental determination of the partial cross-section for each ΔN transition from a single rotational level.

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