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EXCITATION OF ATOMIC NITROGEN BY ELECTRON IMPACT

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Excitation of Atomic Nitrogen by Electron Impact

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Abstract

Absolute cross sections have been measured for the excitation of the NI($\lambda\lambda$1134Å, 1164Å, 1168Å, 1200Å, 1243Å, and 1743Å) multiplets by electron impact on atomic nitrogen. The λ1134Å and λ1200Å cross sections are large, reaching 1.8×10^{-16} cm^2 and 2.3×10^{-16} cm^2 at their peaks, respectively. The magnitude of these cross sections is about a factor of two lower than the values reported previously by the authors, due to the discovery of a fault in the apparatus. The presence of vibrationally excited molecular nitrogen in the discharged gas is confirmed, and its effect on the measurements is discussed. The ratio of the oscillator strengths of the λ1200Å and λ1134Å resonance transitions is measured to be 2.6 ± 0.3. The branching ratio for the NI(λ1311Å/λ1164Å) multiplets is measured to be 2.8 ± 0.3. Striking differences in the distribution of intensity between the spectra of atomic nitrogen and molecular nitrogen excited by energetic electrons suggest an optical method for measuring the density of atomic nitrogen in the upper atmosphere.

INTRODUCTION

Radiation from atomic nitrogen in the vacuum ultraviolet is commonly observed in gas discharges, in the polar aurora, and in the terrestrial airglow. The extent to which these emissions are due to each of several processes (electron impact on atomic nitrogen, electron impact dissociation of molecular nitrogen, photodissociation of molecular nitrogen, etc.) is not known, due in part to the fact that many of the relevant cross sections have not been measured. Knowledge of all the electron-impact excitation cross sections would be an important contribution to the understanding of these natural and laboratory phenomena, and would also provide a tool for analysis of the composition of the upper atmosphere.

Cross sections for dissociative excitation of N_2 by electron impact have been measured by several investigators[1-3] and are now fairly well established. Cross section measurements for direct excitation of atomic nitrogen by electron impact have been reported only in previous work by the authors[4-7]. In this paper we wish to revise values previously given and to report the completed work. We also suggest a means of measurement of the atomic nitrogen density in the upper atmosphere, based on these cross sections.

EXPERIMENTAL

The basic apparatus used in this experiment has been described in detail in an earlier paper[4]. Only several important modifications to this system will be discussed here. The most important modification was the redesigning of the optically thin helium-discharge light source used in the atomic density measurements. In order to be more certain that the $\lambda1200\overset{o}{A}$ triplet of atomic nitrogen emitted by the lamp was optical thin, the new light source was formed as a pinched "T" (see Figure 1). High purity helium flowed through the discharge region at a pressure of about 0.15 torr, and a trace of nitrogen was added through a leak valve. The axis of the RF discharge was perpendicular to the optical axis of the experiment, and the discharge was constricted at the point of observation to provide a thin layer of excited atoms. The direction of the flow of gas was away from the monochromator, so that atoms produced were quickly swept out of the field of view. The new light source was found to be much more stable than the old one, so that it was possible to scan the monochromator at high resolution slowly over the $1200\overset{o}{A}$ triplet to observe its profile. In the absence of self-reversal, the intensity ratio of the three lines should be 6:4:2. A sample scan of the triplet is shown in Figure 2.

A new photomultiplier tube (EMR 541GX) which has a quantum efficiency about three times that of the old one was installed, thus improving the counting statistics significantly.

An attempt was made to use a constriction in the gas flow tube just downstream from the dissociating discharge in order to increase the density of nitrogen atoms in the experimental region. While this

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technique worked well for an experiment with atomic oxygen it had no appreciable effect on the atomic nitrogen experiment, and was not used.

The experiment was conducted as before, by counting photons with the discharge on and off, alternately. The counting period was 30 seconds in each mode, which was short enough to hold drifts in pressure and beam current to 1 or 2% at most. The difference in photon count rate per unit of electron beam current per unit of pressure, with appropriate adjustments for the loss of N_2 density due to dissociation and absorption of the atomic signal by ambient N atoms in the collision chamber, was assumed to be the contribution from direct electron impact excitation of atomic nitrogen.

Values for the absolute cross sections were obtained by comparing the observed photon count rates for each excitation per unit of electron beam current per unit of gas density in the collision chamber per unit of solid angle subtended by the entrance slit of the monochromator with count rates for dissociative excitation processes whose cross sections are well known. The known cross section was taken to be 11.0×10^{-18} cm^2 for dissociative excitation of molecular hydrogen by 100 eV electrons to produce Lyman alpha ($\lambda 1215.6 \overset{o}{A}$) radiation,[8,9] or, equivalently, 6.1×10^{-18} cm^2 for dissociative excitation of molecular nitrogen by 100 eV electrons to produce the NI($\lambda 1200 \overset{o}{A}$) triplet. The latter (relative to the former) was confirmed in the course of this experiment. For dissociative excitation the gas density was determined using an ionization gauge (Varian Millitorr) which was calibrated against a McLeod Gauge. For direct excitation, atomic nitrogen densities were determined by observing resonance absorption of the NI($\lambda 1200 \overset{o}{A}$) triplet in the collision chamber.

The relative spectral response of the optical system was determined by the method of molecular branching ratios, using the LBH band system of molecular nitrogen[10] and Lyman-band fluorescence in hydrogen deuteride.[11,12]

For the resonance absorption measurement the relation between the absorption of light and the column density of the atomic gas was calculated from the formulas of Mitchell and Zemansky.[13] The oscillator strength used was the mean of the values of Lawrence and Savage[14] and of Lin, Parkes, and Kaufman,[15] giving f = 0.12, 0.08, and 0.04 for the three lines of the λ1200Å triplet. The source and absorption lines were assumed to have pure Doppler profiles. The temperature of the light source was taken to be 400°C, and the temperature of the absorbing gas to be 320°C. The latter figure is based on the fact that the discharged gas is cooled by passage through 30 cm of room-temperature Pyrex tubing before entering the collision chamber, whose temperature is about 25°C above room temperature. The light source temperature was estimated as quite low for an RF discharge, since the discharge was very weak (barely visible to the eye). The pyrex tube in which the discharge took place remained only warm to the touch, requiring no special cooling.

It was assumed that the λ1134Å and λ1200Å signals produced in direct excitation of N atoms were subject to resonance absorption within the collision chamber, but that the same emissions produced in dissociative excitation were not, due to the very broad Doppler profiles for such excitation. We attempted to estimate the possible error incurred through this assumption, using examples of emission line profiles for dissociative excitation of metastable atoms calculated by Wells.[16] In the worst case (λ1200Å, high gas density, relatively slow atoms) the cross section reported

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here would be too high at 100 eV by about 7% relative to its value at dissociative threshold. In all likelihood the true correction is much smaller.

The cross sections reported in this paper for the excitation of atomic nitrogen are more than a factor of two below those that we have reported previously. This revision reflects the discovery of a malfunction in a now-discarded electronic counter that was used during the resonance-absorption measurements of the atomic density. This instrumental malfunction resulted in an underestimate of the atomic nitrogen density and hence in an overestimate in the magnitude of the absolute cross sections.

EXCITED STATES IN THE DISCHARGED GAS

Unlike the O_2 molecule, molecular nitrogen has no low-lying (less than 6 eV) electronically excited states. The principal metastable species formed in the discharge are the $A^3\Sigma_u^+$ state with a lifetime on the order of 2 seconds, and the $a^1\Pi$ state, with a lifetime of about 100 microseconds. The $a^1\Pi$ state does not live long enough to reach the collision chamber from the discharge, and both states appear to be efficiently de-excited by wall collisions. In an auxiliary experiment, we have examined the translational energy spectra of metastable species produced by a pulsed beam of electrons impacting on N_2 in the present apparatus. The resulting spectra are completely consistent with a model based on the Boltzman velocity distribution and free-molecule expansion from a point source. We also observed the decay of optical radiation from the $a^1\Pi$ state (LBH bands) as excited within the collision chamber by a pulsed electron beam. The decay of radiation is characterized by a lifetime ($\sim$ 24 microseconds) consistent with thermal motion out of the field of view of the monochromator and with no (< 5%) reflection from the chamber walls. (The wall surface is a Dag graphite dispersion.) These tests lead us to conclude that there is no significant component of the discharged gas in electronically excited molecular states. We expect that the same probably applies to excited states of the nitrogen atom, and the flowing afterglow work of Lin and Kaufman[17] supports this view.

However, there is evidence of vibrational excitation of the ground $N_2(X^1\Sigma)$ state. We observe changes in photon emission rates in the $N_2(a^1\Pi - X^1\Sigma)$ LBH band system when the discharge is turned on, indicating that the apparent cross sections for excitation to the various vibrational

levels of the upper $a^1\Pi$ state are altered by the discharge. Table I indicates the observed changes, and compares them with LBH Franck-Condon factors. The comparison clearly implies that the discharge-on excitation involves initial vibrational levels above $v = 0$. This effect confirms the results of numerous other studies[18,19] on the presence of vibrational excitation in discharged N_2 which is long-lived with respect to wall de-excitation.

The presence of the vibrationally-excited ground state molecules implies that there will be a weak onset of dissociative excitation with the discharge-on at one or two electron volts lower energy than the threshold for dissociative excitation with the discharge off. This will tend to exaggerate the height of apparent atomic cross sections in the energy range 16 - 40 eV, particularly where the atomic signal is small. We are not able to assess quantitatively the effect of such an early threshold on our atomic cross sections, and have not attempted to correct for it. The fact that the observed cross sections rise rapidly to a peak from the expected threshold for direct excitation argues that the early dissociation effect accounts only for a small part of the total.

RESULTS

A. Atomic Cross Sections

Figures 3 through 6 show excitation functions for direct excitation of the atomic nitrogen multiplets at 1134$\overset{o}{A}$, 1200$\overset{o}{A}$, 1243$\overset{o}{A}$, and 1743$\overset{o}{A}$. The error bars shown represent $\pm$ 1 standard deviation in the reproducibility of the data. Note that the λ1200$\overset{o}{A}$ excitation function shape differs slightly from that reported previously.[4] The absolute values of the peak cross sections and the estimated total probable errors at the peak for these transitions and those at 1164$\overset{o}{A}$ and 1168$\overset{o}{A}$ are summarized in Table II. Contributing to the estimated errors are a number of factors: (1) the statistical error in the photon count rate, generally 10% or less, (2) statistical error in the measurement of atom density in the collision region, 10%, (3) stated possible error in the dissociative cross sections and optical oscillator strengths used as standards, about 15% each, (4) possible error in the estimated temperature of the light source, contributing up to 10%, (5) uncertainty in the length of the column of absorbing gas in the collision chamber, 10%, (6) uncertainty about the extent of the contribution to the measured cross sections of vibrationally excited ground-state molecules, 10% to 50%, and (7) uncertainty in the relative spectral response calibration, 0% to 15%. Moreover, the probable error increases as higher electron energies are approached, as indicated by the error bars and scatter in the data in Figures 3 - 6.

All cross section values given in this paper are total cross sections for electron impact, including all fast cascade transitions. They are calculated on the assumption that the spatial distribution of the excited radiation is isotropic (the radiation is observed at right angles

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to the electron beam). This assumption is justified only when the upper state of the transition is an S state; hence the NI cross sections are subject to revision if the radiation is eventually determined to be strongly anisotropic.

B. Dissociative Excitation Cross Sections

Table II also contains three cross sections measured for dissociative excitation of molecular nitrogen, which were determined in the course of measuring atomic cross sections. The dissociative cross section for the $\lambda1134\text{Å}$ transition has not been reported previously in the literature. Its excitation function is shown in Figure 7. The transitions at 1164Å and 1311Å result from excitation of the same $2p^23d(^2D)$ upper state, and the branching ratio between them has been in dispute. Labuhn[20] found that the ratio of transition probabilities $A(1311\text{Å})/A(1164\text{Å})$ was $2.8 \pm 10\%$, while Aarts and de Heer[3] arrived at a value of 0.4. The present results are in (fortuitously) exact agreement with Labuhn's value. The ratio found by Aarts and de Heer is presumably erroneous due to their method of obtaining the relative spectral response of their instrument at short wavelengths.[11]

C. $2s^22p^3(^4S) - 2s2p^4(^4P)$ Oscillator Strength

A simple modification of the apparatus permitted us to compare the ratio of the transition probabilities for the $\lambda1134\text{Å}$ and $\lambda1200\text{Å}$ transitions. A pyrex tube, about a foot long and one inch in diameter, was place along the optical axis between the light source and the monochromator. Nitrogen gas which had passed through the discharge flowed into the tube at its center and out its open ends. By comparing

the ratio of the absorption of the two resonance triplets, the ratio of the oscillator strengths was determined. Figure 8 is a graph of the resultant data, taken at various pressures. Horizontal error bars indicate $\pm$ 1 standard deviation in the statistical error for the weaker absorption. Vertical error bars were too small to illustrate. The graph exhibits a distinct curvature, considerably in excess of what can be accounted for on the basis of deviation from linearity expected in resonance line absorption (solid line). The excess deviation is probably due to absorption features in the discharged molecular nitrogen which do not appear in the undischarged nitrogen, since the vibrational temperature of the discharged gas is quite high[21],[22].

The ratio of oscillator strengths is obtained as the extrapolated initial slope of the graph,

$$\frac{f(\lambda\ 1200\overset{o}{A})}{f(\lambda\ 1134\overset{o}{A})} \quad = \quad 2.6 \pm 10\%$$

This is in good agreement with the results of Lawrence and Savage,[14] who found the ratio to be 2.7, and with Labuhn,[20] who obtained the value 2.6.

A comparison of the spectra produced by electron impact on N and N_2 shows striking differences in the relative magnitudes of the signals observed at 1134Å, 1200Å, and 1493Å. Spectra taken in the present apparatus in the discharge-on mode at 16 eV (atomic excitation) and in the discharge-off mode at 100 eV (molecular excitation) are shown in Figure 9. The vertical scale (photon counting rate) is uncorrected for relative spectral response of the optical system. In the atomic spectrum, the two resonance transitions are overwhelmingly larger than any of the other emission features. In the dissociative spectrum, the 1134Å feature is much reduced, while the 1493Å feature is enhanced, relative to the triplet at 1200Å. This difference in the relative magnitudes of the 1134Å, 1200Å, and 1493Å multiplets can serve as the basis for the measurement of atomic nitrogen density in the upper atmosphere.

Although several papers[23] based on the chemistry of nitric oxide have calculated atomic nitrogen densities for various daytime conditions, there is only one report, by Ghosh et al.,[24] of a direct mass spectrometer observation of atomic nitrogen. However, this result has not been confirmed in a large number of other mass spectrometer experiments, which have established an upper limit on the atomic nitrogen concentration of less than 1/4 percent of the ambient N_2 density. Conventional mass spectrometers are limited in their ability to measure N in the presence of large quantities of N_2 due to interference problems caused by the dissociative ionization of N_2. Two optical approaches to this problem are discussed below.

A sounding rocket equipped with a scanning monochromator or

suitable photometers can measure the overhead brightness of uv emission features as a function of altitude. In the absence of significant resonance trapping of radiation and under static conditions, the local photon emission rate is proportional to the derivative of the overhead brightness with respect to altitude. The local emission rate E for an atomic nitrogen emission feature of wavelength λ as a function of altitude z can be written as

$$E^i(z) = k(\lambda) \quad (n_1 F_1^{\ i}(z) + n_2 F_2^{\ i}(z))$$

where k is a function of wavelength containing the collection and detection efficiencies of the experimental optics, and n is a species number density. Subscripts 1 and 2 refer to atomic and molecular nitrogen, respectively, and superscript i labels the particular emission feature. F is a photon production efficiency given by

$$F^i(z) = \int \sigma^i(E) \ \phi(E,z) \ dE$$

where σ is the total electron impact cross section as a function of the electron impact energy E, and ϕ is the flux of energetic electrons $(cm^{-3} \ eV^{-1})$. Selecting a pair of NI emission features ($\lambda 1134\overset{o}{A}$ and $\lambda 1200\overset{o}{A}$ or $\lambda 1200\overset{o}{A}$ and $\lambda 1493\overset{o}{A}$), the equations for local emission rates can be solved simultaneously to yield the density of atomic nitrogen (n_1) and the relative spectral response $(k(\lambda^i)/k(\lambda^j))$ of the instrument if the flux ϕ and the density of molecular nitrogen (n_2) as a function of altitude are known.

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Unfortunately, this optical method suffers from several significant liabilities. First, modern monochromators suitable for satellite or sounding rocket experiments exhibit comparatively low sensitivity below 1150Å. This characteristic makes it difficult to observe the λ1134Å feature effectively. Perhaps this problem can be dealt with or mitigated by the use of windowless optics or by more careful treatment of the lithium fluoride windows and surfaces commonly employed. Second, the use of the emission feature at 1493Å has the disadvantage that it is heavily overlapped by strong LBH band radiation. Although the NI line intensity may be obtained in principal by a suitable deconvolution process, the absolute accuracy of the result may be poor. Finally, it is supposed that only electron impact on atomic and molecular nitrogen contributes to the excitation of the NI vacuum ultraviolet emission features and that no other sources (e.g., resonance scattering of solar photons) are significant. Hence, detailed fore-knowledge of the geophysical situation is required in practice before the results could be considered unique.

An alternative method for measuring the atomic and molecular nitrogen densities in the upper atmosphere would involve the use of a programmed electron gun in a suitable open chamber and the examination of the optical signal produced at 1200Å with the electron beam energy at 16 eV, 100 eV, and off. This method takes advantage of the striking differences in the shape and absolute magnitude of the atomic and molecular excitation cross sections. A comparison of the excitation functions for the λ1134Å feature shown in Figures 3 and 7, and the absolute magnitude data presented in Table II, for example, illustrates the point amply. A review of available flight instrumentation shows that atomic nitrogen concentrations

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as low as 10^5 atoms/cm^3 can be measured in this manner. The presence of molecular nitrogen does not affect the atomic measurements because the energy spectrum of the exciting electrons is controlled so that the experiment clearly discriminates between the atomic and molecular excitation channels.

ACKNOWLEDGEMENTS

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Table I

Variation in LBH Excitation Cross Sections Caused by Discharge

Band Observed* (v',v'')	Signal Change due to Discharge	Franck–Condon Factors			
v' v'' =		0	1	2	3
(0, 2) 0	+ 65%	.043	.152	.249	.250
(1, 1) 1	− 3%	.116	.193	.080	.001
(2, 0) 2	− 23%	.171	.097	.003	.108
(3, 0) 3	− 27%	.183	.012	.076	.068
(4, 0) 4	− 25%	.160	.006	.096	.000
(5, 0) 5	− 18%	.122	.046	.047	.034

* Electron energy 23 eV

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Table II

Measured Cross Sections

Multiplets		Target Species	Electron Energy (eV)	Cross Section (cm²)	Probable Error
Wavelength	Array				
1134Å	$2s^2 2p^3 (^4S^0) \leftarrow 2s2p^4 (^4P)$	N	22 (peak)	1.8×10^{-16}	40%
1164Å	$\leftarrow 2p^3 (^2D^0)$	N	18 (peak)	$3. \times 10^{-18}$	50%
1168Å	$\leftarrow 2p^3 (^2D^U)$	N	18 (peak)	1.2×10^{-17}	60%
1200Å	$\leftarrow 2p^3 (^4S^0)$	N	32 (peak)	2.3×10^{-16}	35%
1243Å	$\leftarrow 2p^3 2s' (^2D)$	N	17 (peak)	$5. \times 10^{-18}$	60%
1744Å	$\leftarrow 2p^3 (^2P^U)$	N	17 (peak)	1.2×10^{-17}	50%
1134Å		N_2	100	1.14×10^{-18}	25%
1164Å		N_2	100	1.14×10^{-19}	20%
1311Å	$2p^3 (^2P^0) \leftarrow 2p^3 3p (^2D)$	N_2	100	3.17×10^{-19}	30%

Figure 1   Diagram of the light source used in the resonance absorption measurement of atomic density.

Figure 2   Scan of the NI(λ1200Å) triplet produced by the light source.

Figure 3   Excitation function for electron impact on atomic nitrogen to produce the λ1134Å triplet. The peak of the curve corresponds to a cross section of 1.8×10^{-16} cm^2.

Figure 4   Excitation function for electron impact on atomic nitrogen to produce the λ1200Å triplet. The peak of the cross section corresponds to 2.3×10^{-16} cm^2. Above 50 eV the curve is drawn to follow a $\ln(E/10.4)/E$ dependence (Born approximation).

Figure 5   Excitation function for electron impact on atomic nitrogen to produce the λ1243Å multiplet. The peak of the curve corresponds to a cross section of 5×10^{-18} cm^2. The solid line is drawn by hand and is very approximate.

Figure 6   Excitation function for electron impact on atomic nitrogen to produce the λ1743Å multiplet. The peak of the curve corresponds to a cross section of 1.2×10^{-17} cm^2. The solid line is drawn by hand and is very approximate.

Figure 7   Excitation function for electron impact on molecular nitrogen to produce the λ1134Å triplet of atomic nitrogen. The peak of the curve corresponds to a cross section of 1.14×10^{-18} cm^2.

Figure 8   Relation between resonance absorption at 1134Å and 1200Å for various gas pressures. The solid line indicates the deviation from linearity which can be accounted for solely on the basis of resonance absorption by N atoms.
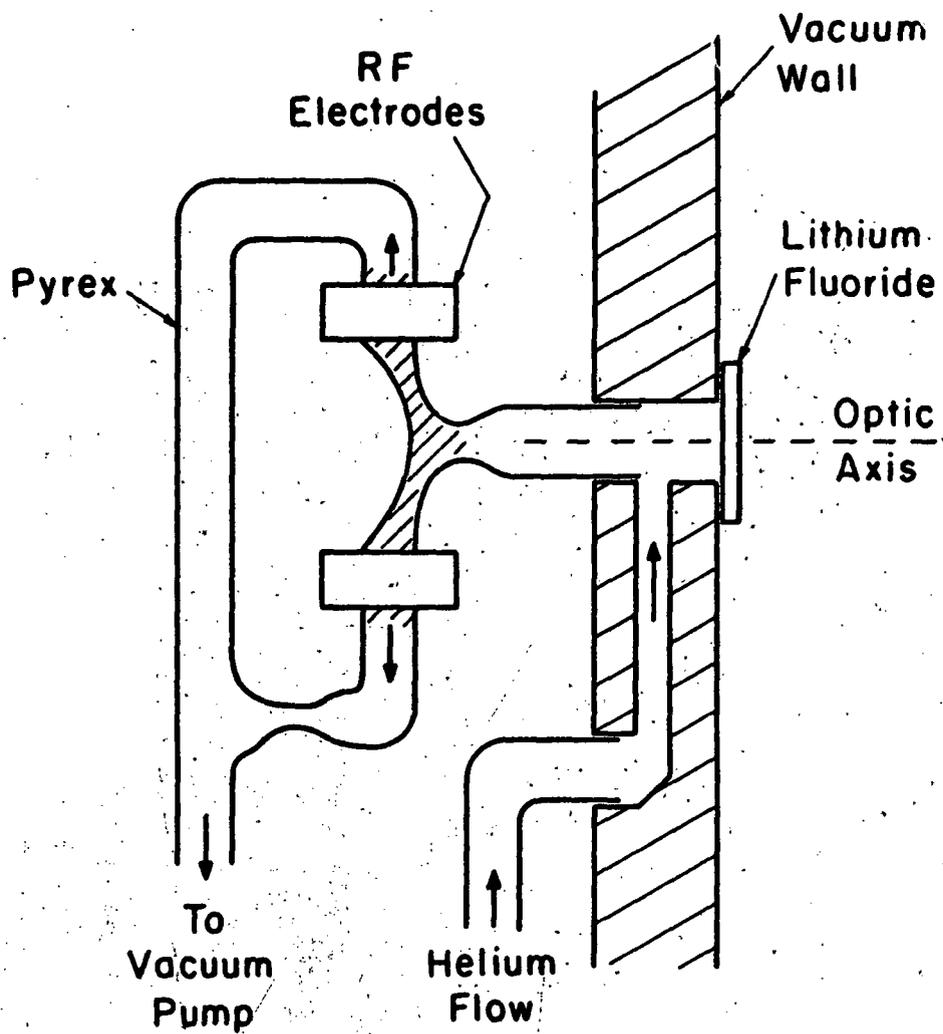
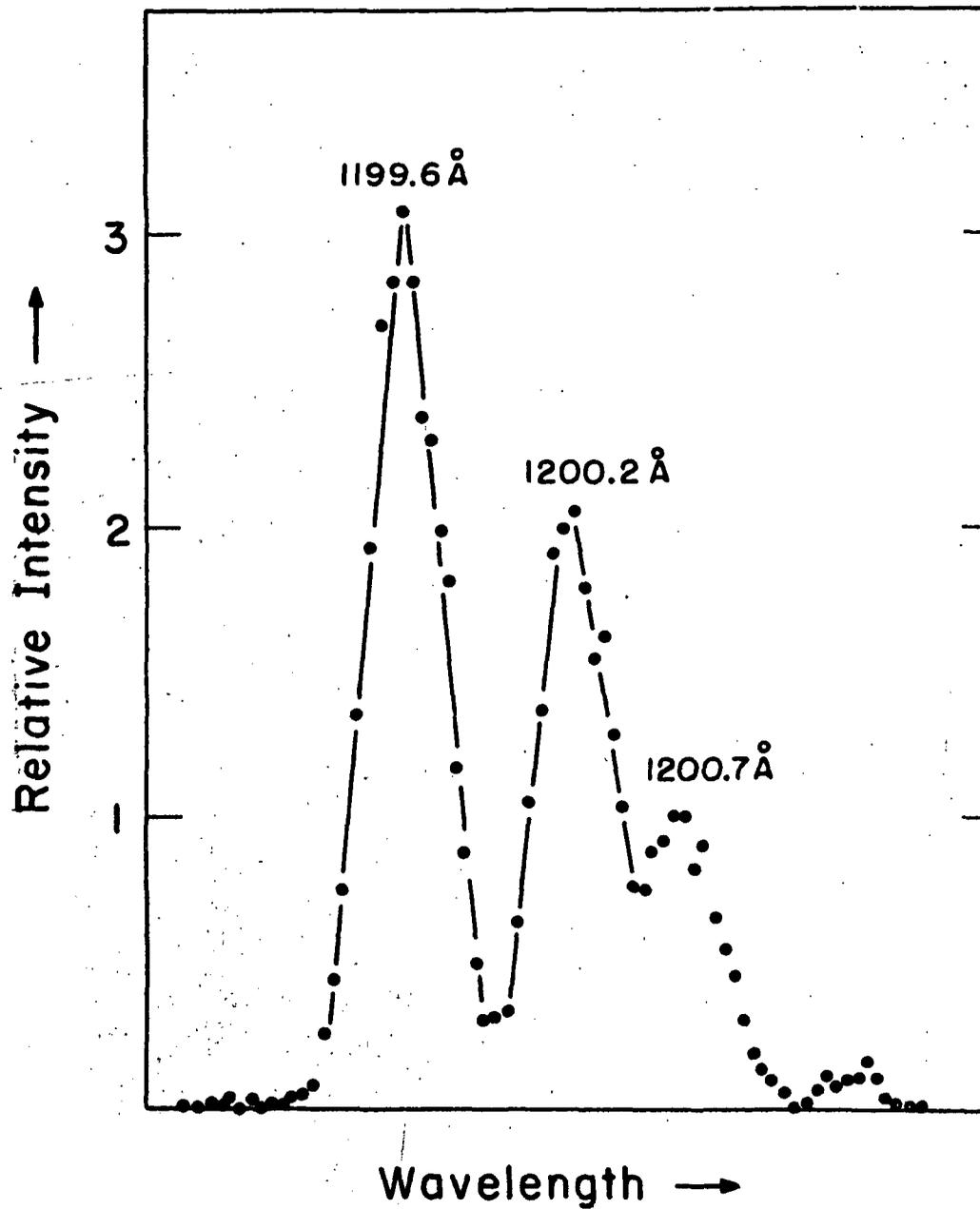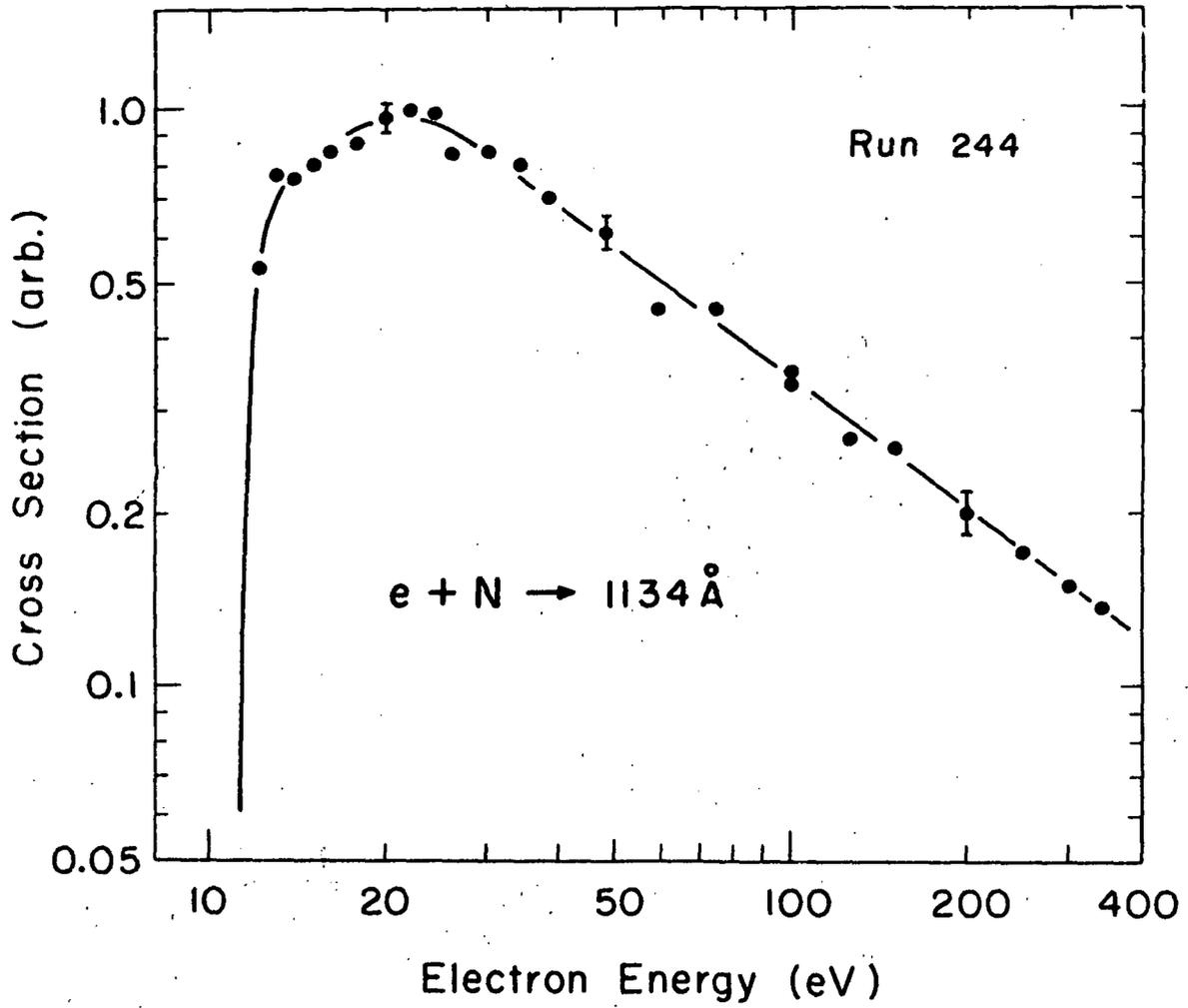Figure 9 Comparison of the vacuum uv spectra of atomic and molecular nitrogen.
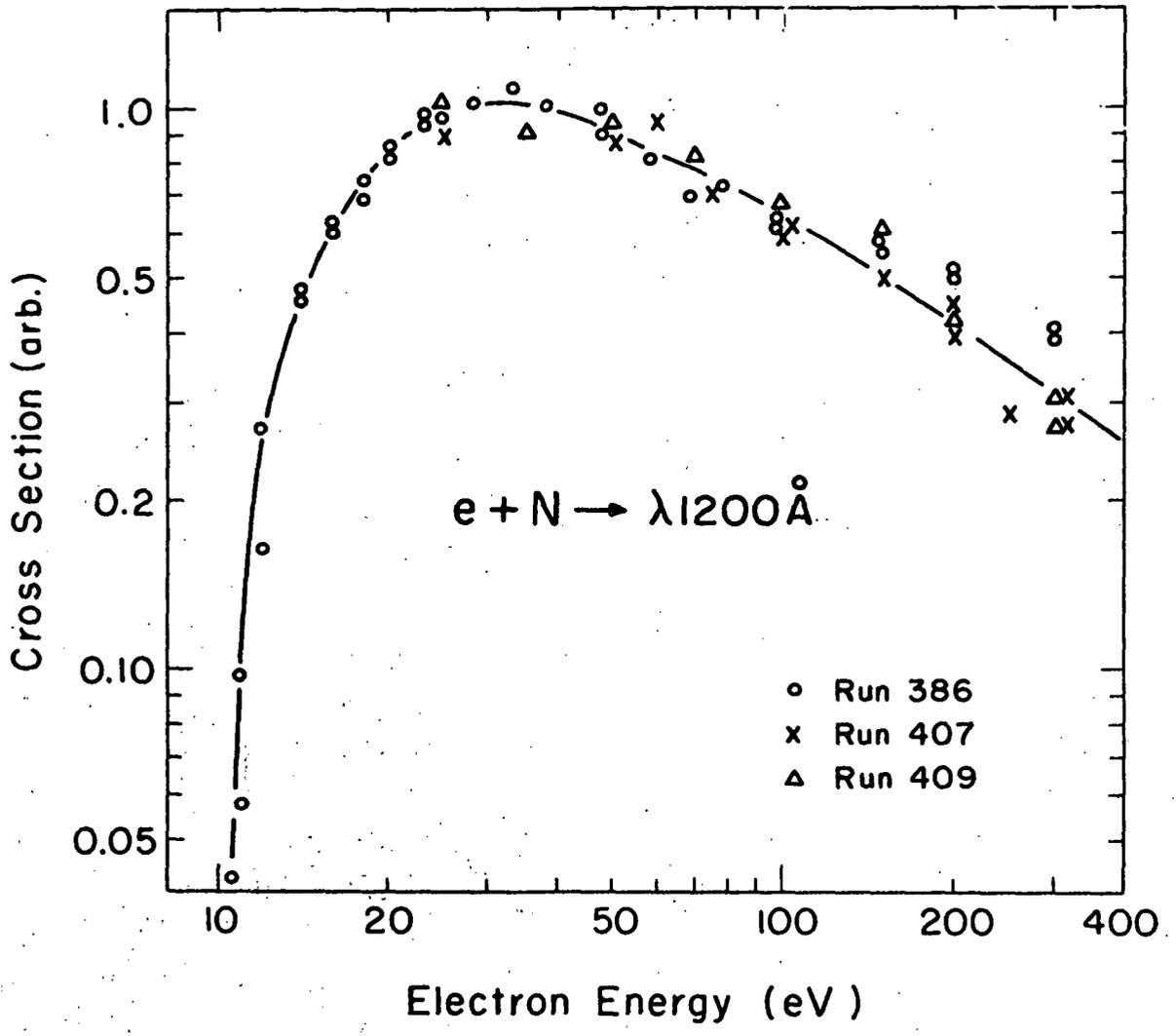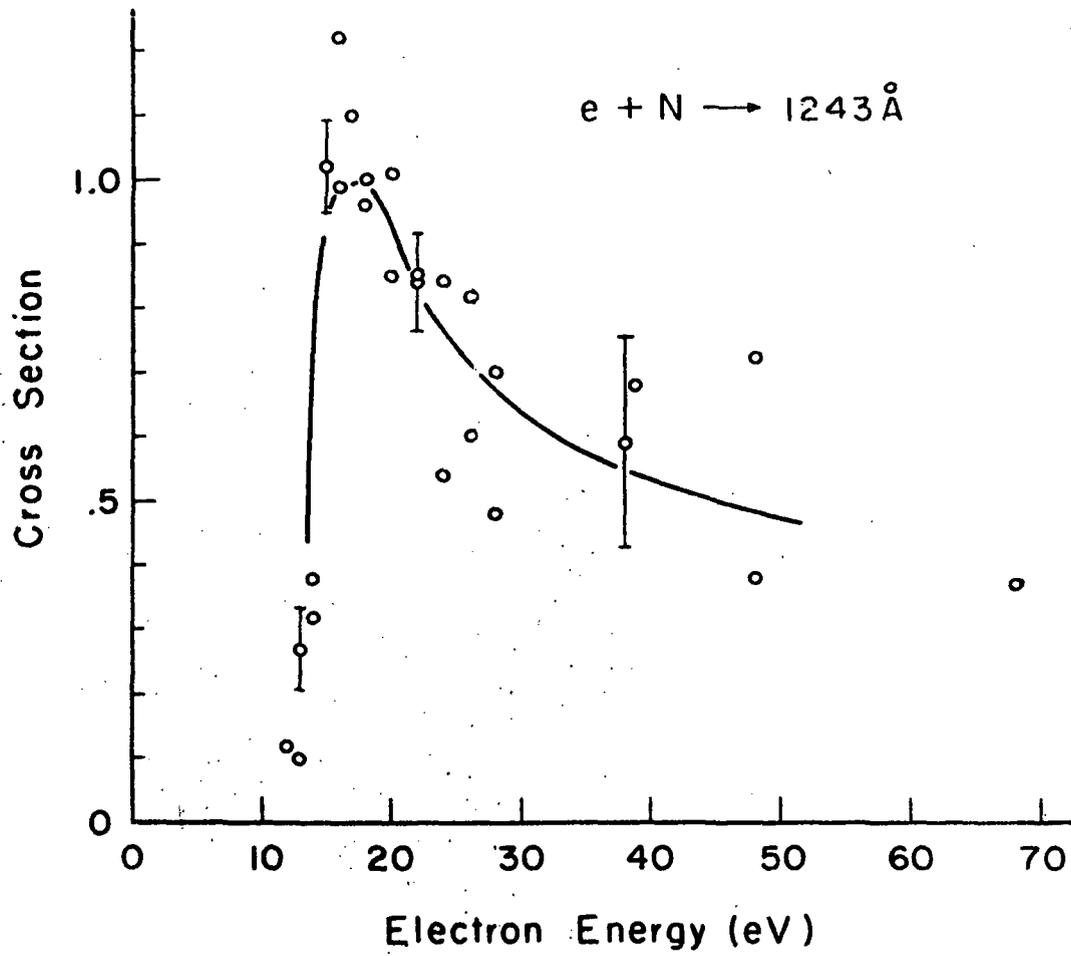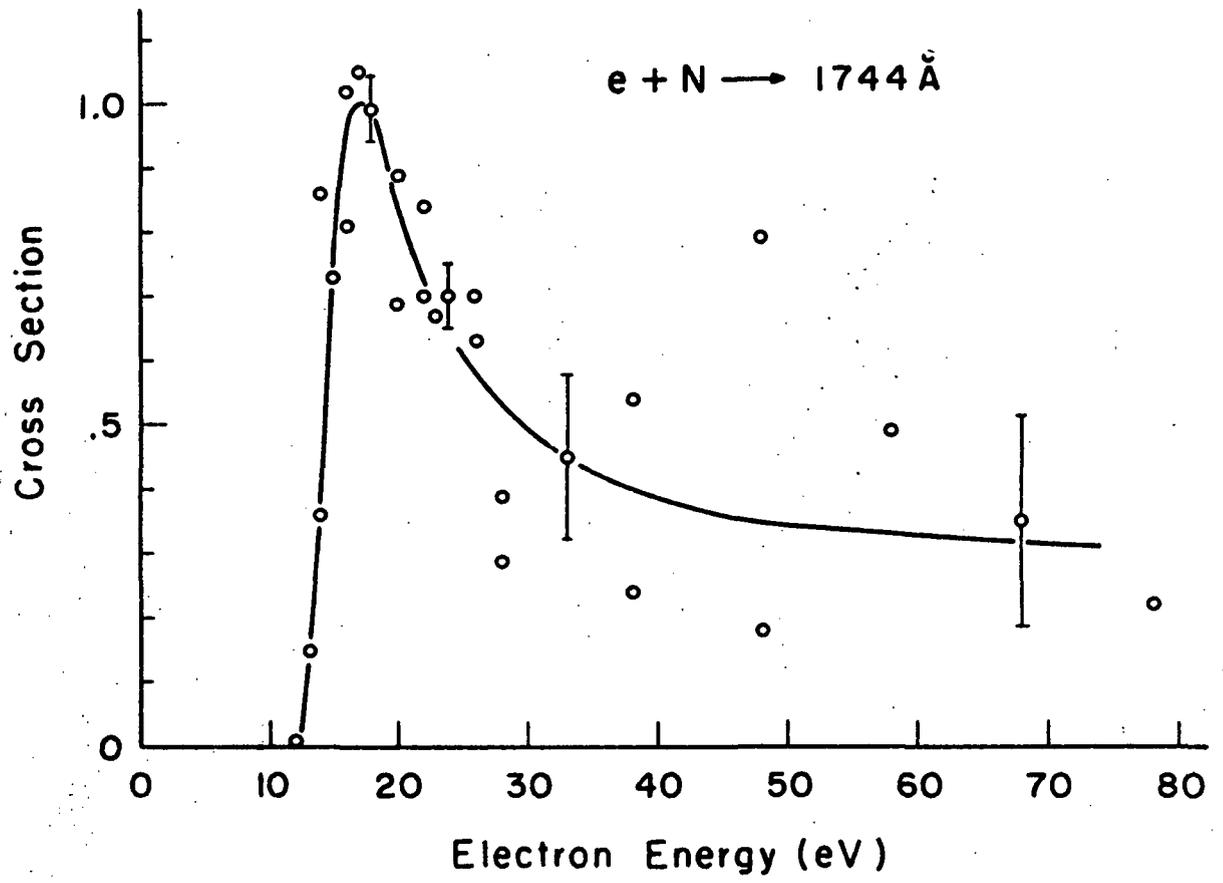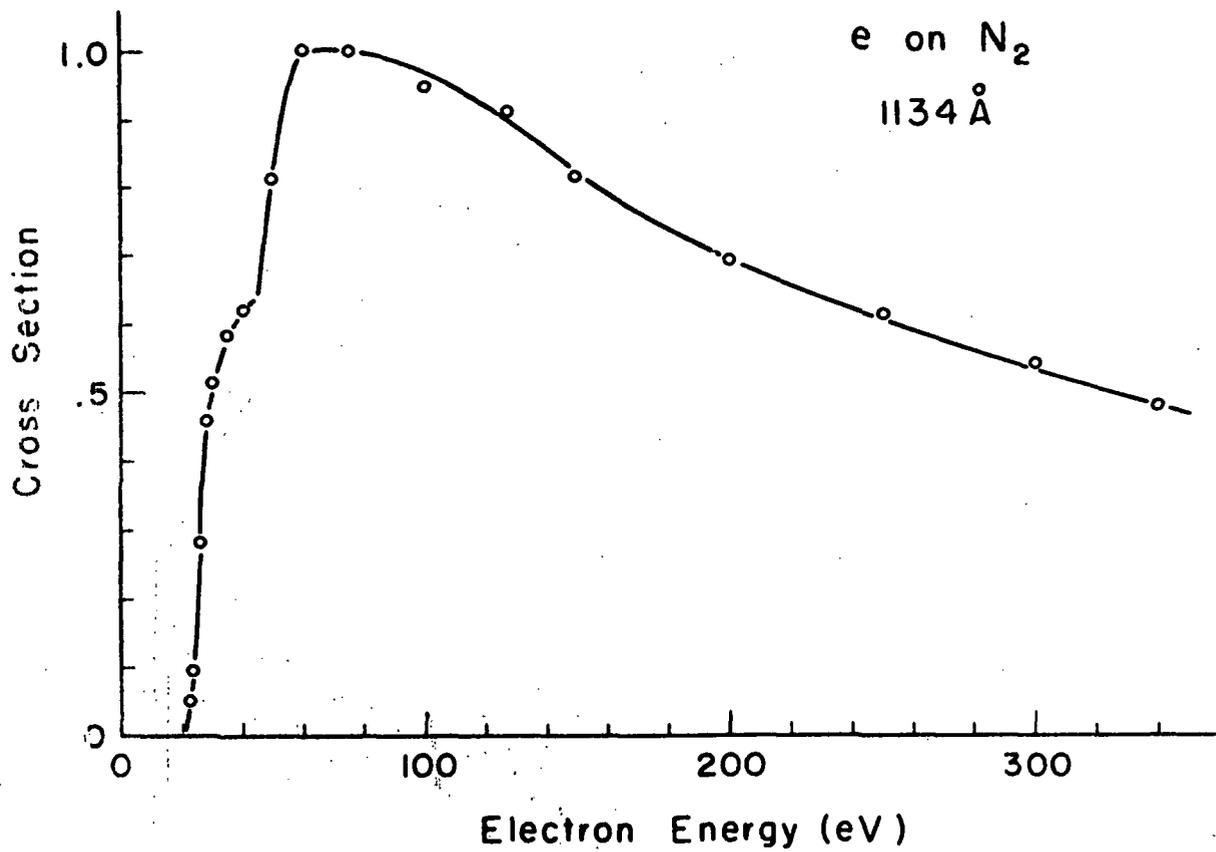
Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

e on N₂
1134 Å

Figure 7

Figure 8

e + N (16 eV)

1593 Å

1243 Å

1200 Å

1134 Å

1085 Å

e + N₂ (100 eV)

LBH Bonds

Intensity (Linear Scale)

Wavelength (Å)

Figure 9