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COLLISION QUENCHING EFFECTS IN NITROGEN AND HELIUM EXCITED BY A 30-keV ELECTRON BEAM

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16. Abstract

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SUMMARY

The quenching cross section for the 0-0 first negative band of nitrogen is determined for temperatures between 78 K and 300 K. As the temperature increases above 78 K, the quenching reaches a maximum at approximately 140 K and then decreases as 300 K is approached. At temperatures on the order of 5000 K, quenching is reported to increase with temperature and must therefore reach a minimum at some intermediate temperature between 300 K and 5000 K. By comparison, quenching of the 5016 Å helium line increases continuously over the temperature range 78 K to 300 K.

INTRODUCTION

Interest in hypersonic flight at high altitudes has fostered the development of measurement techniques to study high-speed, low-density flows. One of the more successful methods is the electron-beam fluorescence technique for measuring densities (see ref. 1), which has the important advantage of interference-free measurements. A collimated beam of moderate-energy electrons is directed through the flow field to excite a fraction of the molecules, on the order of 10^{-4} or less, along the path of the beam. The excited molecules decay spontaneously giving rise to the characteristic fluorescence spectrum of the gas. In nitrogen or air at low densities the most intense bands in the visible spectrum are the first negative bands of nitrogen. For densities less than 10^{22} molecules/meter^3 the intensities of the first negative bands are proportioned to the density. At higher densities an increasing number of excited molecules undergo nonradiative transitions upon collision with a ground-state molecule, resulting in a nonlinear relation between density and intensity. This effect is referred to as quenching. In the limited context of this paper, quenching refers only to collisions between excited molecules or atoms and ground-state molecules or atoms of the same species, although quenching collisions can occur between molecules of different species. To determine density when quenching is appreciable, a calibration

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curve of intensity plotted against density is obtained at some convenient temperature, usually room temperature. This curve is not a unique calibration unless the quenching rate is independent of temperature. Theoretical studies (see refs. 2 and 3) predict that for temperatures of a few hundred degrees or less, the quenching rate should be constant, but there is no experimental confirmation of this.

This paper is concerned primarily with the quenching of the 0-0 first negative band in pure nitrogen. Several investigators (refs. 4 to 7) have determined the quenching cross section at room temperature, and Camac (ref. 8) has made measurements at temperatures on the order of 5000 K. For aerodynamic studies these measurements must be extended down to the low free-stream temperatures achieved in high Mach number wind tunnels, and the present experiments cover the range from 78 K to 300 K.

Although air and nitrogen are the most common gases for aerodynamic tests, several high Mach number wind tunnels operate on helium. When low-density helium is excited by an electron beam, the 5016 Å line is the most intense part of the visible spectrum. Preliminary results of the quenching of this line are presented to illustrate the different trends for quenching in helium and nitrogen as a function of temperature.

SYMBOLS

A_{jk}	transition probability between states j and k
A_t	sum of the transition probabilities originating from state j
C_T	proportionality constant relating i_p and I_{00} at temperature T
C_{296}	C_T at reference temperature of 296 K
c	velocity of light
F_j	excitation function defined by equation (1)
h	Planck's constant
I_{jk}	band intensity corresponding to transitions from state j to state k
I_{00}	band intensity corresponding to transitions from the zero vibrational level of $N_2^+B^2\Sigma_u^+$ to the zero vibrational level of $N_2^+X^2\Sigma_g^+$

I_{5016} intensity of 5016 Å line of helium

i beam current

i_p photomultiplier output current

$K = hc\nu A_{jk}F_j / A_t$

N number density

n_j population of state j

$N_2^+B^2\Sigma_u^+$ upper electronic state of nitrogen ion N_2^+

$N_2^+X^2\Sigma_g^+$ ground state of nitrogen ion

$Q(T)$ quenching cross section

q quenching term, $\frac{2Q(T)}{A_t}\sqrt{\frac{4RT}{\pi}}$

R gas constant for nitrogen

R_L load resistance in figure 2

T temperature

$\overline{V}$ mean molecular velocity

ν frequency

THEORETICAL BAND INTENSITY

When a beam of moderate-energy electrons (10 to 100 keV) passes through a gas, some molecules are raised to excited states which decay spontaneously giving rise to the characteristic spectrum for the gas. For steady conditions the number of molecules in the excited state, and hence the intensity of the spectrum, is determined by a balance between the populating and depopulating processes for that state. Muntz (see ref. 1) has derived a general expression for a band intensity and the reader is referred to that paper for a more complete discussion of the possible populating and depopulating mechanisms. For the first negative bands of nitrogen Muntz (ref. 1) concluded that high-energy electrons excite ground-state molecules directly to the $N_2^+B^2\Sigma_u^+$ states. These states either decay spontaneously to the $N_2^+X^2\Sigma_g^+$ states or are involved in quenching collisions with ground-state molecules which remove part or all of the excitation energy in radiationless transitions. If no other processes are significant, equating the rate of excitation to the rate of depopulation for a particular state j yields the equation

$$F_jNi = A_tn_j + \sqrt{2}Nn_jQ(T)\overline{V} \tag{1}$$

On the left-hand side of the equation, i is the beam current and F_j is an excitation function defined so that F_jNi molecules are excited to the j state per unit volume per second. The total number density is N, and n_j is the population of the j state. The sum of the transition probabilities originating from state j is denoted by A_t, so that A_tn_j is the rate of depopulation of the j state through all the spontaneous transitions. In the third term Q(T) is the quenching cross section and $\overline{V}$ is the mean molecular velocity. Apart from the notation, this term is the same as that derived by Kennard (ref. 9, p. 112) for the collision rate between two species of molecules both assumed to have a Maxwellian distribution of velocities. In writing equation (1) the assumption is made that the sum of all the populations of the excited states is negligible compared with N. For typical electron-beam excitation conditions used in this study the population of the excited states is on the order of 10^{-4} N. If the band observed corresponds to a transition from state j to a lower state k, then from reference 10 and equation (1) the band intensity is given by

$$I_{jk} = hc\nu A_{jk}n_j = \frac{hc\nu A_{jk}F_jNi}{A_t + \sqrt{2}NQ(T)\overline{V}} \tag{2}$$

Equation (2) can be written

$$I_{jk} = \frac{KNi}{1 + \frac{\sqrt{2}N\overline{V}Q(T)}{A_t}} \tag{3}$$

where $K = \frac{hc\nu A_{jk}F_j}{A_t}$ and depends on the electron-beam energy.

The most intense band in the visible nitrogen spectrum at densities below 10^{23} molecules/meter^3 is the 0-0 first negative band corresponding to transitions between the zero vibrational levels of the $N_2^+B^2\Sigma_u^+$ state and the $N_2^+X^2\Sigma_g^+$ state. For density measurements this band is spectrally isolated from the rest of the spectrum and the intensity measured with a photomultiplier through a suitable optical system. The photomultiplier output current i_p is proportional to the band intensity and can be written as $i_p = C_T I_{00}$ where C_T depends on the photomultiplier sensitivity and the geometry of the apparatus and optical system. From equation (3), after substituting for $\overline{V}$ and normalizing the intensity by the beam current, a nondimensional equation is obtained:

$$\frac{i_p}{i} = \frac{C_T I_{00}}{i} = \frac{C_T K N}{1 + \frac{2NQ(T)}{A_t}\sqrt{\frac{4RT}{\pi}}} \tag{4}$$

The variation of the quenching cross section $Q(T)$ with the relative velocity of the colliding molecules is governed by the potential function for a neutral molecule colliding with a nitrogen molecular ion. For thermal collisions at temperatures of a few hundred degrees or less, the long range part of the interaction potential dominates the quenching rate. This interaction consists of a charge-quadrupole term and an induced-dipole term, but Wolf (see ref. 2) states that the charge-quadrupole interaction is only important at extremely low energies. The potential for the induced dipole varies as the inverse fourth power of the distance between the molecules. With this potential, semiclassical analyses (refs. 2 and 3) predict a cross section varying inversely as the relative velocity. For thermal collisions the relative velocity varies as $\sqrt{T}$, and it can be shown using equation (4) that the quenching rate should be independent of temperature. One result of the present experiments is that they permit an examination of validity of these simplified theoretical considerations of the quenching of the 0-0 band of nitrogen; also they improve the measurements of density over a range of temperatures.

EXPERIMENTAL DETAILS

The experimental measurements were made in a flowing stream of nitrogen or helium using a 30-keV electron beam with currents of 200 microamperes (μA) or less. Figure 1 is a schematic diagram of the apparatus showing the heat exchanger used to cool the gas and the position of the electron beam. The heat exchanger is enclosed in a bath of 2-methylbutane, which is thermally insulated by the surrounding vacuum. A controlled flow of liquid nitrogen through a coil immersed in the 2-methylbutane permits the cooling of the heat exchanger to any desired temperature between 300 K and 113 K. Alternatively, liquid nitrogen is used in place of 2-methylbutane to obtain temperatures close to 78 K. The heat exchanger consists of two chambers separated by a valve which controls the pressure in the first chamber. This chamber acts as a reservoir of cooled gas for the main chamber to ensure that the gas is cooled to the temperature of the heat exchanger even at the lowest pressures. The gas temperature is assumed to be the mean value recorded by thermocouples on the last baffle and along the exit pipe from the heat exchanger (located as indicated by x in fig. 1).

The coolant bath surrounding the exit pipe maintains the temperature uniform along the pipe to better than 1 K. Velocity profiles across the exit pipe are similar to those for developing pipe flow. For pressures greater than about 1 N/m^2 mechanical vacuum pumps maintain an axial velocity along the pipe of 50 m/sec or more. At lower pressures diffusion pumps are used to maintain this flow speed. With the drift tube and Faraday cup located as shown, this flow speed is sufficient to prevent convective heating of the gas in the exit pipe. For all conditions of these experiments the intensity measurements did not vary with time and increased linearly with beam current. This would not be the case if the hot drift tube or Faraday cup were heating the gas. In this respect the present apparatus is superior to a static test chamber, where convective heating of the test gas might be a serious problem.

A pressure transducer, calibrated against a McLeod gage, recorded the test-chamber pressure through an orifice in the chamber wall adjacent to the exit pipe. The wall temperature remained close to ambient, and thermal transpiration along the tubing was negligible. The density of the flowing gas was calculated from the measured pressure and temperature.

Details of the electron gun are given in reference 11. Vacuum pumps maintain the electron gun at a high vacuum and the electron beam enters the test region through a narrow orifice in the drift tube. For pressures below 130 N/m^2 the orifice diameter is 0.1 cm, but for high pressures the diameter is reduced to 0.05 cm to enable the required vacuum to be maintained in the electron gun.

The photomultiplier output must be normalized by the beam current, which is difficult to measure because of its scatter as it passes through the test chamber. The test chamber is grounded and used as the current collector, but to minimize the emission of secondary electrons and the reflection of primary electrons from the chamber surfaces, a carbon Faraday cup is placed at the bottom of the chamber, as shown in figure 1. Figure 2 is a schematic circuit diagram showing the electron gun, drift tube, and high-voltage supply connected to the test chamber only by the resistor R_L, which should therefore carry the beam current. With this arrangement the drift tube is at a potential $R_L i$ above the test chamber. As the electron beam produces a plasma in and around the end of the drift-tube orifice, the drift tube draws a net electron current and thus leaves positive ions in the plasma which diffuse to the test-chamber walls and, hence, reduce the indicated beam current. To minimize this effect, a grounded shield was placed around the drift tube with a small opening left around the drift-tube orifice. Provided the drift tube was not more than 0.02 volt above the test-chamber potential, the intensity varied linearly with beam currents. With $R_L = 100$ ohms the maximum allowable beam current was 200 μA. Within this current range measurements were performed to confirm that intensity varied linearly with beam current for the complete range of temperatures and densities used in the experiment.

A lens system focused the fluorescence from the beam into the entrance slit of a spectrometer. Initially a 2-mm-long slit was orientated parallel to the length of the beam image and the lens system adjusted to observe a point 2.8 cm from the end of the drift tube. The lens system was traversed to move the beam image across the entrance slit while recording the photomultiplier output. This showed that for a 30-keV electron beam traversing nitrogen at a density of 1.2×10^{23} molecules/meter^3, approximately 1 percent of the electrons were scattered beyond a diameter of 1.5 cm. A different lens system was then used to form a reduced image of the beam perpendicular to the slit, the length of which determined the width of the field of view. Varying the field-of-view width from 1.5 cm to 1.9 cm caused less than a 1-percent increase in photomultiplier output. Thus, the previous measurement of beam width was confirmed. The experiment was performed with a field-of-view width of 1.9 cm and entrance slit dimensions of 0.63 cm by 0.25 cm.

The 0-0 band of nitrogen was scanned while the beam current and intensity were recorded. The 1-1 first negative band at 3884 Å and the 3-6 second positive band at 3895 Å overlap the 0-0 band. To minimize the effect of these bands, the "tail" of the 0-0 band was extrapolated smoothly to zero, using theoretical rotational-line intensities as a guide. This leads to no more than a 2-percent error in the 0-0 band intensity since the 3-6 band is weak at the densities considered and the 0-0 band has a low intensity beyond 3884 Å even at room temperature. Integrating the area under the adjusted intensity curve and normalizing by the beam current gives the required band intensity.

Similar measurements were performed on the 5016 Å line of helium. The exit slit of the spectrometer was wider than the entrance slit so that symmetrical, flat-topped intensity traces were obtained. The line intensity was found to be proportional to the peak value of this trace.

QUENCHING CROSS SECTIONS

Equation (4) can be rearranged as

$$\frac{N}{i_p/i} = \frac{1}{C_T K} + \frac{1}{C_T K} \frac{2NQ(T)}{A_t} \sqrt{\frac{4RT}{\pi}} \tag{5}$$

Therefore, $N/(i_p/i)$ plotted against N should be a straight line and the quenching cross section can be determined from the ratio of the slope to the intercept on the $N = 0$ axis.

The function C_T depends on photomultiplier sensitivity, which varied less than 0.5 percent during the course of measurements at one temperature. However, over the several weeks required for completion of the experiment, C_T varied appreciably. For convenience in interpretation of the data, the values of C_T at a reference temperature of 296 K are used in the presentation of data and were determined in the following manner. Values of $C_T K$ were obtained from plots of $N/(i_p/i)$ against N since the intercept determines $1/C_T K$. These data were then normalized by multiplying i_p/i measured at a different temperature by C_{296}/C_T. Typical plots of $I_{00}C_{296}/i$ against N are shown in figures 3 and 4 for nitrogen and helium, respectively. Typical plots of $Ni/I_{00}C_{296}$ against N are shown in figure 5 for nitrogen. The data define straight lines except at low densities, as seen on the detailed plot for 78 K, which is typical of all the data.

To examine the different trend at low densities, values of $C_T K$ obtained from the the intercepts of $N/(i_p/i)$ plotted against N were compared with the values of the slope of the initial linear portion of the curve of i_p/i against N. Values of $C_T K$ obtained from these two methods differed by as much as 16 percent, which could not be accounted for by any combination of systematic errors. The different trend at low densities cannot be due to secondary electrons emitted from the test chamber walls or to reflected primary electrons. If secondary or reflected electrons entered the region in which measurements are made, the band intensity would be increased, whereas the observed results correspond to low intensities. One possibility is that collisional quenching is more complicated than assumed in deriving equation (5), and further studies at low densities are needed. In the absence of a more detailed knowledge of the quenching mechanism, the cross sections have been calculated using only data for densities between 1×10^{22} and 12×10^{22} molecules/meter^3, for which $N/(i_p/i)$ plotted against N defines a straight line. The values obtained are plotted in figure 6 and listed in table 1, but it should be emphasized that these values apply only to this limited density range. Figure 6 also shows values obtained at 300 K by several other investigators. (See refs. 4, 5, 6, and 8.)

Only one or two measurements of the cross sections were made at each temperature and reliable standard deviations cannot be calculated. The values in table 1 are mean values, and the error shown was estimated from the accuracy of the individual measurements. Neutral-density filters were used to confirm that the photomultiplier output was a linear function of intensity to better than 1 percent. For beam currents less than 200 μA, the intensity varied linearly with beam current to within 1 percent. Pressure measurements were accurate to ± 2 percent or better. The most significant source of error is in the determination of the low temperatures, and at 78 K the error could be 4 percent.

The intensity of the 5016 Å helium line is influenced by resonance absorption of the 537 Å line and excitation by secondary electrons. (See ref. 12.) These secondary processes were not investigated for the present apparatus and reliable quenching cross sections cannot be deduced from the data of figure 4 since plots of $Ni/I_{5016}C_{296}$ against N were nonlinear. However, resonance absorption of the 537 Å line and secondary electron excitation should not be a function of temperature, and the present data show that quenching increases with temperature over the range 78 K to 300 K.

DISCUSSION

The cross sections plotted in figure 6 decrease smoothly as the temperature increases toward 300 K, while Camac's measurements (ref. 8) at temperatures on the order of 5000 K vary as $T^{-1/6}$. For the aerodynamicist using equation (4) to relate band intensity to local number density, the variation of the quenching term $q = \frac{2Q(T)}{A_t}\sqrt{\frac{4RT}{\pi}}$ is of more direct importance. This term varies as $\sqrt{T}Q(T)$ and with increasing temperature reaches a maximum at about 140 K and then decreases as plotted by the dashed curve in figure 6. Camac's results (ref. 8) show that for temperatures on the order of 5000 K the quenching term increases at $T^{1/3}$ and, therefore, must be a minimum at some temperature between 300 K and 5000 K. Since the quenching rate is simply qA_t, the theoretical predictions of Wolf (ref. 2) and Gioumousis and Stevenson (ref. 3) are not in agreement with the present results. This indicates that the quenching mechanism observed in this work is more complicated than that described by a collision under the influence of an interaction potential varying as the inverse fourth power of the distance between the molecules. Brocklehurst and Downing (ref. 7) discuss some possible quenching mechanisms for nitrogen and point out that there is a strong excitation exchange interaction between like molecules. The products of such quenching are uncertain; the molecule may dissociate or be converted into another bound state. Any attempt to include these interaction forces in the theoretical calculations is beyond the scope of this work, and the aerodynamicist must rely on measured quenching cross sections to correct wind-tunnel data.

Before the data plotted in figure 6 can be used to determine the local density in a flow field, the local temperature must also be known. Although there are several methods of measuring temperature, the electron-beam probe (see ref. 1) can be used and offers several advantages. The probe does not interfere with the flow (see refs. 1 and 13) and the density and temperature can be measured simultaneously at the same point in the flow. Translational temperature can be determined with the electron-beam probe, but rotational temperature can be more easily measured and, in most flow fields of interest, is equal to the translational temperature. The measured rotational temperatures must be corrected for the effect of secondary processes and since these corrections vary with density, an iterative procedure is required to obtain corrected densities and temperatures, as discussed in detail in reference 13.

CONCLUDING REMARKS

The quenching cross section and quenching term for the 0-0 first negative band of nitrogen were determined for densities between 10^{22} and 10^{23} molecules/meter^3. These two parameters allow a room-temperature calibration curve to be used for the determination of local densities in the temperature range 78 K to 300 K. The quenching term reaches a maximum at approximately 140 K and decreases to a minimum at a temperature between 300 K and 5000 K. The low-temperature results and qualitative discussion of the high-temperature data provide the aerodynamicist with a means of interpreting electron-beam density measurements in nitrogen at densities on the order of 10^{23} molecules/meter^3, where quenching is appreciable.

In contrast to nitrogen, the quenching of the 5016 Å line of helium increases continuously over the temperature range 78 K to 300 K, but further studies are required to determine the quenching cross sections.

Langley Research Center,
National Aeronautics and Space Administration,
Hampton, Va., September 11, 1973.

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TABLE 1.- CROSS SECTIONS FOR QUENCHING OF THE $v' = 0$ LEVEL

OF THE $N_2^+ B^2\Sigma_u^+$ STATE OF N_2

T, K	Cross section, m^2	T, K	Cross section, m^2
296	4.8 (±0.2) × 10^{-19}	162	8.7 (±0.6) × 10^{-19}
247	5.6 (±0.3)	123	9.8 (±0.7)
208	6.7 (±0.3)	118	10.0 (±0.8)
168	7.7 (±0.5)	78	11.0 (±0.9)

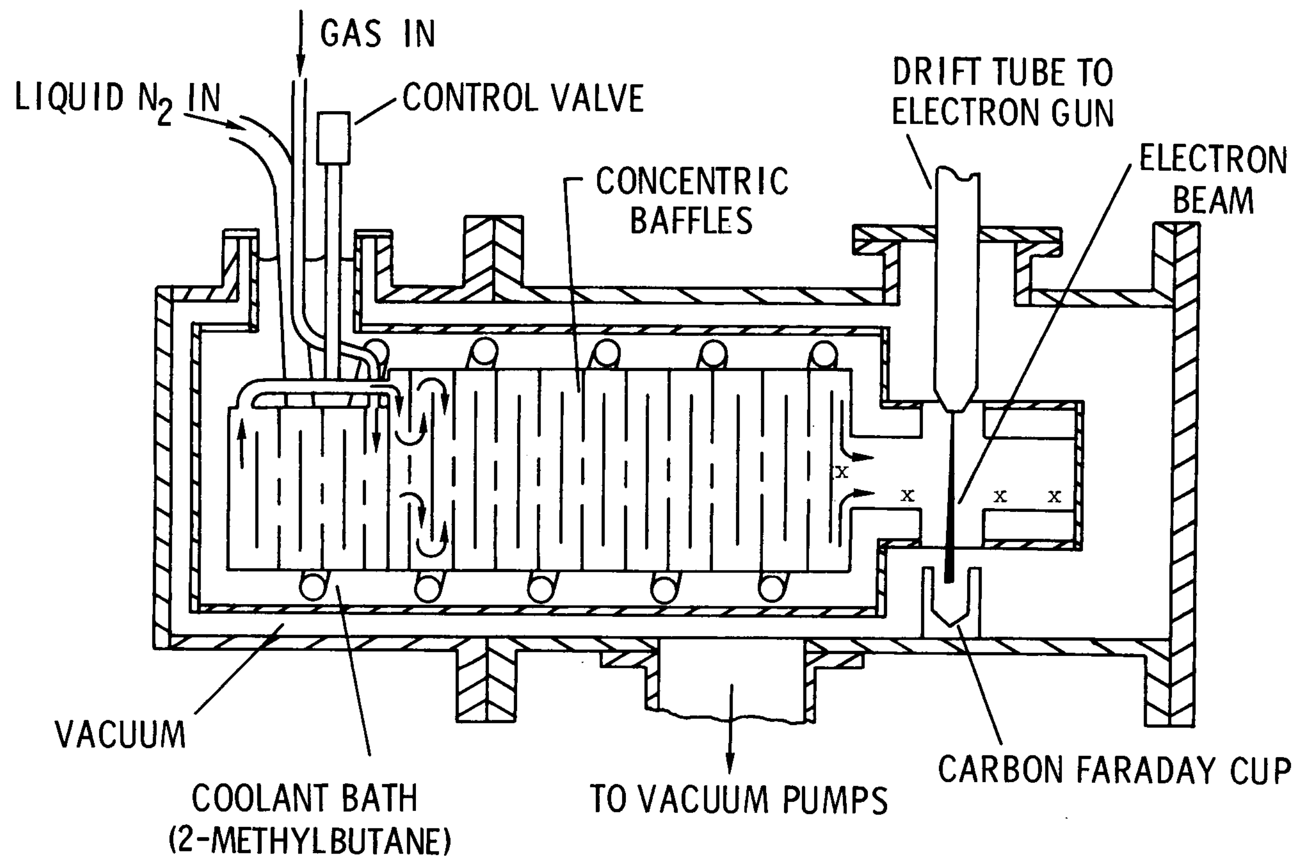

Figure 1.- Schematic diagram of apparatus. (Thermocouple locations denoted by x.)

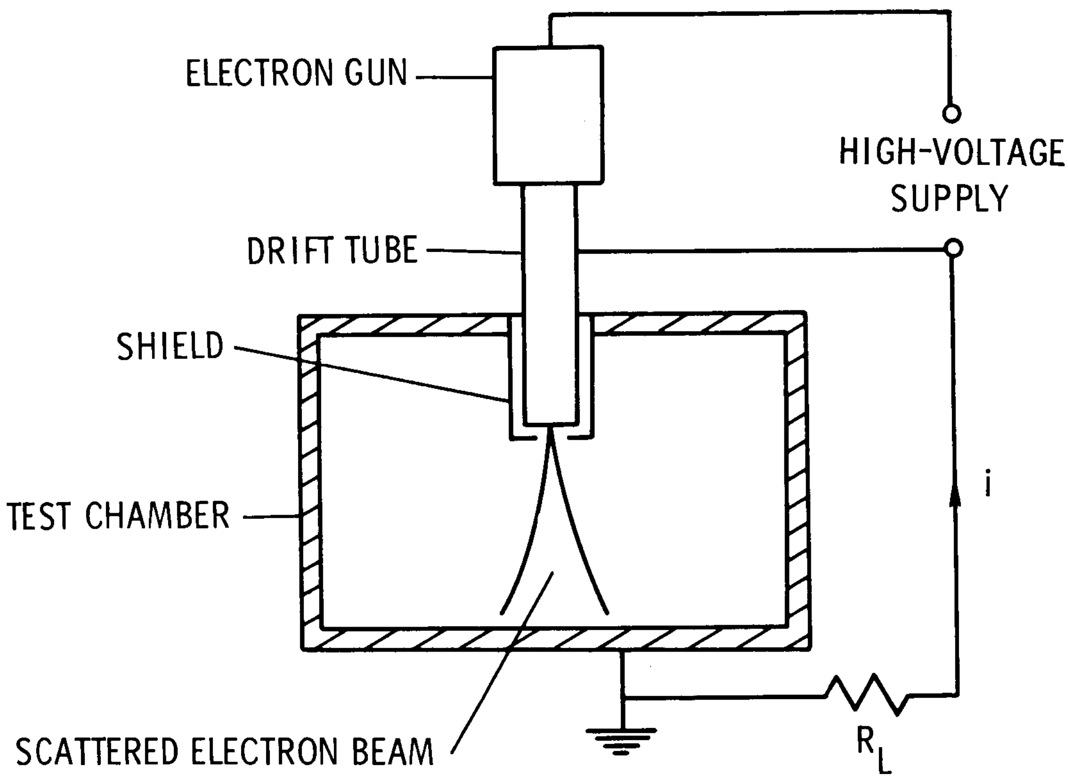

Figure 2. - Schematic circuit diagram.

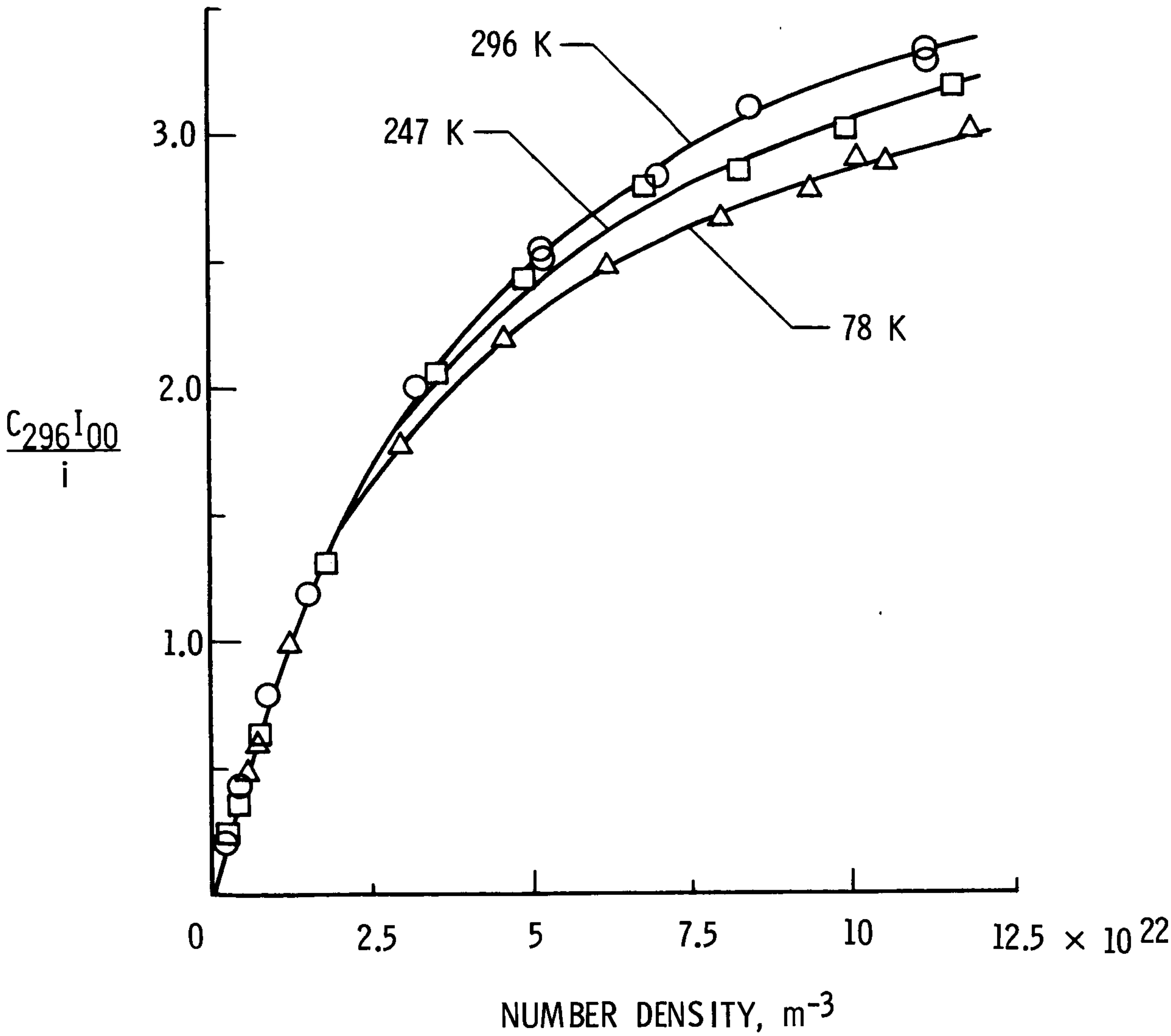

Figure 3.- Quenching of the 0-0 first negative band of N_2^+.

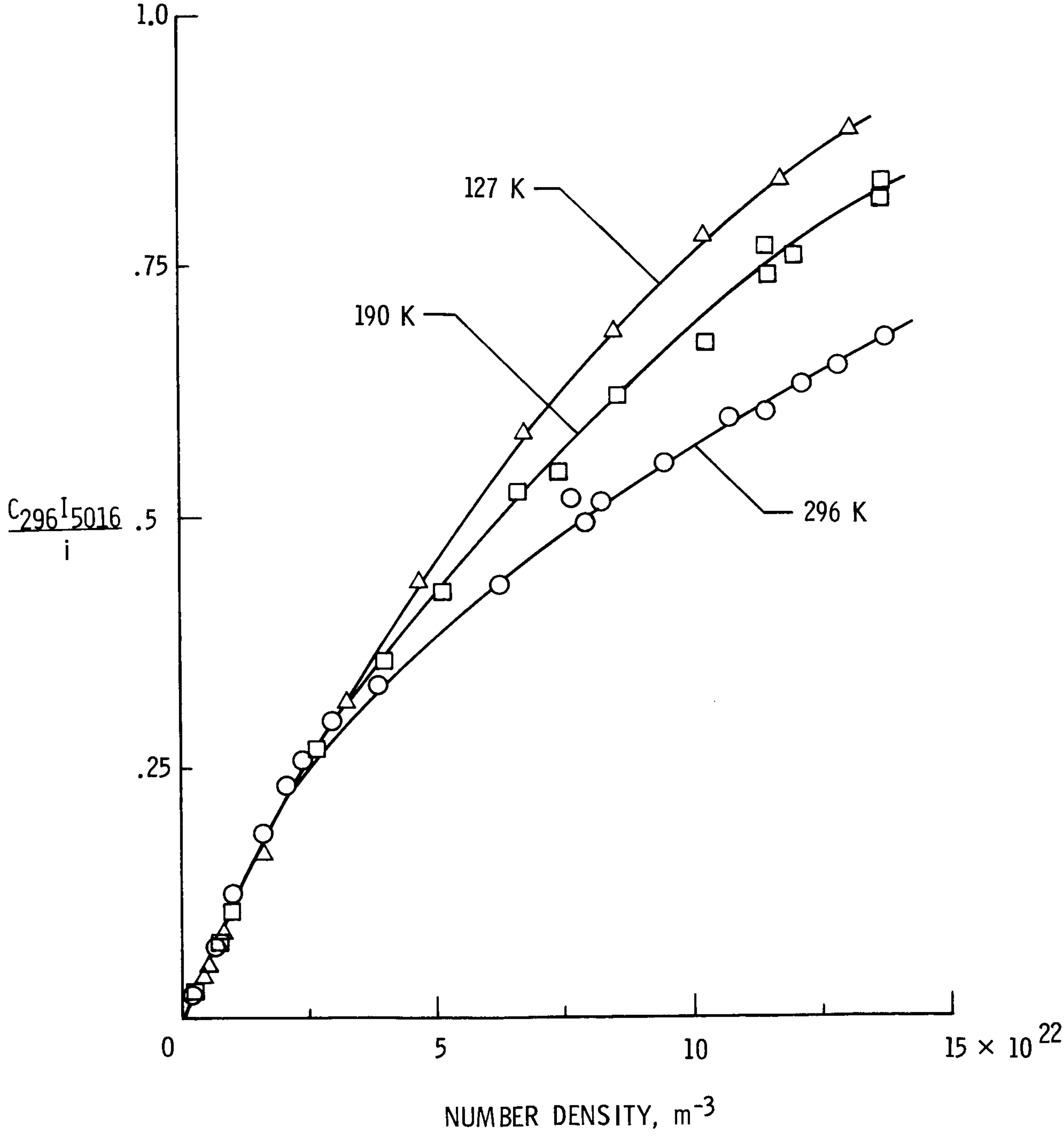

Figure 4.- Quenching of the 5016 Å line for helium.

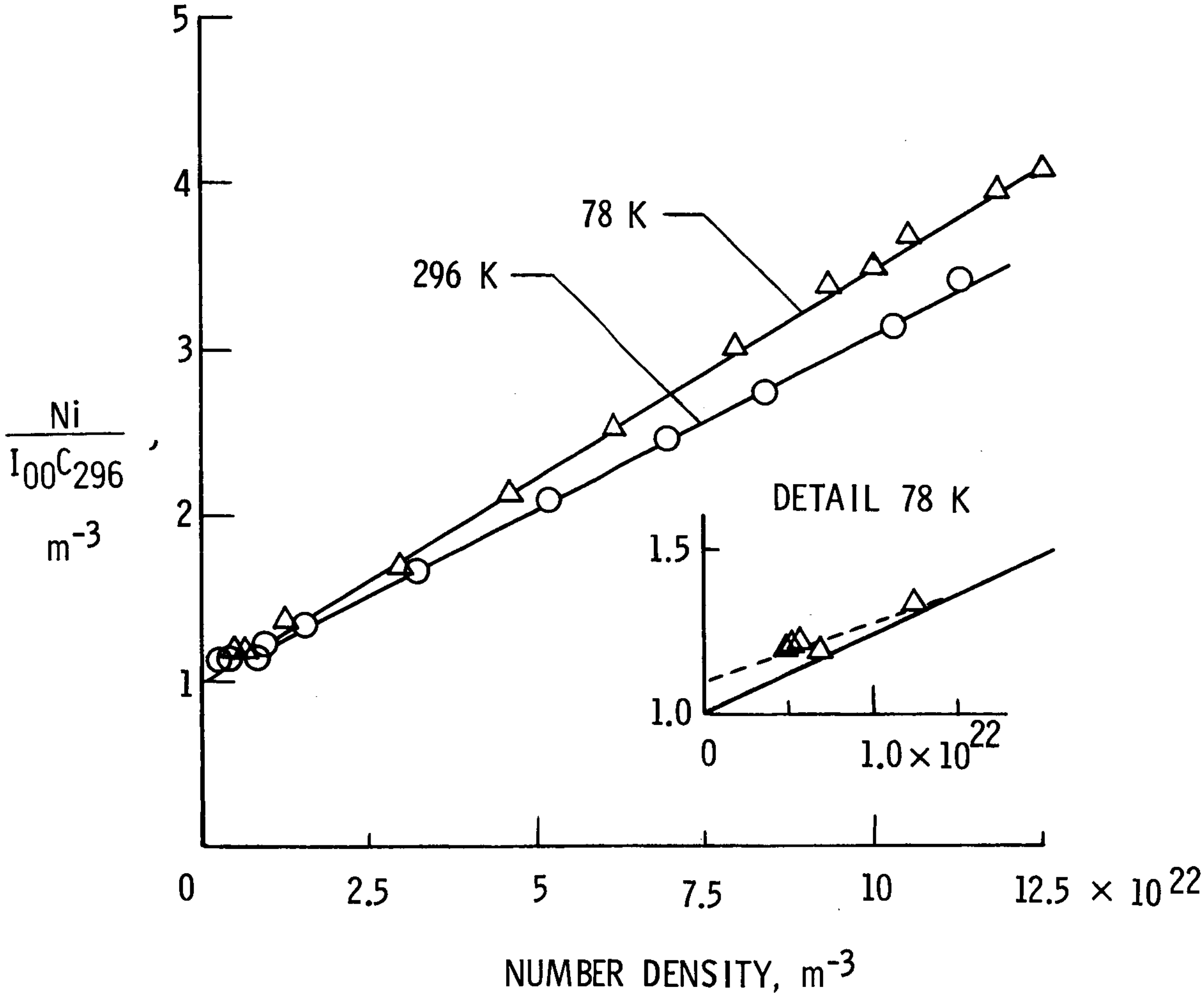

Figure 5.- Variation of $Ni/I_{00}C_{296}$ with number density for nitrogen.

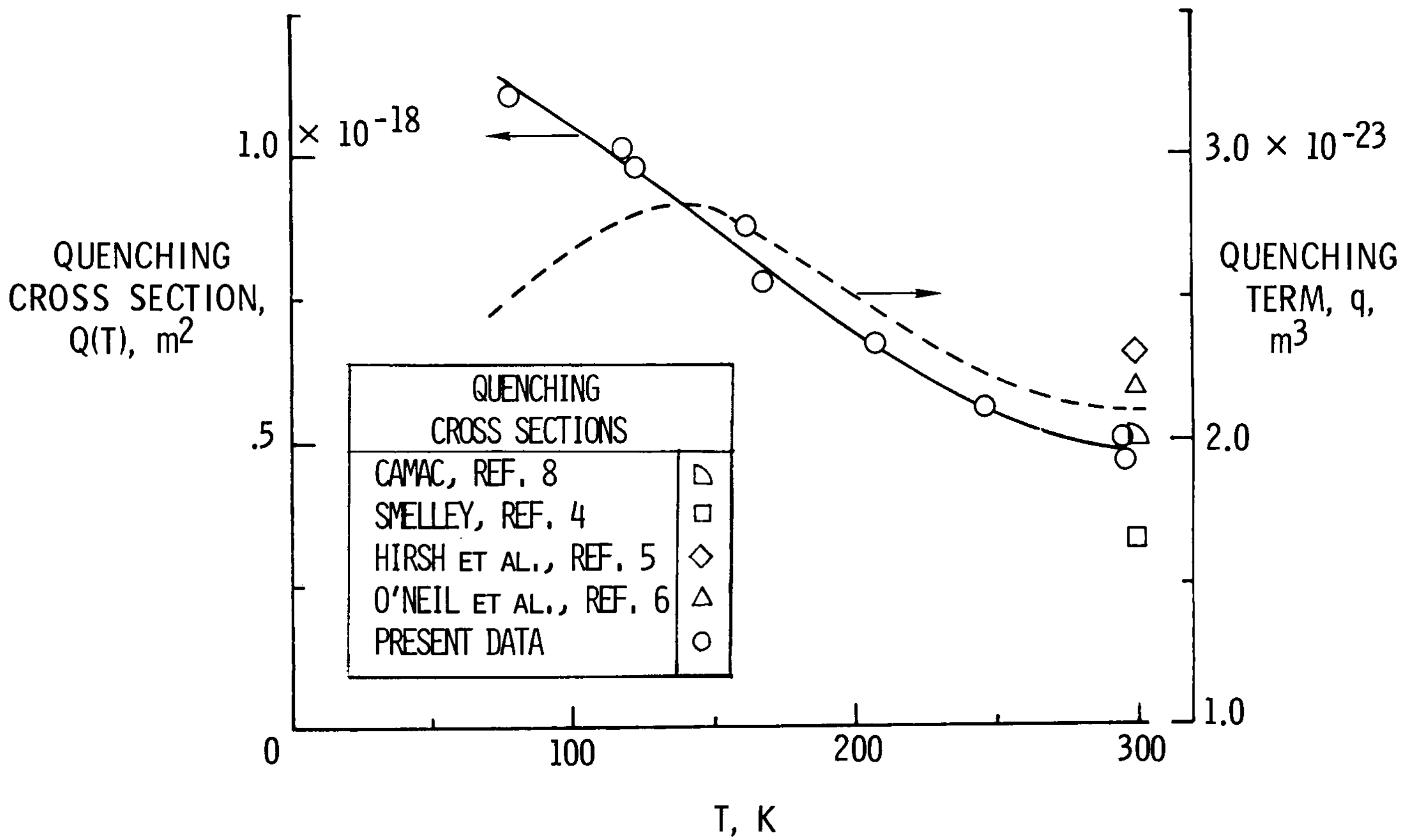

Figure 6.- Quenching cross sections for the 0-0 band of nitrogen.