Composite Model Describing the Excitation and De-excitation of Nitrogen by an Electron Beam

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Based on recent studies, the effect of re-excited ions in the emission of electron beam induced fluorescence in nitrogen has been estimated. These effects are included in the formulation of a composite model describing the excitation and de-excitation of nitrogen by an electron beam. The shortcomings of previous models, namely the dependence of the measured temperature on the true gas temperature as well as the gas density, are almost completely eliminated in the range of temperatures and densities covered by the available data.

Nomenclature

A = collision cross-section between a ground state ion and an exciting electron
B = density coefficient
C = mean thermal speed of the molecular ions
D = debye length in plasma or dipole transitions
F = excitation function of the neutral molecules or ions by a primary beam electron
G = transition factor of ground state molecules or ions
I = intensity of the rotational line in the rotational spectrum of nitrogen
K = quantum number of the rotational line of the excited ion before emission
L = distance required for an ionizing collision between a primary beam electron and the neutral gas molecules
P = overall transition probability of the ground state molecules or ions
Q = quadrupole transition
T = temperature of the gas
b = intercept of the least squares line fit
d = diameter of the electron beam
n = number density of the gas molecules, ions, or plasma electrons
p = gas pressure
$\propto$ = a proportionality constant
β = exponent relating the plasma electron number density to the gas molecule number density
γ = a proportionality constant
θ = characteristic rotational temperature of the gas
ν = frequency of the rotational line
ρ = gas density in grams per cubic centimeter
σ = standard error in the measurement of the average temperature

Subscripts
D = dipole transition
I = ions
K = quantum number
Q = quadrupole transition
e = plasma electron
r = rotational mode of the molecules

I. Introduction

BECAUSE electron beam fluorescence can be used to obtain information about a flowing rarefied gas with a minimum of disturbance of gas flow, it has been used extensively in recent years.[1-4] A good bibliography of the work done up to 1968 may be found in Ref. 5. In principle, information can be obtained about a localized region in the flow. This region is known as the observation volume, and it lies on the intersection of the electron beam and the detection system optical axis.

The intensity of the rotational lines in the rotational structure of the 0-0 band of the first negative system of the nitrogen molecular ion carries information about the rotational equilibrium distribution which is related to the gas temperature. Muntz[6] was the first to propose an analytical model to describe the excitation and de-excitation of nitrogen by an electron beam, in order to predict the intensity of the rotational lines. His model used two successive dipole transitions and is hereafter called the D-D model. Hickman[7] proposed a different model to interpret data obtained at low temperatures. This model described the excitation de-excitation of nitrogen by a combination of quadrupole-dipole transitions and is hereafter called the Q-D model. Ashkenas[8] attempted to evaluate the ability of the two models to predict the true gas temperature. He found that the calculated temperature obtained from either model depends upon the gas density, the true gas temperature, and the number of rotational lines used in the calculations. These conclusions were later confirmed by Williams.[9] Attempts have been made to correct the measured temperature employing either a correction factor or new methods of data analysis without qualitative explanation.[10-12] Previous electron beam temperature measurements reveal, upon examination, both experimental and data analysis errors. When these errors are accounted for,[13] the apparent dependence of the calculated temperature on gas density and gas temperature is partially reduced, while the dependence upon the number of rotational lines used in the calculations is eliminated.

This paper proposes a new model based upon recent electron beam rotational temperature measurements and upon the data of Ashkenas.[8] The new model is a composite of the Q-D and D-D models. In the past some authors have assumed that there are quenching collisions which somehow depopulate some rotational levels faster than others. If this is true, then one must be able to determine, in some detail, the processes of differential depopulation of rotational states. In the composite model presented here, all rotational levels are presumed to be quenched, without preference, to an equilibrium distribution. It is further presumed that the discrepancy in observed spectra is due to re-excitation of the ground state ions by either secondary or primary electrons. For gas temperatures between 70 K and 400 K and gas densities between 10^{+17} and 10^{+13} molecules/cm^3, this model is shown to eliminate some of the shortcomings of the previous models, namely their strong dependence upon the true gas temperature and the gas density. Additional data is necessary before the validity of the model can be extended to temperatures below 70 K.

Received March 7, 1974; revision received October 25, 1974. This work has been partially supported by the National Science Foundation Grant GK-15200 and the UCSB Research Fund. Also, we wish to thank Dr. Ashkenas for the generous contribution of his data bank.

Index categories: Atomic, Molecular, and Plasma Properties; Research Facilities and Instrumentation.

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II. Formulation

The D-D model overpredicts the temperature of the gas when the true gas temperature is about 300 K. The difference between the spectrally measured temperature and the true gas temperature increases as the true gas temperature is reduced to 78 K.[13] The Q-D model predicts approximately the true gas temperature at 300 K but predicts a lower gas temperature at 78 K. The Q-D model takes into account the fact that the homonuclear molecule of nitrogen in the ground state $N_2X^1\Sigma_g{}^+$ does not possess a dipole field. This model postulates a combination of quadrupole-dipole transitions of the neutral molecule of nitrogen from the ground state $X^1\Sigma_g{}^+$ to the excited state of the ion $B^2\Sigma_n{}^+$. Based upon the ability of the Q-D model to predict the approximate correct temperature of the gas at 300 K, it is reasonable to use it as a basis for a composite model. The remaining shortcomings, namely the density dependence and the prediction of a lower value for the gas temperature at 78 K, may be due to contributions from molecules or ions which arrive at the emission state by paths not postulated by the Q-D model. This can be represented by the equation

$$I_K \propto (nFP_{\text{Q-D}} + \text{other terms}) \tag{1}$$

where I_K is the intensity of the rotational line with quantum number K, n is the number density of the gas molecules, F is the function for ionization of the gas by a fast moving beam electron, and $P_{\text{Q-D}}$ is the transition probability of the Q-D model. This can be expressed by

$$P_{\text{Q-D}} \propto G(K, T_r) \exp(-K[K+1]\theta/T_r) \tag{2}$$

where

$$\begin{aligned} G(K,T_r) = \left(\frac{v}{v_o}\right)^4 & K\{K(K-1)(K-2)/(2K-1)(2K-3)(2K+1) \times \\ & \exp[6\theta(K-1)/T_r] + \\ & [(K+1)(K+2)(K+3)/(2K+1)(2K+3)(2K+5)] \times \\ & \exp[-6\theta(K+2)/T_r] + \\ & [2K^2(K-1)/3(2K+1)^2(2K-3) + K(K+1)^2/ \\ & (2K+1)^2(2K+3)] \exp(2\theta K/T_r) + \\ & [2(K+1)^2(K+2)/3(2K+1)^2(2K+5) + \\ & K^2(K+1)/(2K+1)^2(2K-1)] \times \\ & \qquad \exp[-2\theta(K+1)T_r] \end{aligned} \tag{3}$$

v is the frequency of the rotational line under consideration; v_o is a reference frequency; θ is the characteristic rotational temperature of nitrogen.

A review of Muntz's[6] work indicates that a potential cause of the experimental discrepancies may be due to the re-excitation of the ground state ions from the $X^2\Sigma_g{}^+$ state to the $B^2\Sigma_u{}^+$ state followed by an emission to the $N_2{}^+X^2\Sigma_g{}^+$ ground state of the ion. Muntz's analysis assumed a dynamic balance between the production of ions by an inelastic collision between the primary electron and the neutral molecule in the ground state, and loss of ions by diffusion to the walls of the chamber from a cylinder of a given height and a diameter equal to the beam diameter. From an estimate of the ratio of the collision cross-section of the fast beam electron with the ion and of the fast beam electron with the neutral molecules it is possible to calculate the ratio of the rate of re-excitation of ions to the rate of direct excitation of the neutral molecules. For steady-state conditions this represents the ratio of the number of re-excited ions to the number of directly excited ions. Muntz estimated this ratio to be between 4% and 0.004%. Thus it seemed that the re-excited ions had a small effect and therefore could be neglected. However, if the beam region is considered as a plasma, then from the theoretical work of Dunn and Self[14] it is possible to find an ion number density that is a thousand times larger than the number density assumed by Muntz, for pressures as low as 10^{-3} Torr. This result is confirmed by Hedvall's[15] measurements. On the other hand, it appears that Muntz's estimate of the collision cross-section between fast beam electrons with a 17.5 KeV energy and the neutral molecules is larger than the data obtained by Barnett et al.[16]

From this information, a reasonable starting hypothesis is that the dynamic balance used by Muntz underestimates the proportion of ions re-excited by either the primary or secondary electrons which contribute to the spectrum. Therefore, the effect of the re-excited ions may cause the density and temperature dependent errors present in either excitation model.

Before an expression describing the re-excitation of the ion can be obtained it is necessary to determine the rotational distribution of the unexcited ions in the $N_2{}^+X^2\Sigma_g{}^+$ state. In considering the thermal diffusion of the unexcited ion, it is known that the time required for it to move from the center of the beam to the outer rim is of the order of 10^{-6} sec. for a beam of a diameter of a few mm. It is also known that the lifetime of the excited state of the ion is of the order of 10^{-8} sec. From the work of Gadamer,[17] Camac,[18] and Maguire,[10] evidence is abundant that radiationless quenching collisions significantly affect the intensity of the rotational lines at pressures as low as 300 mTorr. That is to say, collisions between some of the just-excited ions and the neutral molecules of the gas take place before these ions emit, i.e., during the lifetime of the excited ions, which is 10^{-8} sec. In the time required for diffusion at least ten collisions take place between an unexcited ion and the neutral gas molecules before the ion escapes the beam perimeter. From the theoretical studies of Lordi, et al[19] and Parker,[20] which have some experimental confirmation,[21-23] the number of collisions required for complete rotational equilibration of nitrogen varies from about two collisions at 100 K to about 4 collisions at room temperature. Based on the preceding, it is reasonable to assume that the ions in the ground state $N_2{}^+X^2\Sigma_g{}^+$ are in rotational equilibrium with the neutral molecules. With this assumption, the rotational equilibrium distribution of the unexcited ions can be represented by a Boltzman rotational distribution at the true gas temperature.

Knowing the rotational equilibrium distribution of the ground state ions, and assuming that the primary electron interacts with the dipole field of the ions, the excitation and de-excitation path of the ions becomes identical to the path described by the D-D model. The transition probability for the ground state ions is therefore given by

$$\begin{aligned} P_{\text{D-D}} \propto \left(\frac{v}{v_o}\right)^4 & \{(K+1)\exp[-2(K+1)\theta/T_r] + \\ & K\exp(2K\theta/T_r)\} \exp[-K(K+1)\theta/T_r]K/(2K+1) \end{aligned} \tag{4}$$

The rate of excitation of ions from the ground state to the excited state can be expressed, in terms of n_I, the number density of ions, and F_I, the excitation function of the ground state ion by a primary electron. Equation (1) is therefore rewritten as

$$I \infty (nFP_{\text{Q-D}} + n_I F_I P_{\text{D-D}}) \tag{5}$$

In describing Eq. (5), we have neglected the effect of quenching collisions upon the total intensity.

To estimate the number density of ions, let us consider the dynamic balance between the rate of creation of ions by ionizing collisions between a neutral molecule and a fast moving beam electron, and the rate of depletion of ions by thermal diffusion to the walls of the chamber. The balance can be described by the equation

$$nF = \langle C \rangle dL\pi n_I/4 \tag{6}$$

where n is the gas number density; n_I is the ion number density; d is the beam diameter; L is the mean distance required for an ionizing collision; and $\langle C \rangle$ is the mean thermal speed. The ratio n_I/nF is expressed as

$$n_I/nF = \gamma/\langle C \rangle L \tag{7}$$

where γ is a constant depending on the electron beam configuration. $\langle C \rangle$ is proportional to the square root of the ion's temperature, which is assumed to be equal to the gas temperature. From Grün's experiment,[24] L is found to be inversely proportional to the gas number density.

The excitation function of the ions F_I is proportional to the

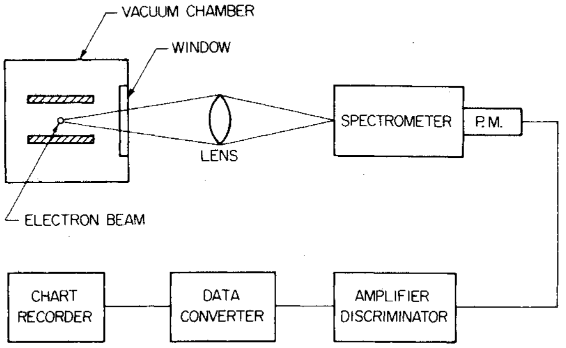

Fig. 1 A schematic of the experimental apparatus used to obtain the rotational spectra of nitrogen.

collision cross-section between the exciting electron and the ground state ion. The collision cross-section A_I for a given energy of the exciting primary electron is assumed to be proportional to the square of the Debye length D. The ion excitation function is then expressed by[25]

$$F_I \propto A_I \propto D^2 \propto (1/n_e) \tag{8}$$

where n_e is the number density of the plasma electrons. Physically this means that the field of the ground state ion is shielded from the exciting electron by the plasma electrons. The effectiveness of the shielding increases proportionally to the increase in the number density of the plasma electrons. This means that the D-D model provides a significant proportion of the emission at low density even though quenching collisions are not significant in the total emission. From Harbour's[26] analysis the number density of the plasma electrons can be empirically related to the number density of the gas molecules by the equation

$$n_e \propto (n)^\beta \tag{9}$$

where the value of the exponent β is approximately 1.5. Using Eqs. (7–9), the ratio $n_I F_I/nF$ can be expressed as

$$n_I F_I/nF \propto 1/\langle C\rangle L n^\beta \propto T^{-1/2}/n^{\beta-1} \tag{10}$$

Equation (5) can then be rewritten in terms of the gas pressure in millitorr and the gas temperature in Kelvin by

$$I = nF\left[P_{\text{Q-D}} + \alpha\frac{T^{(\beta-1.5)}}{p^{(\beta-1)}}P_{\text{D-D}}\right] \tag{11}$$

where α and β are constants to be found from the analysis of the available data. At this point it must be admitted that an excitation model with two adjustable empirical constants may be able to describe the data without describing the real physical process. The postulated model is at least plausible and does not rely upon a completely empirical treatment.

III. Experimental Apparatus and Procedure

Figure 1 shows a schematic diagram of the experimental arrangement used for the current data. A test chamber contains gaseous nitrogen at a low pressure. An electron beam generated by a Brad Thomson Industries welding gun is directed through a high vacuum system and through a 2 mm differentially pumped orifice into the test chamber. The electron beam is operated at about 15 kv and with a nominal current of 0.5 mA. The beam is collected by a Faraday cup, and the beam current is obtained by measuring the voltage drop across a 10 ohm resistance. The

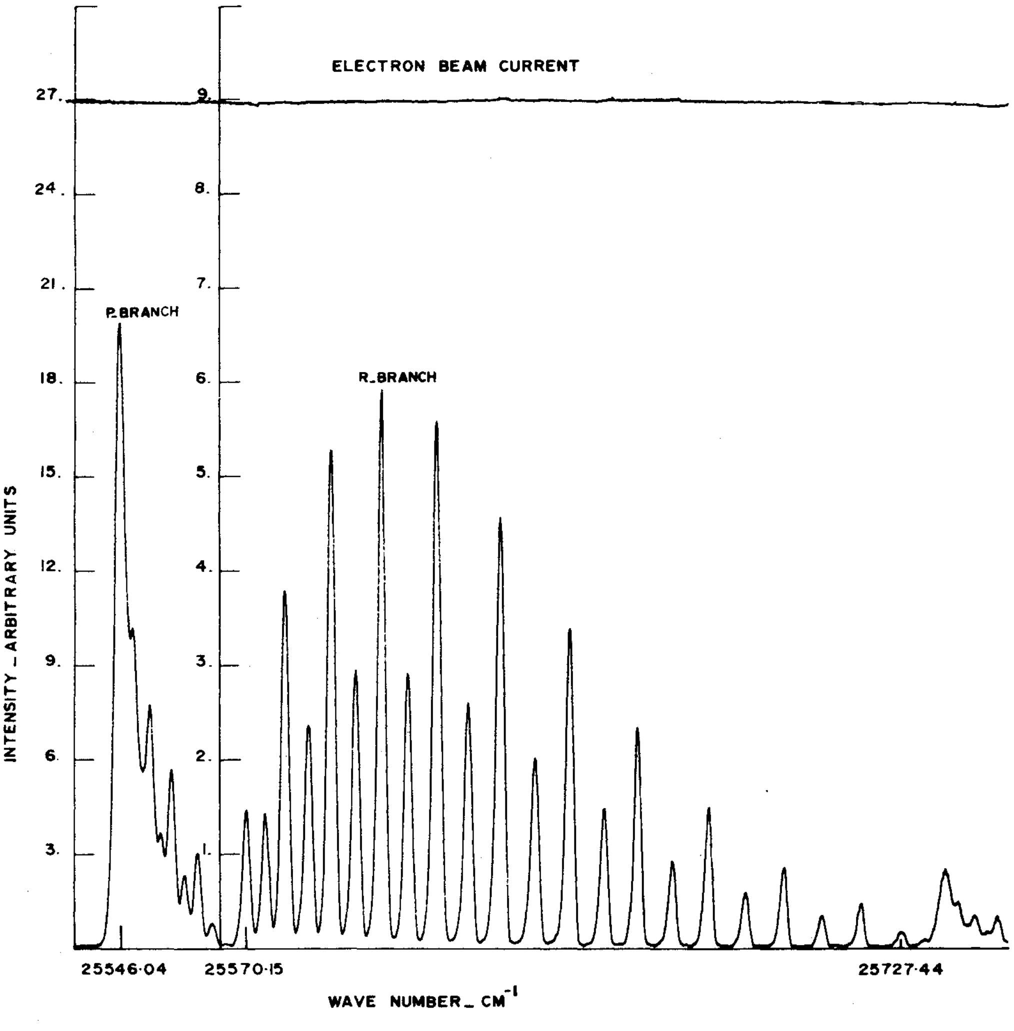

Fig. 2 A spectrum of the 0–0 band, first negative system of nitrogen, at 30 mtorr.

Table 1 Variations of predicted temperature for selected values of β and α (present data)

Exponent β	Value of α	Calculated value of coefficient B	$T_{calc.}/T_{true}$ average	σ Standard error in the average $T_{calc.}/T_{true}$
1.1	5.0	0.0263	1.0114	±0.0274
1.2	4.5	0.0231	1.0137	±0.0253
1.3	4.0	0.0199	1.0159	±0.0233
1.4	3.5	0.0171	1.0177	±0.0216
[a]1.45	3.0	0.0157	1.0174	±0.0208[a]
1.5	2.875	0.01448	1.0198	±0.0210
1.6	2.00	0.0124	1.0173	±0.0189

[a] These values represent the best fit for both room temperature and 78 K data.

current was held within 1% of the nominal value throughout the data collection periods.

Two parallel brass plates inside the vacuum chamber, separated by 2.54 cm, are centered about the beam. The plates are fitted with two copper-constantan thermocouples each to monitor the ambient temperature. The entire vacuum chamber and the brass plates are held at room temperature. A maximum of a 1% spread between all the thermocouples existed through all experimental runs.

The fluorescence produced by the beam is collected by a lens, which focuses the image of the beam on the entrance slit of a monochromator. The slit is placed parallel to the beam. A Jarrell-Ash Model 82-000 half-meter scanning monochromator is used, with a 1200 G/mm grating blazed at 5000 Angstroms, and a dispersion of 0.3 Å. A Centronic Q 4249 BA selected photomultiplier tube is attached to the exit slit of the monochromator, and the output of the photomultiplier is measured by an SSR photon counting system. This system consists of a model 1120 amplifier discriminator and an 1105 data converter. The spectral data is plotted on one channel of a Soltec two-pen recorder, and the beam current is recorded on the other channel.

To keep a constant instrument function, both the entrance and the exit slit were kept at a constant width of 30 μ. Neutral density filters are placed in front of the entrance slit of the monochromator to maintain a constant signal strength when the gas density was changed.

The pressure of the chamber was changed from 1–320 mtorr. During data collection the variation in the pressure is kept within 2% of the nominal value. Seventy-four rotational spectra were recorded. Figure 2 represents a typical spectrum of the 0–0 band of the first negative system of nitrogen obtained at a pressure of 30 mtorr.

IV. Results and Discussion

Using the spectra obtained in this study, and Eqs. (2–4) and (10), a least square analysis is employed to calculate the rotational temperature of the gas.[13] The calculated temperature is then normalized by the true gas temperature. A least square data fit is then calculated for the temperature ratio $T_{calc.}/T_{true}$ vs the logarithm of the gas density ρ, where the density ρ is expressed in g/cm^3. This is done to test the sensitivity of the model to density. When the predicted temperature loses its density dependence, one of the major goals has been met. The equation representing the relation between temperature and density is

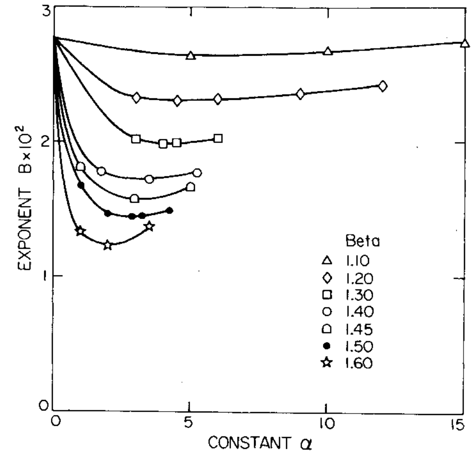

Fig. 3 The change in the value of B vs the change in the value of α, with β as a parameter, for room temperature data.

$$T_{calc.}/T_{true} = b + B \log(\rho) \tag{12}$$

If b were one and B zero, the model would be perfect.

The value of β in Eq. (11) was varied from 1.1–1.6. For each value of β a curve representing the change in the value of α versus the change in the value of the coefficient of the density dependence B was drawn. This is shown in Fig. 3. B exhibits a minimum for each value of α. For this particular value of α, the average value of the temperature ratio $T_{calc.}/T_{true}$, the standard error in the value of the temperature ratio and the value of the coefficient B are computed. The values obtained are tabulated in Table 1. It may be seen that the value of the coefficient B decreases as the value of β is increased. To be able to choose a particular value for β, the same method of data analysis was employed using Ashkenas' data. This data covers two temperature regions, the room temperature region (300 K), and the liquid nitrogen region (78 K). The density varies from 10^{-5}–10^{-9} g/cm^3 in each temperature region. The data amounts to about 200 spectra.

Table 2 summarizes the results for Ashkenas' data. For the liquid nitrogen temperature region the temperature ratio $T_{calc.}/T_{true}$ varies from 1.04 to about 0.987 as the value of β varies from 1.1–1.6. The value of the standard error as well as the value of the coefficient B decreases as β is increased. For the room temperature data, the value of the temperature ratio, the

Table 2 Variations of predicted temperature for selected values of β and α (Ashkenas' data)

Exponent β	Value of α	Calculated value of coefficient B		$T_{calc.}/T_{true}$ average		σ Standard error in $T_{calc.}/T_{true}$	
		300 K	78 K	300 K	78 K	300 K	78 K
1.1	10.0	0.0346	0.0217	1.0249	1.0417	±0.0269	+0.0498
1.2	4.5	0.0317	0.0094	1.0238	1.032	±0.0258	±0.0484
1.3	4.0	0.0290	−0.0010	1.0227	1.0224	±0.0249	±0.0476
1.4	3.5	0.0267	−0.0090	1.0215	1.0129	±0.0241	±0.0470
[a]1.45	3.0	0.0259	−0.0108	1.0202	1.0049	±0.0237	±0.0468[a]
1.5	2.875	0.0251	−0.0125	1.0206	1.0039	±0.0238	±0.0482
1.6	2.00	0.0241	−0.0125	1.0173	0.9870	±0.0237	±0.0458

[a] These values represent the best fit for both room temperature and 78 K data.

Table 3 Comparison of models

	Composite model	Q-D model	D-D model
Coefficient B			
Present data	0.0157	0.0276	0.0308
Ashkenas 300 K	0.0259	0.0361	0.0389
Ashkenas 78 K	−0.0108	0.0373	0.0363
Average $T_{\text{calc}}/T_{\text{true}}$			
Present data	1.0174	0.9904	1.0454
Ashkenas 300 K	1.0202	1.0070	1.0633
Ashkenas 78 K	1.0049	0.9388	1.1659
σ standard error in $T_{\text{calc}}/T_{\text{true}}$			
Present data	± 0.0208	± 0.0283	± 0.0310
Ashkenas 300 K	± 0.0237	± 0.0294	± 0.0287
Ashkenas 78 K	± 0.0468	± 0.0533	± 0.0544

standard error σ in the temperature ratio, and the value of the coefficient B all decrease as the value of β is increased.

From the previous two tables given it appears that the best choice for the value of β is 1.45. This choice is a compromise between contending with the approximate correct value of the temperature ratio $T_{\text{calc}}/T_{\text{true}}$ for the liquid nitrogen temperature region and the decreasing value of the coefficient B and the standard error σ in the measurement of the temperature ratio. This value of β is in reasonable agreement with the value obtained from Harbour's analysis which was quoted earlier.

Table 3 is given to facilitate the comparison of results obtained using the composite model vs the results obtained using the two models described earlier in this study. It is clear that the dependence of the predicted temperature on the true gas temperature has been largely eliminated and the standard error σ in the measured temperature ratio $T_{\text{calc}}/T_{\text{true}}$ has been reduced. The dependence of the measured temperature on the gas density has been significantly reduced but not eliminated. A change of three orders of magnitude in the gas density will produce a maximum uncertainty of about 4% which is comparable to the standard error in the measurements. More data are necessary before the final value of the density coefficient can be determined. Figures 4 and 5 represent a least-squares line fit of the temperature ratio $T_{\text{calc}}/T_{\text{true}}$ vs the logarithm of the density ρ in g/cm^3 for Ashkenas' liquid nitrogen temperature data using the composite model and the Q-D model, respectively. This is the worst case for all the data used in this study. The individual points are also shown.

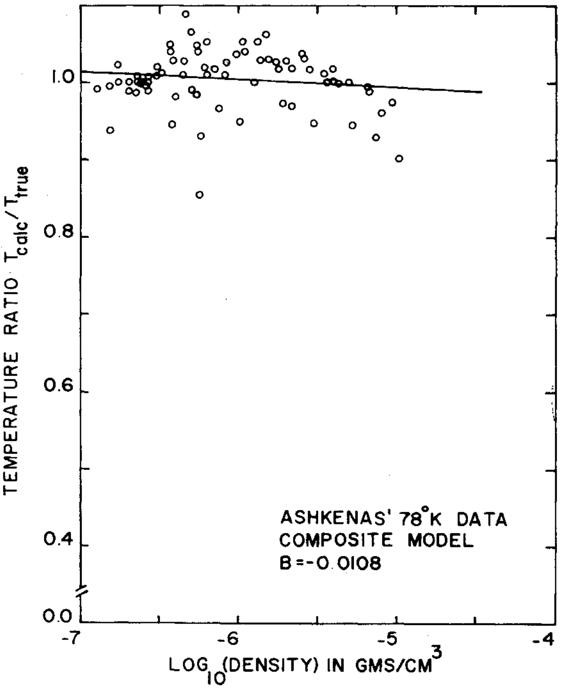

Fig. 4 Least squares straight line fit of $T_{\text{calc}}/T_{\text{true}}$ vs $\log_{10}$ (ρ) for Ashkenas' liquid nitrogen data, using the composite model.

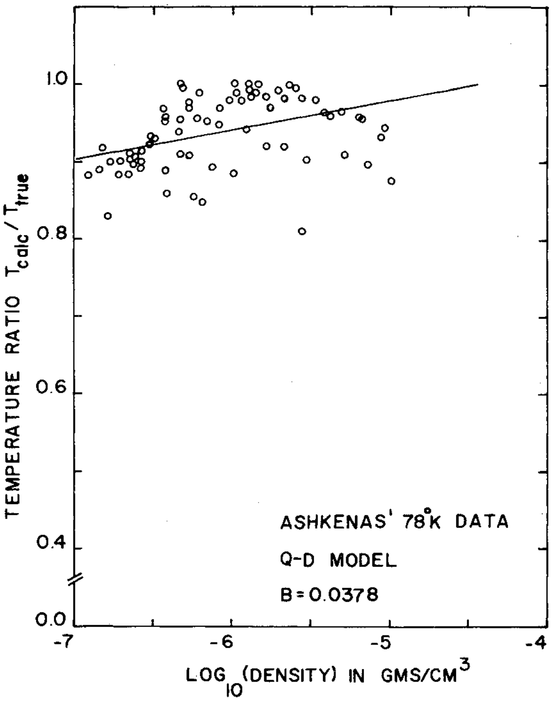

Fig. 5 Least-squares straight line fit of $T_{\text{calc}}/T_{\text{true}}$ vs $\log_{10}$ (ρ) for Ashkenas' liquid nitrogen data, using the Q-D model.

In summary, it is evident that the composite model may explain simply, but with sufficient accuracy, the effects that the re-excited ions have on the measurement of rotational temperature using electron beam induced fluorescence. By accounting for the emission from the ions, the value of the calculated rotational temperature becomes independent of the true gas temperature for temperatures of 300 K and 78 K. Similarly, the dependence of the calculated temperature on the gas density has been almost completely eliminated.

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