Research Article

On the Pressure and Temperature Dependence of the Absorption Coefficient of NH_3

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The effects of pressure and temperature on the absorption coefficient of ammonia (NH_3) gas self-perturbed and perturbed by nitrogen (N_2) gas have been measured. We varied the gas pressure from 10 to 160 Torr and the temperature from 235 to 296 K in order to study the absorption coefficient at the center and the wings of lines in the ν_4 band of NH_3. These measurements were made using a high resolution ($0.0038 \ cm^{-1}$) Bruker Fourier-transform spectrometer. These spectra have been analyzed using the method of multipressure technique permitting to succeed to an evolution of the absorption coefficient with the pressure and the quantum numbers J and K of the NH_3 molecule. The results show that the absorption coefficient varies as a quadratic function of the pressure at the center of a given line. However, it has a linear evolution in the wings of the line. Moreover, the absorption coefficients are inversely proportional to temperature in the wings when NH_3 lines are broadened by N_2. The retrieved values of these coefficients were used to derive the temperature dependence of N_2 broadening NH_3 lines. The absorption coefficients were shown to fit closely the well-known exponential law.

1. Introduction

The infrared spectroscopic investigations of the atmospheres of stars, planets, and their satellites, using infrared spectroscopy, not only provide valuable information about the chemical elements that they consist of, but also about the horizontal and the vertical distribution of their minor constituents. Due to the complexity of the line profiles used to model the spectral shapes (absorption, broadening, intensity), it is necessary to determine experimentally the line parameters of the spectra in order to test the models being used.

Several studies in the literature have investigated the spectral properties of NH_3 in several infrared bands. Aroui et al. [1] have studied the self-broadening and line intensities, Nouri et al. [2] have studied the temperature dependence of pressure broadening, and other authors [3, 4] were interested in the absorption coefficient at the line centers of NH_3. Experimentally absorption coefficients for broadband ArF excimer radiation laser were determined for NH_3 at temper-atures up to 3500 K [5]. Measurements of the NH_3

absorption coefficients at CO_2 laser wavelengths have been done by Zelinger et al. [5] using photoacoustic spectroscopy. NH_3 absorption coefficients were also measured by Allario and Seals [6] using several transitions of a CO_2 laser for small concentrations of NH_3 perturbed by N_2. The influence of CO_2 Laser line width on the measured absorption coefficients of atmospheric ammonia has been studied by Voitsekhovskaya et al. [7].

The focus of the present study is to present absorption line profile measurements of NH_3 in the $6 \ \mu m$ region ($^P P$ branch of the ν_4 band). In this range, we resolved the spectra for different J and K quantum numbers. We have determined the absorption coefficients in the centers and in the wings of NH_3 lines self-perturbed and perturbed by N_2 at various pressures (10–160 Torr). The measurements were made for different gas temperatures 235, 245, 268, and 296 K. The analysis was made as a function of J and K quantum numbers, and the results were compared to the previous investigations.

Studied gas	Temperature (K)	Pressure of the perturbed gas (mbar)	Cell length (cm)
NH_3–NH_3	296	10~141	2.5
NH_3–N_2	235, 245, 268, 296	10~141	15

2. Experimental

The measurements were made using a high-resolution Bruker Fourier transform spectrometer (Bruker IFS 120 HR) [8, 9]. This spectrometer is equipped with different components: a Globar source, a KBr beam splitter, a filter eliminating infrared radiation above 2500 cm^{-1}, and a photovoltaic HgCdTe detector cooled at 77 K by liquid nitrogen.

The spectral resolution was about 0.0038 cm^{-1} after apodization with a triangular function. This value is equal to the half-width at half maximum of the apparatus function approximated in the calculations by a Gaussian shape. This approximation has a negligible contribution to systematic errors since the pressure-broadened lines were much larger [2, 10]. Ammonia gas in natural abundances was provided by Air Liquid France with stated purity of 98.5%. The spectra were measured at different pressures covering the lines of the ^PP branch of the ν_4 band of ammonia. NH_3 and N_2 gases were contained in a metallic cell with a 2.5 cm path length for NH_3 self-perturbed, and in a Pyrex glass cell with a 15 cm path length for NH_3 perturbed by N_2. Both of the absorption cells are sealed by CaF_2 windows.

The pressures of the gases were measured accurately using three calibrated capacitive MKS Baratron transducers with full-scale readings of 10, 100, and 1000 mbar. The accuracy of these manometers is 0.5% of the readout. Ammonia sample pressures were allowed to stabilize for sufficient time before the spectrum of the sample was finally recorded.

For NH_3 self-perturbed, the pressure was varied from 5 to 120 mbar, whereas for NH_3 perturbed by N_2 we varied the gas pressure from 10 to 160 Torr. For the present experiments, the temperatures varies from 235 to 296 K. Each temperature was monitored by three calibrated thermocouples (Pt-100).

The measured intensity of the incident light (I_0) and the transmitted light (I) at wave-number σ were converted to transmission spectra $\tau(\sigma, T)$ using Lambert-Beer's law

$$\tau(\sigma, T) = \frac{I(\sigma, T)}{I_0(\sigma, T)}. \tag{1}$$

Table 1 summarizes the experimental parameters. The spectral region for this study is between 1470 and 1600 cm^{-1}. Figure 1 shows short transmittance spectra of NH_3 around 1550 cm^{-1} exhibiting some prominent lines of the ^PP branch. These spectra were recorded at $T = 296$ K. At low pressure, the lines are separated, but when the pressure increases the lines widen and begin to overlap.
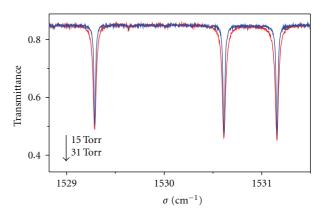

Figure 1: Transmittance spectrum of NH_3 around 1550 cm^{-1} showing some lines of the ^PP branch of the ν_4 band. The pressures of NH_3 are 15 and 31 mb.

3. Fitting Procedures

The absorption coefficient of an isolated line of the ν_4 band is obtained by comparing the recorded line with a synthetic line. The adjustment is performed using the Rosenkranz profile given [11, 12] by

$$\alpha(\sigma) = \frac{P_{NH_3}}{\pi} \sum_{\text{line } k} S_k \frac{PY_k(\sigma - \sigma_k) + P\gamma_k}{(\sigma - \sigma_k)^2 + (P\gamma_k)^2}, \tag{2}$$

where P_{NH_3} is the NH_3 pressure, P is the partial pressure of the perturbing gas, k represents the line $\nu_i J_i \rightarrow \nu_f J_f$, S_k its intensity, σ_k its wave number including the collisional shift δ ($\delta = \sigma_k - \sigma_0$, σ_0 the unperturbed or zero pressure wave number), γ_k its broadening coefficient, and Y_k its mixing parameter.

The modelled transmission $\tau(\sigma)$ is the result of a convolution of $\alpha(\sigma)$ with a Doppler profile (α_{Dop}) and the apparatus function of the spectrometer (F_{App}) [10].

$$\tau(\sigma)$$
$$= \int_{-\infty}^{+\infty} F_{App}(\sigma - \sigma') \exp\left[-l \int_{-\infty}^{+\infty} \alpha_{Dop}(\sigma' - \sigma'')\alpha(\sigma'')d\sigma''\right]d\sigma', \tag{3}$$

where l represents the length of the absorption cell.

The differences between the experimental and calculated spectra were minimized by adjusting the parameters PS_k, σ_k, $P\gamma_k$, and PY_k using a nonlinear least-squares multipressure fitting technique where all spectra at various pressures are successively adjusted using (3). An example of multipressure fit in the case of $^PP(7, 3)_s$ line for four NH_3 pressures (30, 60, 91, and 120 mbar) is shown in Figure 2. Residual (measured minus calculated) spectra are shown in the lower part of graphs.

As illustrated by this figure, the theoretical model given by (3) proves sufficiently accurate to fit very well the observed spectral lines without accounting for Dicke narrowing and/or speed dependence and demonstrates at the same time that the line coupling cannot be neglected.
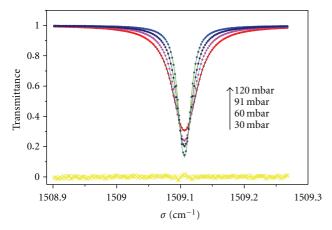
FIGURE 2: Results of multipressure fits for the $^PP(7,3)_s$ line in the ν_4 band of NH_3 self-perturbed at 296 K. (—) and (•) are the measured and calculated values, respectively. Residual (measured minus calculated) spectra are shown in the lower part of the graphs.
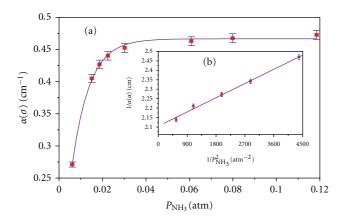

FIGURE 3: Variation of the absorption coefficient and its inverse in the center of the $^PP(8,3)_s$ line versus P_{NH_3} and $1/P_{NH_3}^2$, respectively, at $T = 296$ K.

4. Results Analysis

4.1. Line Center Absorption Coefficients. We have made measurements of the absorption coefficients for 60 isolated lines of NH_3 in the spectral region between 1470 and 1600 cm^{-1}. For illustration, the evolution of the absorption coefficient à $T = 296$ K for the $^PP(8,3)_s$ line at various pressures of NH_3 is shown in Figure 3(a).

In the low-pressure regime, we notice that the absorption coefficient of a given line varies as a quadratic function of pressure as given by the following equation:

$$\alpha = C^{te}P_{NH_3}^2, \tag{4}$$

where C^{te} is a constant value depending on the individual line.

This figure shows that at high-pressure region, the absorption coefficient tends towards a constant value and hence becomes independent of pressure.
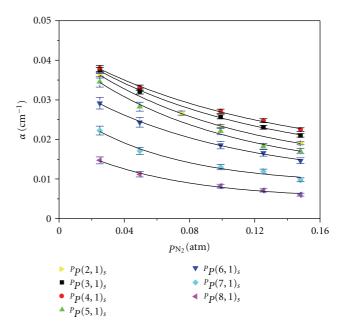

FIGURE 4: Variation of the absorption coefficient in the line center versus the N_2 pressure (P_{N_2}) for the $^PP(J,1)_s$ lines.

These results are in agreement with those published in [4], where the author studied some lines of the ν_2 band of NH_3.

The inverse of the absorption coefficient $(1/\alpha)$ as a function of the square of the NH_3 pressure is plotted in Figure 3(b) for the $^PP(8,3)_s$ line. Indeed, one can see that these two parameters vary linearly. The slope of the obtained straight line is related to the line intensity and self-broadening coefficient of the considered line [4].

Results obtained for NH_3 perturbed by N_2, show that the peak absorption coefficient, for the 60 lines studied in this work, is decreasing with the N_2 pressure. Figure 4 illustrates the variation of the line center absorption coefficients versus the N_2 pressure for the $^PP(J,1)_s$ lines at $T = 296$ K. This figure also shows that the absorption coefficient α decreases with the pressure of the perturbing gas. Moreover, it illustrates that α decreases with quantum number J for a given K. For example, the α of $^PP(2.1)_s$ line is greater than that of $^PP(8,1)_s$ line.

Figure 5 represents the evolution of the absorption coefficient α with the quantum number J in the case of NH_3–NH_3 gas mixture at $T = 296$ K and $P = 32$ Torr. The coefficient α has a maximum at $J = 4$ for the $^PP(J,1)$ lines and decreases monotonically for $K \geq 2$ with increasing J.

According to the literature [10, 13], the line intensity S_0 depends on the statistical weight factor g_s, which is related to the quantum numbers J and K. The intensities of the transition having a statistical weight $g_s = 2$ (i.e., for $K = 3n$, $n = 1, 2, 3, \ldots$) are generally higher than of those lines having a statistical weight $g_s = 1$ for $K \neq 3n$. As illustrated by Table 4, one can conclude that the absorption coefficients and the line intensities vary in the same way as a function of the quantum numbers.

TABLE 2: Self-absorption coefficient of the two wings of $^PP(5,2)_s$ and $^PP(7,4)_s$ lines as a function of the square of NH_3 pressure ($P^2_{NH_3}$) at $T = 296$ K.

$P^2_{NH_3} \times 10^4$ (atm^2)	$\alpha \times 10^4$ (cm^{-1})			
	$^PP(5,2)_{s\text{ left}}$	$^PP(5,2)_{s\text{ right}}$	$^PP(7,4)_{a\text{ left}}$	$^PP(7,4)_{a\text{ right}}$
2.28	14.9	14.4	—	1.64
18	61	—	48.5	—
37	123.8	125.9	80.5	81.5
63	191.5	191.5	122.1	121.5
140	384.4	382.3	256.5	256.3

TABLE 3: Absorption coefficient of two wings of the $^PP(7,2)_a$ for NH_3–N_2 mixtures as a function of N_2 pressure (P_{N_2}) at $T = 268$ K.

$P_{N_2} \times 10^3$ (atm)	$\alpha/P_{NH_3} \times 10^2$ (cm^{-1} atm^{-1})	
	$^PP(7,2)_{a\text{ right}}$	$^PP(7,2)_{a\text{ left}}$
29.6	15.1	13.8
59.2	19.4	18.4
89.6	24.1	23.9
121.9	27.7	26.7
138.9	30.8	30.1

FIGURE 6: Variation of self-absorption coefficient in the right wings of $^PP(5,2)_s$ and $^PP(7,4)_a$ lines as a function of the square of NH_3 pressure ($P^2_{NH_3}$): Case of NH_3–NH_3 gas mixture at $T = 296$ K.
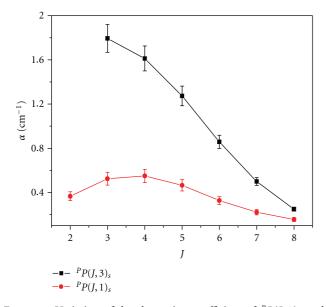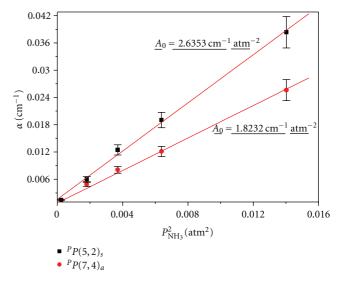

FIGURE 5: Variation of the absorption coefficient of $^PP(J,3)_a$ and $^PP(J,1)_s$ lines with the quantum numbers J and K. Case of NH_3–NH_3 gas mixture at $P = 32$ Torr for $T = 295$ K.

4.2. Line Wing Absorption Coefficients. According to our analysis, we observe a quadratic dependence of the absorption coefficient as a function of pressure of the NH_3 gas that can be modeled as follows [14]:

$$\alpha = A_0(\sigma, T)P^2_{NH_3}, \qquad (5)$$

$A_0(\sigma, T)$ is the normalized absorption coefficient depending on the temperature and the wave number σ of the line; P_{NH_3} is the NH_3 pressure. Table 2 gives the self-absorption coefficient of the two wings of $^PP(5,2)_s$ and $^PP(7,4)_s$ lines

as a function of the square of NH_3 pressure ($P^2_{NH_3}$) at $T = 296$ K. Figure 6 shows the variation of self-absorption coefficient of the right wings of $^PP(5,2)_s$ and $^PP(7,4)_a$ lines as a function of the square of NH_3 pressure ($P^2_{NH_3}$). One can see that the wing absorption coefficients increase linearly with pressure.

All the self-absorption coefficients measured for the pressure and temperature ranges considered in this study validate the above quadratic pressure dependence given by (5).

For NH_3–N_2 gas mixtures, a sample of the absorption coefficient α for the two wings of the $^PP(7,2)_a$ line as a function of N_2 pressure (P_{N_2}) at $T = 268$ K is given by Table 3. This table reveals an increasing of α with P_{N_2}. So the coefficient α can be described by the following equation [15]:

$$\alpha = A_0(\sigma, T)P^2_{NH_3} + B_0(\sigma, T)P_{NH_3}P_{N_2}, \qquad (6)$$

where $B_0(\sigma, T)$ is the normalized absorption coefficient.

The $A_0(\sigma, T)$ and $B_0(\sigma, T)$ parameters were determined using (6) and the absorption coefficient values for the right and left wings of the each line considered in this work. The results at $T = 296$ K are given in Table 4 for 32 ro-vibrational antisymmetric lines in the ^PP branch of the ν_4 band along with the estimated errors given in parentheses.

TABLE 4: Values of the normalized absorption coefficients A_0 and B_0 for NH_3 broadened by N_2 at $T = 296$ K.

Transition	σ (cm^{-1})	NH_3–NH_3		NH_3–N_2	
		A_0 (cm^{-1} atm^{-2}) (σ_{left})	A_0 (cm^{-1} atm^{-2}) (σ_{right})	B_0 (cm^{-1} atm^{-2}) (σ_{left})	B_0 (cm^{-1} atm^{-2}) (σ_{right})
$^PP(2,1)_a$	1591.1050	1.467 (0.098)	1.493 (0.063)	2.731 (0.437)	2.448 (0.245)
$^PP(2,2)_a$	1595.0801	6.759 (0.348)	1.045 (0.069)	3.228 (0.233)	2.799 (0.303)
$^PP(3,1)_a$	1572.4882	3.282 (0.139)	3.435(0.065)	2.807 (0.100)	2.956 (0.542)
$^PP(3,3)_a$	1579.6363	16.39 (0.431)	2.216 (0.047)	3.859 (0.274)	4.176 (0.183)
$^PP(4,1)_a$	1554.7340	2.065 (0.047)	2.009 (0.050)	3.490 (0.329)	3.006 (0.207)
$^PP(4,2)_a$	1558.2494	1.344 (0.046)	1.370 (0.011)	2.815 (0.269)	2.923 (0.218)
$^PP(4,3)_a$	1561.3835	4.217 (0.086)	4.1741 (0.150)	9.782 (0.707)	9.632 (0.462)
$^PP(4,4)_a$	1564.0825	10.40 (0.186)	0.566 (0.042)	2.338 (0.203)	5.111 (0.511)
$^PP(5,1)_a$	1538.0102	0.860 (0.083)	0.838 (0.058)	3.693 (0.372)	3.343 (0.171)
$^PP(5,2)_a$	1541.0041	2.520 (0.075)	2.272 (0.021)	2.883 (0.540)	2.893 (0.534)
$^PP(5,3)_a$	1543.8550	2.468 (0.07)	2.990 (0.029)	5.638 (0.248)	5.391 (0.583)
$^PP(5,4)_a$	1546.3315	5.805 (0.194)	4.610 (0.270)	4.829 (0.028)	4.013 (0.539)
$^PP(5,5)_a$	1548.4290	13.527 (0.381)	2.471 (0.142)	2.920 (0.220)	3.510 (0.208)
$^PP(6,1)_a$	1522.3847	1.370 (0.033)	1.518 (0.024)	2.020 (0.384)	1.021 (0.134)
$^PP(6,2)_a$	1522.7763	—	—	2.006 (0.399)	1.894 (0.064)
$^PP(6,3)_a$	1527.0615	2.822 (0.093)	2.583 (0.053)	4.232 (0.364)	4.313 (0.372)
$^PP(6,4)_a$	1529.2897	1.524 (0.015)	1.553 (0.058)	2.424 (0.146)	2.172 (0.221)
$^PP(6,5)_a$	1531.1590	1.571 (0.07)	1.4164 (0.056)	3.784 (0.522)	3.003 (0.625)
$^PP(6,6)_a$	1532.6831	20.697 (0.570)	2.250 (0.118)	4.430 (0.076)	9.116 (0.919)
$^PP(7,2)_a$	1509.4479	1.818 (0.053)	1.863 (0.108)	1.415 (0.044)	1.445 (0.055)
$^PP(7,3)_a$	1511.3125	1.169 (0.064)	1.510 (0.047)	1.183 (0.195)	1.312 (0.247)
$^PP(7,4)_a$	1513.0439	1.823 (0.094)	1.888 (0.074)	1.657 (0.131)	1.431 (0.442)
$^PP(7,5)_a$	1514.6055	2.319 (0.176)	2.145 (0.141)	1.810 (0.157)	1.500 (0.104)
$^PP(7,6)_a$	1515.8819	3.013 (0.035)	3.074 (0.162)	2.200 (0.179)	1.946 (0.226)
$^PP(7,7)_a$	1516.8521	8.994 (0.217)	1.248 (0.070)	2.080 (0.120)	2.632 (0.091)
$^PP(8,1)_a$	1494.2421	0.375 (0.038)	0.334 (0.009)	1.116 (0.113)	1.180 (0.101)
$^PP(8,2)_a$	1493.5269	—	—	1.145 (0.142)	1.328 (0.215)
$^PP(8,3)_a$	1496.1740	0.546 (0.105)	0.597 (0.072)	2.357 (0.153)	2.785 (0.261)
$^PP(8,4)_a$	1497.6750	1.982 (0.104)	2.123 (0.071)	4.054 (0.261)	4.521 (0.276)
$^PP(8,6)_a$	1499.8028	2.199 (0.022)	2.877 (0.264)	2.710 (0.236)	2.499 (0.917)
$^PP(8,7)_a$	1500.5103	11.869 (1.307)	10.742 (1.414)	—	—
$^PP(8,8)_a$	1500.9439	7.227 (0.231)	2.221 (0.335)	1.423 (0.349)	1.799 (0.368)

These errors correspond to the statistical errors expressed as one-time standard deviation for all spectroscopic parameters determined in this work. As expected, the errors vary widely with the lines. The stronger and less blended ones are better determined. For the lines which are too weak or too strongly blended, no reliable fit could be obtained. For this reason, some lines have been disregarded. The assignments of the lines considered in this table are taken from [16]. Within the experimental errors, as seen by Table 4, the values of $A_0(\sigma, T)$ and $B_0(\sigma, T)$ are practically identical for the two wings.

Figure 7 shows the variation of the absorption coefficient with the N_2 pressure for the two wings of the $^PP(7,2)_a$ line for NH_3–N_2 mixtures at $T = 268$ K. As shown by this figure, the variation of the absorption coefficient divided by the NH_3 pressure (α/P_{NH_3}) with the N_2 pressure is linear. Also, we observe that the values of α/P_{NH_3} and $A_0(\sigma, T)$ parameters are practically the same for the left and right wings of the lines.

4.3. Temperature Dependence. Spectra of the ν_4 band of NH_3 perturbed by N_2 were recorded at four temperatures 235, 245, 268, and 296 K for different pressures of nitrogen (P_{N_2}). For these spectra, the pressure of NH_3 was fixed. Variation of the absorption coefficient α at the line center as a function of P_{N_2} for the $^PP(5,2)_s$ line for $T = 235$ and 265 K is shown by Figure 8 which illustrates a decreasing of the absorption coefficient with temperature.

According to Shi and Zhang and Bauer et al. [17, 18], the temperature dependence of absorption coefficient $\alpha(T)$ could be presented by the simple power law:
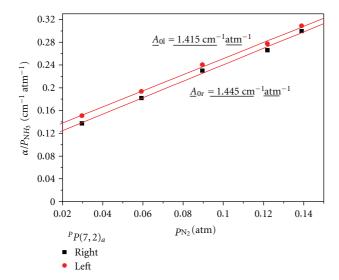
$$\alpha = \alpha(T_0)\left(\frac{T}{T_0}\right)^A, \quad (7)$$

FIGURE 7: Variation of α/P_{NH_3} versus the N_2 pressure (P_{N_2}) for the two wings of the ${}^P P(7,2)_a$ line perturbed by N_2 at $T = 268$ K.
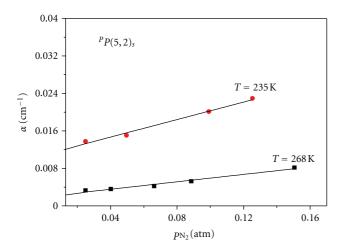

FIGURE 8: Variation of the absorption coefficient as a function of the N_2 pressure (P_{N_2}) for the ${}^P P(5,2)_s$ line for the temperatures $T = 235$ and 265 K.

where A is the temperature exponent and T_0 is the reference temperature. In our case $T_0 = 235$ K.

From the measured values of the absorption coefficients for the considered temperatures, one can determine the values of the exponent A as the slope of least square fits from the graphs of $\ln(T/\alpha(T_0))$ as a function of $\ln(T/T_0)$. The straight lines obtained for all the transitions considered in this work validate (7) within the indicated temperature range. Figure 9 illustrates the variation of $\ln[\alpha(T)/\alpha(235)]$ as a function of $\ln(T/235)$ for the ${}^P P(5,2)_a$ line. The variation is linear; the slope of the line obtained from a linear regression gives a temperature exponent $A = -0.619 \pm 0.016$.

This dependence is in agreement with the work performed by Bauer et al. [18] in their study of the temperature dependence of the absorption coefficient of H_2O transitions.
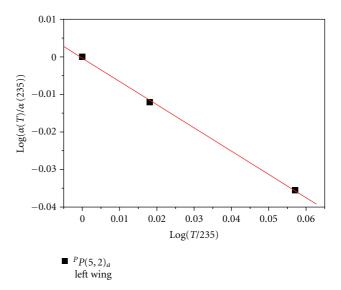

FIGURE 9: Variation of $\ln[\alpha(T)/\alpha(235)]$ as a function of $\ln(T/235)$ for the left wing of ${}^P P(5,2)_a$ line. The slope of the line obtained from a linear regression represents the temperature exponent $A = -0.619 \pm 0.016$.

5. Conclusion

Using spectra recorded using a Fourier transform spectrometer and a multispectrum fitting technique, we have determined the absorption coefficients at the center and in the wings of 60 lines pertaining to the ${}^P P$ branch of the ν_4 band of NH_3 as a function of NH_3 and N_2 pressures for different temperatures. The present results are in agreement with other recent measurements. In the low-pressure region, the self-absorption coefficient at the center of a given line varies with pressure as a quadratic function. At higher pressures, this coefficient tends to become constant. For NH_3–N_2 mixture, the values of the absorption coefficients exhibit a decreasing with N_2 pressure.

In the wings of the lines, these coefficients show an increasing with the square of NH_3 pressure, while for NH_3-N_2 gas mixtures they increase with N_2 pressure.

For this gas mixture, we have studied the temperature dependence of absorption coefficient which fits closely an exponential low. The temperature exponents of this law were derived for the ${}^P P(5,2)_a$ line.
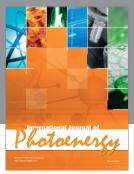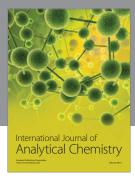
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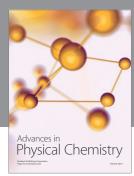
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[18] A. Bauer, M. Godon, J. Carlier, Q. Ma, and R. H. Tipping, "Absorption by H_2O and H_2O-N_2 mixtures at 153 GHz," *Journal of Quantitative Spectroscop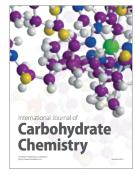y and Radiative Transfer*, vol. 50, no. 5, pp. 463–475, 1993.