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AN APPROXIMATION TO TRUE PEAK ABSORBANCE
FROM OBSERVED PEAK ABSORBANCE FOR
GAS PHASE FOURIER TRANSFORM SPECTROSCOPYKEVIN L. McNESBY
ROBERT A. FIFER

SEPTEMBER 1990

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I. INTRODUCTION

In our lab, we have been using low resolution FTIR absorption spectroscopy to measure rotational temperatures of gases from ambient to flame temperatures (300-2000 K) at total pressures below one atmosphere. By low resolution, we mean spectra obtained on instruments capable of achieving an optical retardation of less than 10 cm.

In order to obtain temperatures by fitting to a population distribution the peak absorbances of a group of rotational lines for which $\Delta v = \pm 1$, it is necessary to correct for distortions to peak absorbances brought about by the spectrometer. This distortion varies from line to line because of the dependence of the peak absorbances on the half width at half height (HWHH) of the absorbing line.¹ For a FTIR spectrometer using triangular apodization of the interferogram, the observed minimum transmittance, T_m , of a single pressure broadened line m with a Lorentzian absorption profile is given by:²

$$T_m = \int_{-\infty}^{\infty} \exp \left[-2.303 A_p \frac{\alpha_m^2}{(v - v_m)^2 + \alpha_m^2} \right] \frac{\Delta \sin^2 [\pi \Delta (v - v_m)]}{[\pi \Delta (v - v_m)]^2} dv \quad (1)$$

where

- T_m = Observed minimum transmittance
- α_m = Half width at half height (HWHH) of line m
- ν_m = Center of frequency of absorbance of line m
- Δ = Optical retardation
- A_p = True peak absorbance of line m at ν_m

Equation (1) is inconvenient to use because the true peak absorbance, A_p , is part of the integrand. A simple equation relating A_p , T_m , Δ , and α_m has been given by Griffiths:³

$$A_p = 0.91 A_{obs}^2 (2 \Delta \alpha_m)^{-2} \quad (2)$$

where $A_{obs} = -\log T_m$. This equation yields a good approximation to true peak absorbance for a strong absorbing line ($A_{obs} \approx 1$) being measured by a spectrometer whose resolution is nominally larger by an order of magnitude than α_m ; i.e., when the true linewidth is such that the reported lineshape is just the instrument lineshape.

Much of our work involves measuring absorbances under the conditions outlined above. However, for some low pressure flame work, the absorbances are very weak. Therefore, we have extended the approximation given in Reference 3 to include weakly absorbing lines and have obtained equations of the form of Eq. (2) for several common values of optical retardations. It should be emphasized that this approximation is only useful for pressure broadened spectra obtained with low resolution instruments using triangular apodization of the interferogram.

II. CALCULATIONS

We have fit input parameters and the result of the numerical integration of Eq. (1) to an equation of the form:

$$A_p = k A_{obs}^l \rho^n \quad (3)$$

where k , l , and n are constants and ρ is the spectral resolution parameter¹ and is equal to the ratio of the spectrometer resolution (given as $1/\Delta$) to twice α_m . The integral in Eq. (1) was evaluated numerically for all combinations of optical retardations of 1, 2, 4, 8, and 10 cm with true peak absorbances of 0.1 to 10 (in intervals of 0.1) with absorbing lines for which α_m ranged from 0.01 to 0.2 cm^{-1} (in intervals of 0.01 cm^{-1}). Equation (3) was linearized and a weighted linear least squares routine was used to fit the data.

The integration routine assumed that ν_m was located at 2000 cm^{-1} and evaluated Eq. (1) using Simpson's⁴ rule for 50 cm^{-1} on either side of ν_m . The interval to be numerically integrated was divided into 10,000 pieces. The choice of value as the center of frequency is arbitrary, as may be verified by substituting for the expression $\nu - \nu_m$ in Eq. (1). To check and see if numerical integration gave values which were dependent on the center of frequency in Eq. (1), a numerical integration for a range of centers of frequency from 500 to 3500 cm^{-1} showed the value of the integral to be independent of this value for the number pieces used in the integration.

Applying our numerical integration routine to the function $\sin^2 x/x^2$ (which is similar in behavior to Eq. (1) yet has an analytical solution) using similar parameters as in the numerical integration of (1) gave results within 0.001% of the analytical solution. We believe our numerical integration of Eq. (1) gives results of similar accuracy.

III. RESULTS

Table 1 gives values of k , l , and n which provide the best approximation to true peak absorbance from observed peak absorbance for a given range of true peak absorbances for a known linewidth of from 0.01 to 0.1 cm^{-1} and a given spectrometer resolution. The value obtained for A_p using Eq. (3) is typically within 10% of the value input into Eq. (1) prior to numerical integration. In general, the quality of the fit decreases as the range of A_p increases. The quality of the fit is improved if absorbing lines with α_m less than 0.03 cm^{-1} are ignored. We have chosen to include lines with α_m less than 0.03 cm^{-1} because our interest lies in obtaining the true absorbance for weakly absorbing lines at high temperature. In Table 1, some results for absorbance ranges are not reported for optical retardations of from 4 to 10 cm. This is because the fit of Eq. (3) to the data generated by the integration of Eq. (1) was judged to be too poor to be of use. In general, the average relative error $(1/N \{ |(A_p - A_{pd})| / A_{pd} \})$, where A_p is the value obtained from Eq. (3), A_{pd} is the value input into Eq. (1) and N is the number (> 100) of points used in the fit, had to be less than 0.1 for that approximation to be included in Table 1.

Table 1. Values of the Coefficients k , l , and n Used to Fit Eq. (1) in the Text to Eq. (3) in the Text. Standard deviations in parenthesis.

True Absorbance Range	Optical Retardation (cm)	$\ln k$	l	n	RMS Deviation from Actual Absorbance
0.1-1	1	0.211(.066)	1.287(.004)	1.208(.005)	0.011
0.1-1	2	0.298(.008)	1.227(.007)	1.111(.007)	0.02
0.1-1	4	0.455(0.12)	1.116(.011)	0.941(.011)	0.011
0.1-1	8	0.588(0.17)	1.077(0.15)	0.698(0.13)	0.049
0.1-1	10	0.611(0.18)	1.056(0.16)	0.616(0.13)	0.052
1-3	1	0.172(.008)	1.703(.004)	1.634(.011)	0.027
1-3	2	0.331(0.15)	1.661(.028)	1.539(.027)	0.068
1-3	4	0.625(.022)	1.571(.066)	1.301(.058)	0.161
1-3	8	0.906(.025)	1.428(.106)	0.927(.075)	0.256
1-3	10	0.956(.027)	1.426(.110)	0.835(.071)	0.265
1-5	1	0.122(.014)	1.818(.014)	1.748(.015)	0.054
1-5	2	0.326(.029)	1.813(.041)	1.665(.040)	0.149
1-5	4	0.748(.047)	1.734(.110)	1.375(.095)	0.38
1-5	8	1.113(.036)	1.655(.178)	0.982(.120)	0.565
3-7	1	0.045(.035)	1.938(.038)	1.862(.037)	0.089
3-7	2	0.343(.106)	1.908(.143)	1.724(.133)	0.322
3-7	4	1.076(.159)	1.419(.346)	1.060(.269)	0.756
3-10	1	0.036(.046)	1.980(.042)	1.892(.042)	0.162
3-10	2	0.418(.144)	1.966(.167)	1.731(.153)	0.591
3-10	4	1.255(.210)	1.536(.400)	1.073(.298)	1.288

For all ranges of values of A_p used in the integration of Eq. (1), the average deviation of the fitted approximation presented here was superior to that from Reference 3. In fairness, the authors of Reference 3 presented their approximation without derivation and in no way implied it to be valid over any range other than that specified in the paper. In addition, for the range over which the approximation of Reference 3 was used, that approximation and the fitted approximation developed here are in good agreement (see Table 1).

IV. CONCLUSION

We have used the fitted approximation to true peak absorbance given above in calculations on data obtained from FTIR absorption spectra to determine rotational temperatures of CO above 673 K with good results. In this case the correction was applied to the P and R branches of CO prior to fitting the peak absorbances to a Boltzmann distribution. The approximations listed above provide a fast and reasonably accurate way of determining true peak absorbances from observed peak absorbances at low resolution in the gas phase when the α_m of the absorbing line is known. However, when there is significant overlap of lines, such as for a Q-branch or for some flame spectra, more accurate methods must be used. Also, to our knowledge, the approximations provide the first fast method of determining true peak absorbances of individual absorption lines in the gas phase from low resolution FTIR spectra for which the observed peak absorbance is small (~ 0.1), a case we have encountered in dealing with absorptions in gases from levels other than the ground vibrational state.

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