Experimental Study of Temperature-Dependence Laws of Non-Voigt Absorption Line Shape Parameters

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Motivation

- Behavior of non-Voigt parameters with temperature remains unclear

- Test of the commonly used power law for temperature-dependence:

\[ X(T) = X(T_{\text{ref}}) \cdot \left( \frac{T_{\text{ref}}}{T} \right)^n \]

→ comparison with other temperature laws

- Test of lineshape theory
  - Ghysels et al.\(^1\):
    \[ a_w = \frac{\Gamma_2}{\Gamma_0} = (1 - n)^{\frac{2}{3}} \frac{m_p}{m_a} \frac{1}{1 + m_p/m_a} \]

Experiment

- CO$_2$ $\nu_3$ band perturbed by N$_2$

- Bruker IFS 125HR FT spectrometer

- Single pass absorption cell, L=0.22 m

9 Temperatures
5 Pressures
2 Mixing Ratios

- T = 190, 200, 220, 240, 260, 280, 296, 310, 330 K
- p = 10, 30, 100, 300, 1000 mbar
- 2 different mixing ratios of CO$_2$ – N$_2$
- pure CO$_2$ spectra to characterize ILS at each T

- MOPD: 1.2 m (1000, 300 mbar), 2.0 m (100 mbar), 2.5 m (30, 10 mbar)
Analysis

- Multispectrum fits in pressure at each temperature

- Instrumental Line Shape (ILS) characterized with LINEFIT software by Hase et al.¹

- Quadratic Speed-Dependent Hard Collision model implemented as in Ngo et al.²

- Fitted $\sigma$, $S$, $\gamma_0$, $\gamma_2$, $\delta_0$, $\delta_2$, $v_{VC}$, $Y_0$

Results – $\gamma_0$

- Power law $\gamma(T) = \gamma(T_{\text{ref}}) \cdot \left(\frac{T_{\text{ref}}}{T}\right)^n$ introduces 0.2% error on linewidths on average
- Power law works well for $\gamma_2$
- $\gamma_0$ and $\gamma_2$ exhibit different behavior in temperature: $\gamma_2/\gamma_0 \neq \text{const.}$
- Assuming $n(\gamma_0) = n(\gamma_2)$ leads to error of $\sim 13\%$ on average in $\gamma_2$ (at 190 K)
Results – $\delta_0$

- Linear law recommended: $\delta(T) = \delta_0(T_{ref}) + \delta'(T - T_{ref})$
Results – $\nu_{VC}$

- Power law is suitable
- $n(\nu_{VC}) = 1.2$ on average,
  → close to theoretical expectation $n(\nu_{VC}) = 1$

\[
\nu_{VC} = \frac{kT}{2\pi cmD} \quad | \quad D \sim T^2
\]

\textsuperscript{1}JQSRT 129, 89 (2013)
Summary

- Evaluation of non-Voigt line shape parameters over 140 K range based on experiments with CO$_2$ perturbed by N$_2$

- Rotational quantum number dependence of non-Voigt temperature-dependencies accessible

- Errors introduced by different temperature-dependence models become quantifiable

- Distinct temperature-dependencies observed for $\gamma_0$ and $\gamma_2$

- Power law suitable to describe temperature-dependence of all parameters, except $\delta_0$
Results – $Y_0$

- Power law better suited than linear law
Error contributions – example: $\gamma_2$