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:

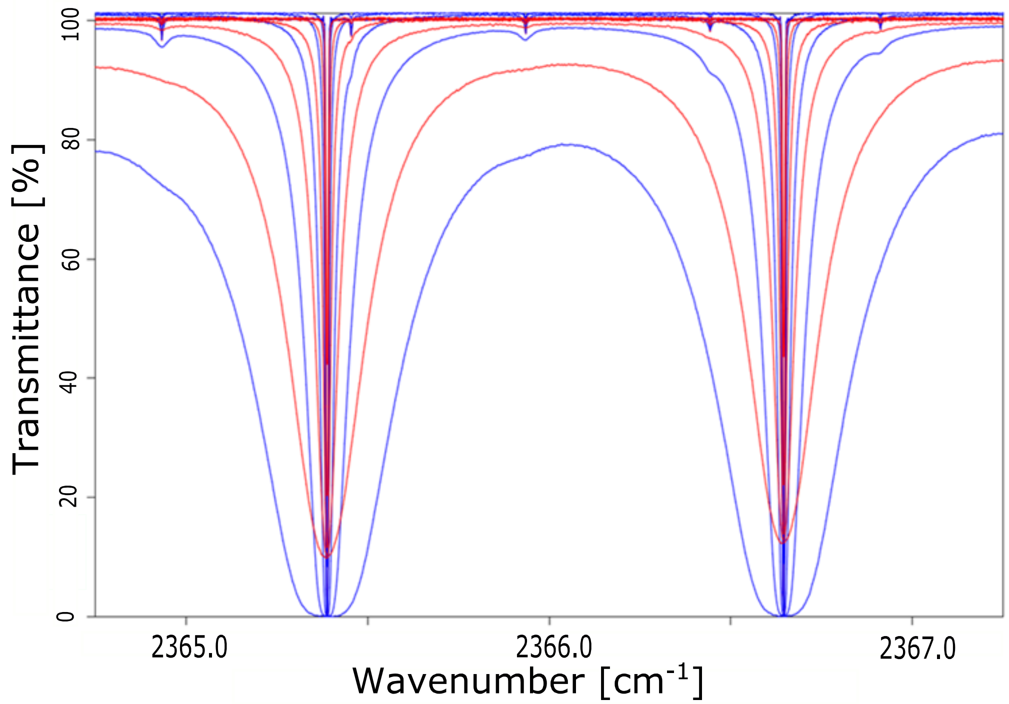
$$X(T) = X(\mathrm{T_{ref}}) \cdot \left(\frac{T_{ref}}{T}\right)^{n}$$

→ comparison with other temperature laws

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

[1] Appl. Phys. B **123**, 124 (2017)

Experiment



- CO_2 ν_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)

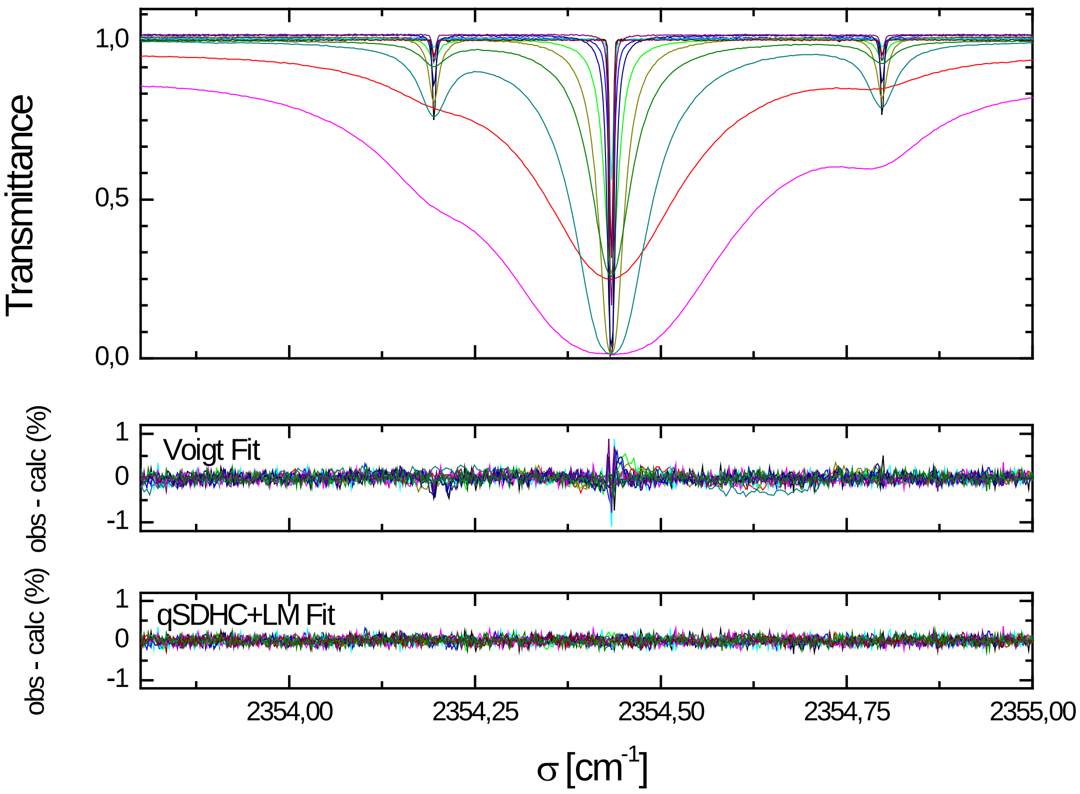
Analysis

- Multispectrum fits in pressure at each temperature

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

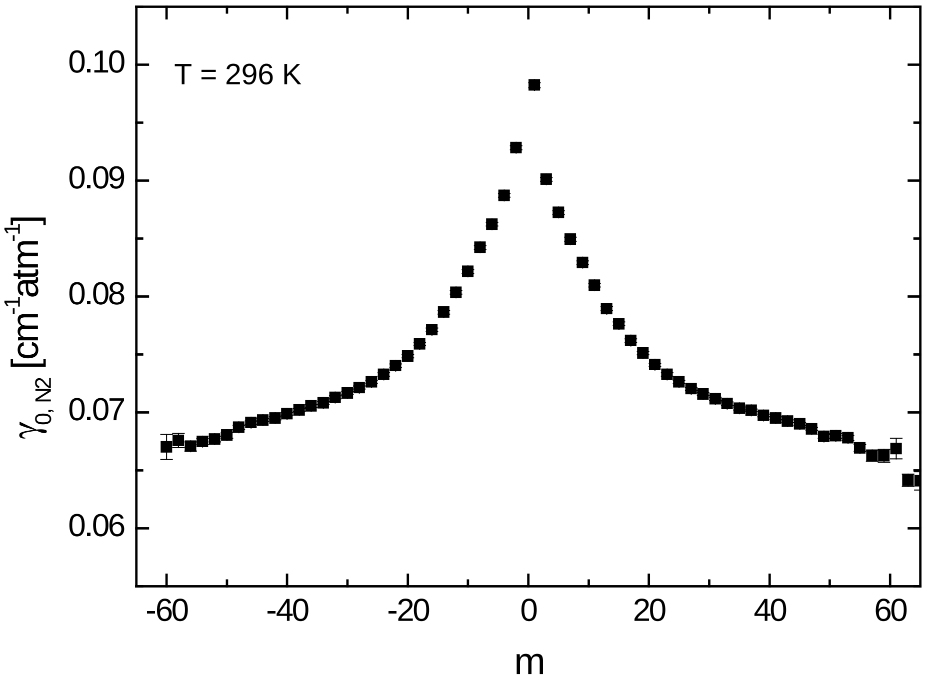
- Quadratic Speed-Dependent Hard Collision model implemented as in Ngo et al.[2]

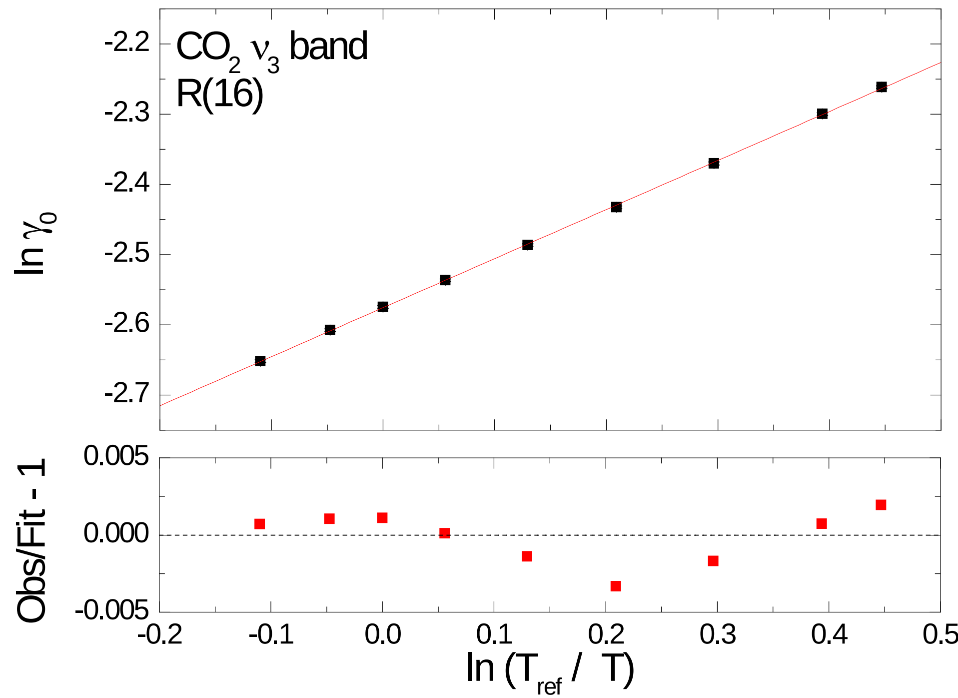
- Fitted
  σ, S, γ_0, γ_2, δ_0, δ_2, ν_{VC}, Y_0



[1] Appl. Opt. **38**, 3417-3422 (1999) [2] JQSRT **134**, 105 (2014)

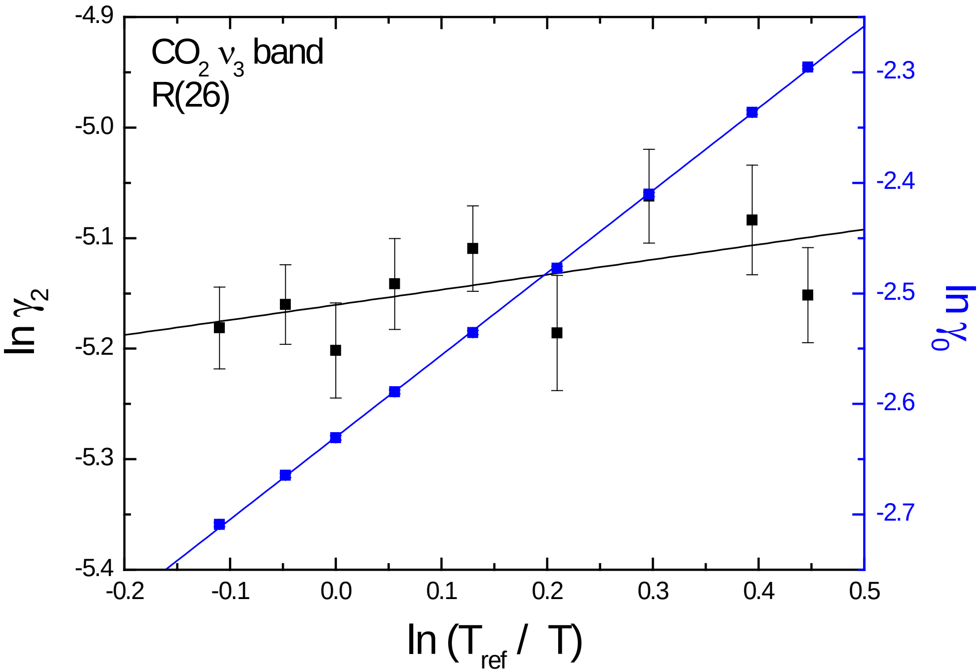
Results – γ_0

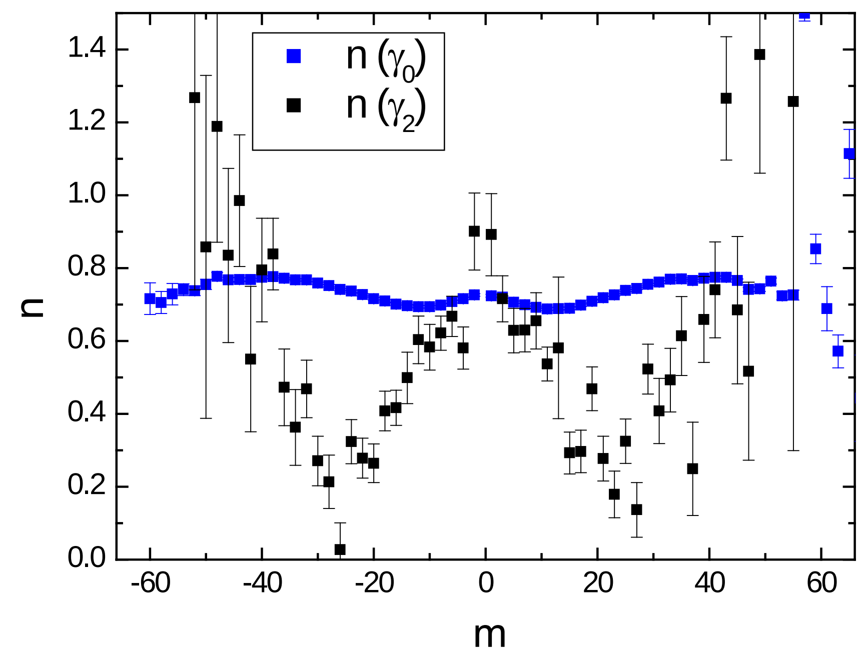




- Power law $\gamma(T) = \gamma(\mathrm{T_{ref}}) \cdot \left(\frac{T_{ref}}{T}\right)^n$ introduces 0.2% error on linewidths on average

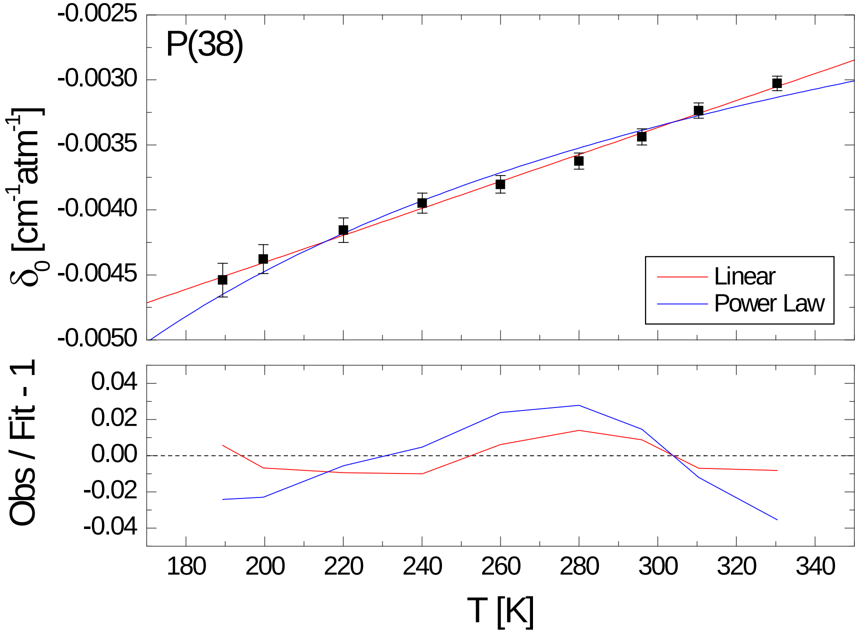
Results – γ_2

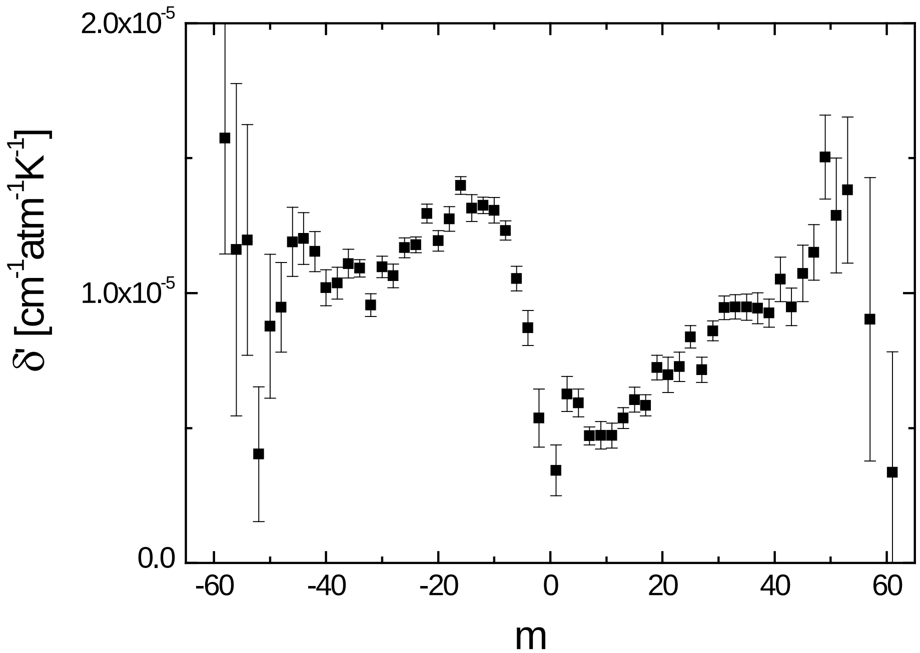




- Power law works well for γ_2
- γ_0 and γ_2 exhibit different behavior in temperature: $\boldsymbol{\gamma_2/\gamma_0 \neq const.}$
- Assuming $n(\gamma_0) = n(\gamma_2)$ leads to error of ~13% on average in γ_2 (at 190 K)

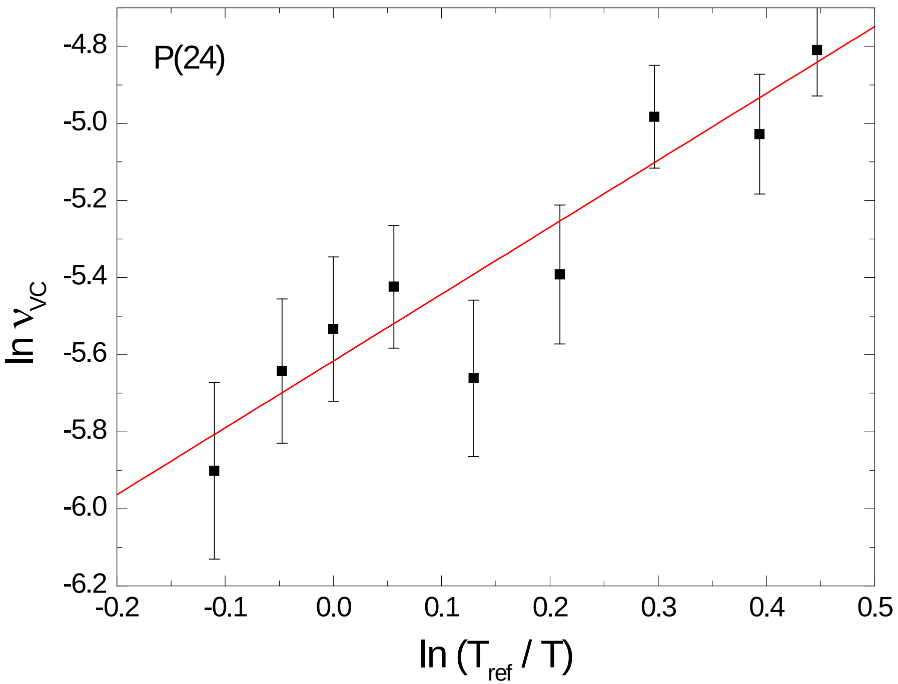
Results – δ_0

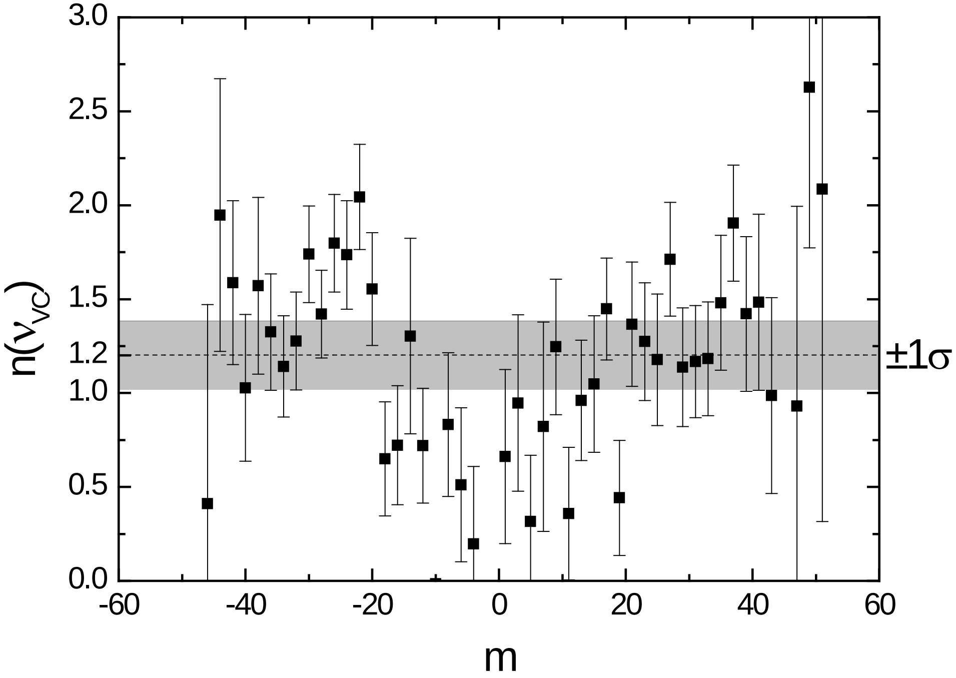




- Linear law recommended: $\delta(T) = \delta_0(T_{ref}) + \delta' \cdot (T - T_{ref})$

Results – ν_{VC}





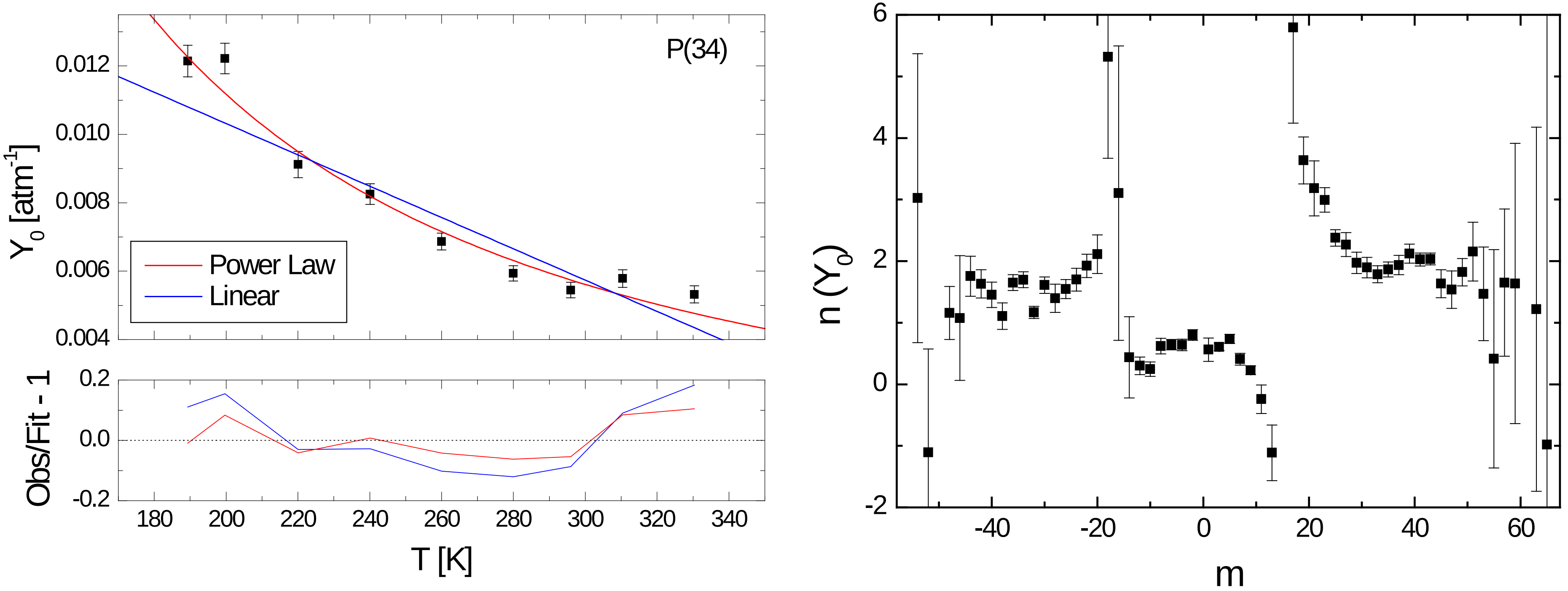
- Power law is suitable
- $n(\nu_{VC})$= 1.2 on average,
  → close to theoretical expectation $n(\nu_{VC})=1$ ($\nu_{VC} = kT / 2\pi cmD$ | $D \sim T^2$)[1]

[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 γ_0 and γ_2
- Power law suitable to describe temperature-dependence of all parameters, except δ_0

Results – Y_0



- Power law better suited than linear law

Error contributions – example: γ_2