

INVESTIGATION OF ORTHO-PARA-DEPENDENT PRESSURE BROADENING IN THE $\nu_1 + \nu_3$ BAND OF ACETYLENE

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Several years ago, Iwakuni et al.¹ reported an unexpectedly strong ortho-para-dependence to the self-pressure broadening in the $\nu_1 + \nu_3$ vibrational band of acetylene. Such an effect can arise because ortho-ortho collisions are statistically more probable than para-para ones and resonant energy transfer processes can make like-molecule collisions more efficient in state-changing, lifetime-shortening, collisions. Subsequently several papers²⁻³ have disputed the observation on the basis that the experimental sensitivity could not be sufficient, and the approximate Voigt lineshape model used in the analysis would lead to systematic errors. However, there has been no reported independent experimental work to verify the results or investigate the refutations. Our group previously reported⁴ a very accurate and precise measurement of the self-pressure broadening for the P(11) transition in the same band, using a comb-stabilized laser spectrometer. We have now resurrected this instrument, and measured additional R-branch lines to investigate the controversial results described above. Measurements of the R(12)–R(15) transitions, which showed significant ortho-para differences in the original work, have been made at pressures ranging from 200 mTorr to 150 Torr. We fit the data to both Voigt and speed dependent Voigt (SDV) lineshape model to determine the pressure broadening coefficients and investigate their rotational and nuclear-spin dependence.

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