OBSERVATION OF ANOMALOUS BROADENING AND SHIFTING OF THE RAMAN Q-BRANCH IN D_2 AND D_2-He MIXTURES

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Using high resolution Raman gain spectroscopy, we have accurately measured the Q(0), Q(1) and Q(2) lines in D_2 and D_2-He mixtures at 250 K, 200 K, 150 K, and 100.7 K. The profiles were assumed to be composed of a strong symmetric Lorentzian component and a weak line-mixing (asymmetric) component. For Q(1) and Q(2) allowance was also made for the weak symmetric depolarized Q-branch component. The widths and shifts of the strong component were extracted for all of the lines. At densities above 10 amagat where the contribution of the Dicke width is negligible, all conventional models and theories predict widths and shifts which vary proportional to the gas density. Surprisingly, above 10 amagat we observed a linear (as opposed to a proportional) variation with density. In other words we observed a constant offset as well as a term proportional to the density for both the widths and the shifts. These constant offsets are functions of temperature, mixture ratio, and rotational quantum number J. We believe these anomalous effects arise from a speed-dependence of the relaxation rates of the optical coherence.