VELOCITY-CHANGING COLLISIONS IN SUB-DOPPLER AND DOPPLER-BROADENED LINES

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The role of velocity changing collisions (VCCs) in pressure-dependent line shapes is revisited, highlighting their contributions to pressure broadening in sub-Doppler saturation line shapes and the conditions required for collisional narrowing in isolated Doppler- and pressure-broadened lines. As reported at last year's meeting (Paper WJ06, 72^{nd} ISMS), we have observed the self-broadening of sub-Doppler saturation dip absorption lines in the $v_1 + v_3$ band of acetylene near $1.5\mu\mathrm{m}$ in frequency comb-referenced measurements. The saturation line shapes are well described by Voigt functions with a fixed, narrow Gaussian component and a Lorentzian component that increases linearly with pressure up to 0.04 mbar. This sub-Doppler pressure broadening exceeds the normal pressure broadening of a full Doppler line observed at higher pressures. Velocity changes following large cross-section, elastic, collisions are dominated by a sharply spiked exponential cusp in the laboratory-frame collision kernel. The VCCs will contribute to the total broadening when the typical change in Doppler detuning associated with small angle elastic collisions exceeds the pressure-dependent homogeneous line width associated with inelastic damping. At higher pressures, the homogeneous width becomes larger than this collisional frequency shift, and the additional damping effect of VCCs becomes negligible. The pressure at which the change in slope of the line width vs. pressure will occur depends on details of the elastic collision kernel. A Monte Carlo sampling model of elastic and inelastic collision rates and cusp-like elastic collision kernels has been developed to generate electric field time correlation functions whose real Fourier transforms depict the pressure dependent line shapes. Useful physical insights follow. In order to produce collisional (Dicke) narrowing, multiple velocity changing collisions must generate large changes in the Doppler shift of a given absorbing molecule prior to its first inelastic collision.

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[b]Work at Brookhaven National Laboratory was carried out under Contract No. DE-SC0012704 with the U.S. Department of Energy, Office of Science, and supported by its Division of Chemical Sciences, Geosciences and Biosciences within the Office of Basic Energy Sciences.