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Precise lineshape parameters from Doppler-free spectroscopy at low pressures

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There have been extensive studies on the pressure dependence of line positions and widths of miscellaneous molecules using Doppler-broadened spectroscopy under moderate pressures ($10^2 - 10^5$ Pa). However, recent studies show that the pressure dependence at lower pressures could be significantly different. The main experimental difficulty in studying this phenomena is that the detection sensitivity, consequently, the signal-to-noise ratio (SNR), decreases with the sample pressure. Moreover, the collision-induced line shift and broadening is more than three orders of magnitudes smaller than the Doppler width. Using comb-locked cavity-enhanced spectroscopy techniques, such as the cavity ring-down spectroscopy, we could observe spectra with sufficiently high SNR even at very low pressures ($10^{-1} - 10^1$ Pa). Meanwhile, by investigating the Doppler-free Lamb-dip spectra which have typical widths of less than 1 MHz, we can determine the line centers and other profile parameters with high accuracy. As a demonstration, we recorded Lamb-dips of ro-vibrational transitions of carbon monoxide broadened by nitrogen near 1.56 μm at low pressures. The results show a clear difference from that reported at higher pressures, indicating that more physics could be explored in this direction.