DIRECT MEASUREMENTS OF COLLISIONALLY BROADENED (CO_2-CO_2) S-BRANCH RAMAN COHERENCE LIFETIMES OF CO_2

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We report direct measurement of S-branch Raman coherence lifetimes of CO_2 due to CO_2-CO_2 collisions by employing picosecond time-resolved coherent anti-Stokes Raman scattering (CARS) spectroscopy. A custom-built, high-peak-power, nearly transform-limited ps laser system offers an ideal combination of frequency and temporal resolution for such measurements. The rotational S-branch transitions of CO_2 ground state [0,0^0,0] with rotational quantum number J= $0 - 52$ were simultaneously excited by using a broadband ($\sim$3-nm) laser pulse with a full-width-half-maximum (FWHM) of $\sim$100 ps. The coherence lifetimes of self-broadened CO_2 for a pressure range of 0.05-1.5 bar were directly measured by probing the rotational coherence with a nearly transform-limited, 80-ps-long laser pulse. The measured linewidth of J=6 and J=50 transitions are found to be $\sim$0.106$\pm$0.0002 and $\sim$0.070$\pm$0.0002, respectively. As expected, the energy-transfer from high J levels has a significantly longer coherence lifetime because of the inertia associated with higher angular momentum. These measurements are very significant for performing accurate thermometry or CO_2 concentration measurements in gas-phase reacting flows.