J-SPECIFIC DYNAMICS IN AN OPTICAL CENTRIFUGE USING TRANSIENT IR SPECTROSCOPY

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Quantum state-specific dynamics are reported for a number of CO_2 rotational states in an optical centrifuge. The optical centrifuge results from combining oppositely-chirped ultrafast laser pulses and spinning CO_2 molecules into extremely high rotational states with J≈220. Collisions of centrifuged molecules induce depletion of population from low-J states (J=0 and 36) and lead to appearance of population in high J states (J=36, 54 and 76). Transient Doppler-broadened line profiles for individual CO_2 states reveal that the depletion populations have narrow velocity distributions with translational temperatures significantly colder than 300 K. Molecules that appear in the higher rotational states have broad velocity distributions, showing that both rotational and translational energy are imparted in collisions of the centrifuged molecules. These results show that substantial amounts of angular momentum persist after many collisions and that translational energy exchange continues for several thousand collisions.