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Spinning CO_2 Molecules into High Rotational States with an Optical Centrifuge AMY S. MULLIN, LIWEI YUAN, SAM TEITELBAUM, ALLISON ROBINSON, Department of Chemistry and Biochemistry, University of Maryland — We have performed the first spectroscopic measurements of molecules in an optical centrifuge. The optical centrifuge is a means to generate molecules in very high rotational states using a pulsed laser. The optical centrifuge consists of two ultrafast laser pulses with reverse chirp and circular polarization that are combined to generate an intense electric field that undergoes angular acceleration. Molecules with polarizability anisotropy are driven by the field into high rotational states. We have used an optical centrifuge to promote CO_2 molecules into high rotational states (J$\sim$200) and monitored the effect of the centrifuge on different quantum states using high-resolution transient IR diode laser absorption at λ=4.3 μm. The depletion of low angular momentum (J=14) states and the appearance (and subsequent depletion) of middle-J (J=76) states that are populated by a collisional cascade have been observed and characterized. Direct detection of CO_2 molecules in states near J=200 will allow further characterization of the centrifuge. Transient signals were observed only in the presence of both optical centrifuge pulses and for pulses with circular polarization. The ability to control molecular rotation using the optical centrifuge opens a new realm of investigation into the behavior of energized molecules.

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