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Spinning CO_2 Molecules into High Rotational States with an **Optical Centrifuge**¹ AMY MULLIN, LIWEI YUAN, ALLISON ROBINSON, SAMUEL TEITELBAUM — We report the first spectroscopic measurements of molecules in an optical centrifuge which generates molecules in very high rotational states. The optical centrifuge combines two ultrafast laser pulses having reverse chirp and circular polarization to generate an intense electric field that undergoes angular acceleration. Molecules with non-uniform polarizability are driven by the field into high rotational states. We have used the optical centrifuge to promote CO_2 molecules into high rotational states (J~200) and monitored the effect of the centrifuge on different quantum states using high-resolution transient mid-IR laser absorption. We have observed and characterized the depletion of low angular momentum (J=14) states and the appearance (and subsequent depletion) of middle-J (J=76) states populated by a collisional cascade. Direct detection of CO₂ molecules in states near J=200 will allow further characterization of the centrifuge. Transient signals are 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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