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Coherently controlled orientation and alignment of molecules in the gas phase by intense THz fields SHARLY FLEISCHER, YAN ZHOU, ROBERT W. FIELD, KEITH A. NELSON, Department of Chemistry, Massachusetts Institute of Technology — We report the use of intense terahertz (THz) pulses to orient polar molecules in the gas phase. Short THz fields exert torques that drive coherent molecular rotational motion, and multiple interactions with strong THz fields can yield multiple-quantum rotational coherences with the prospect of high degrees of orientation (dipoles pointing in the same direction in space) and alignment (molecular axes parallel to each other regardless of dipole orientation). THz-induced molecular orientation offers new possibilities in gas-phase x-ray diffraction, molecular orbital mapping through high harmonic generation and photoelectron angular distribution imaging, and other applications. We demonstrate significantly enhanced coherent control using two THz pulses with an optimized relative time delay. We show in the case of atmospheric water that a short, strong THz field induces long-lived coherent THz emission (free induction decay) that drives significant further rotational responses in a pre-excited polar gas sample. This class of experiments enables broad new capabilities for molecular spectroscopy and control beyond those afforded through molecular alignment by intense optical fields, which do not produce any net orientation.

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