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Controlling rotational revivals in asymmetric tops[1] VINOD KUMARAPPAN, LOTTE HOLMEGAARD, SIMON VIFTRUP, CHRISTER BISGAARD, HENRIK STAPELFELDT, University of Aarhus, EDWARD HAMILTON, TAMAR SEIDEMAN, Northwestern University — We use improved experimental and theoretical tools to demonstrate a novel method for controlling the revival structure of strong-field alignment of asymmetric top molecules. Experimentally, iodobenzene molecules (which is a near-prolate top) are cooled to 1 K using an Even-Lavie supersonic valve and non-adiabatically aligned using 800 nm pulses of durations ranging from 200 fs to 2 ps. The alignment is probed by velocity map imaging of I^+ fragments produced by Coulomb explosion of the molecules using a 25 fs pulse focused tightly to restrict the volume probed. We show that as the fluence of aligning pulse is increased, the revival structure is simplified to a nearly period pattern reminiscent of symmetric tops. Theoretically, non-perturbative solution of the Schrodinger equation demonstrates the generality of the effect, and emphasizes the importance of this new control scheme for the alignment and revival dynamics of asymmetric tops. Classically, the simplified motion at high fluence corresponds to stable rotations about the slowest principal axis (the C-axis) of the molecule.

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