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Optimizing dynamic alignment in diatomic molecules by pulse shaping DANIEL PINKHAM, KAREN MOONEY, ROBERT JONES, University of Virginia — We have used intense laser pulses in combination with a genetic algorithm and laser pulse shaper to explore transient laser alignment of room temperature CO. A molecule with an anisotropic polarizability may undergo periodic, transient alignment following its exposure to an intense ultra-short laser pulse. The laser populates a coherent superposition of rotational states via a sequence of Raman transitions. As this rotational wavepacket evolves, the angular probability distribution periodically aligns parallel and perpendicular to the linear laser polarization. In the impulsive limit, the laser bandwidth exceeds the energy separation between rotational levels, and the degree of alignment depends only on the time-integrated intensity in the pulse. We have used shaped laser pulses in an attempt to optimize the degree of alignment at fixed pulse energy and explore transient laser alignment outside the impulsive limit. In particular, we use phase-only shaping and, using a 30 fsec Coulomb explosion pulse, probe molecular alignment at a delay corresponding to one of the alignment "revivals." By inspecting the time-of-flight of the exploded fragments, we obtain rapid experimental feedback which enables us to test the effectiveness of hundreds of different pulse shapes. In the experiment and corresponding theoretical simulations, we find complex, non-impulsive shapes which align the molecules at least as well as a transform-limited pulse. This work has been supported by DOE BES.

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