Alignment Echoes in Unidirectionally Rotating Molecules

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Echoes are a fundamental element in modern nuclear magnetic resonance and magnetic resonance imaging, have been observed in various non-linear systems. In recent years, laser-induced molecular alignment and orientation echoes have been discovered and extensively studied in molecular gases. Short laser pulses effectively kick the molecules, which then come to alignment along with the field polarization. With time, molecules having different angular velocities step out of phase, and the alignment disappears. If the molecules are kicked again by a second delayed laser pulse, a spontaneous alignment emerges, the so-called alignment echo at twice the delay between the exciting pulses.

In this work, we report the observation of molecular unidirectional rotation (UDR) echo induced by a pair of delayed co-rotating or counter-rotating polarization-twisted pulses. We show that during the echoes, the molecules rotate with a preferred sense. In our experiments, the molecular dynamics is followed using Coulomb explosion through the method of cold target recoil ion momentum spectroscopy (COLTRIMS). The molecular UDR echoes can also be studied in gas cells using all-optical detection. This new type of echo may provide complementary information on the relaxation processes in dense molecular gases, additional to the one offered by the standard rotational echoes generated by parallel linearly polarized pulses.