Ultrafast engineering of the magnetic Hamiltonian in frustrated magnets

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Ultrafast light-matter coupling has emerged as a new mechanism to stabilize novel phases of matter that are not stable in thermal equilibrium [1]. Of particular interest are quantum materials with coupled magnetic and structural instabilities. In these compounds, the targeted manipulation of the crystal lattice via strong THz pulses resonant with IR active phonon modes can decouple the magnetic and structural degrees of freedom and alter the delicate energy balance between them [2]. Observation of this phenomenon in antiferromagnets, which should display faster dynamics and are of interest to spintronic applications [3], is yet to be demonstrated. We deploy ultrafast laser pulses to engineer the magnetic groundstate of an iridium-based frustrated quantum magnet [4] by resonantly pumping an IR active phonon mode that couples to a symmetry-breaking Jahn-Teller mode, we engineer long-lasting changes to the magnetic Hamiltonian as demonstrated by our THz spectroscopy measurements. Our results open new pathways to controlling complex magnetic orders in strong spin-orbit coupled frustrated spin degrees of freedom [5].

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