

Slow dynamics from ultrafast coherent phononic driving in CuGeO₃

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Advances in ultrafast light sources allow the targeted modulation of magnetic interactions through selective excitation of infrared-active phonons [1–4], a mechanism known as magnetophononics. The opportunity to probe dynamical magnetic properties out of equilibrium opens new possibilities for finding both novel phenomena and novel solutions to old but unresolved problems. An example of the latter is the spin-Peierls transition in CuGeO₃. This material hosts quasi-1D antiferromagnetic $S = \frac{1}{2}$ chains, which are unstable towards a lattice distortion that promotes the formation of a gapped, non-magnetic, collective singlet ground state of spin dimers below a transition temperature of $T_{SP} = 14$ K [5]. Despite intensive research into the magnetoelastic coupling mechanism [6–11], neither a soft mode nor the precise phonon or phonons driving the dimerization of the 1D electronic system has ever been identified. We apply intense THz pulses to pump a number of phonon modes that modulate the superexchange paths between neighbouring Cu atoms. We identify extremely slow dynamics triggered by the light pulses when measuring on resonance with the onsite Cu d-d transitions. These dynamics are unique to the spin-Peierls phase and represent the first observation of a distinct difference between the uniform and the spin-Peierls phase in a pump-probe experiment. We investigate the origin of this slow timescale (150 ps) within the physics of the spin-Peierls transition and use the example of CuGeO₃ to discuss the new possibilities that coherent phononic control of magnetic interactions by THz pulses can offer to the field of quantum magnetism.

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