

Band-resolved, phonon-mediated ultrafast demagnetization in MnBi₂Te₄

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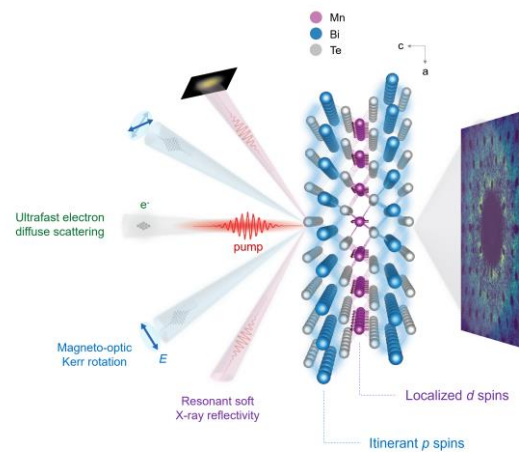


Fig. Multimodal probe of ultrafast demagnetization. Schematic illustrating the time-resolved experimental probes used to characterize ultrafast spin and lattice dynamics in MnBi₂Te₄. The lattice structure of MnBi₂Te₄ is shown with the itinerant *p* spins illustrated schematically using blue clouds, and localized *d* spins illustrated using arrows on Mn ions.

Magnetism in functional quantum materials provides a platform for the discovery of cooperative phenomena with applications in spintronics, magnetic memory, and quantum information. A comprehensive understanding of spins away from equilibrium, including their interplay with lattice and electronic excitations is key to achieving fundamental breakthroughs in this context. We study ultrafast melting of magnetic order in the layered antiferromagnetic topological insulator MnBi₂Te₄. We employ a multimodal experimental approach, wherein ultrafast electron diffuse scattering is used as a momentum-resolved probe of nonequilibrium phonon dynamics, and magneto-optic Kerr rotation and resonant soft X-ray reflectivity are used to directly measure ultrafast dynamics of conduction and localized spins, respectively. Our experiments reveal that ultrafast demagnetization occurs through a *p*-like itinerant spin subsystem, with optical phonons acting as the primary channel for the dissipation of spin angular momentum. Localized Mn *3d*⁵ spins disorder by a distinct, much slower thermalization mechanism, due to the quenched orbital angular momentum. The simultaneous appearance of the disparate demagnetization timescales in both spin subsystems provides evidence of a strong exchange coupling between *3d* localized spins and the *p*-like itinerant bands. Given the *p*-like nature of the topological surface states, our work not only provides a unified picture of nonequilibrium spin dynamics in MnBi₂Te₄, but also sheds light on the interatomic exchange pathways that enable magnetic topological phases.