Interlayer Magnetophononic Coupling in $MnBi_2Te_4$

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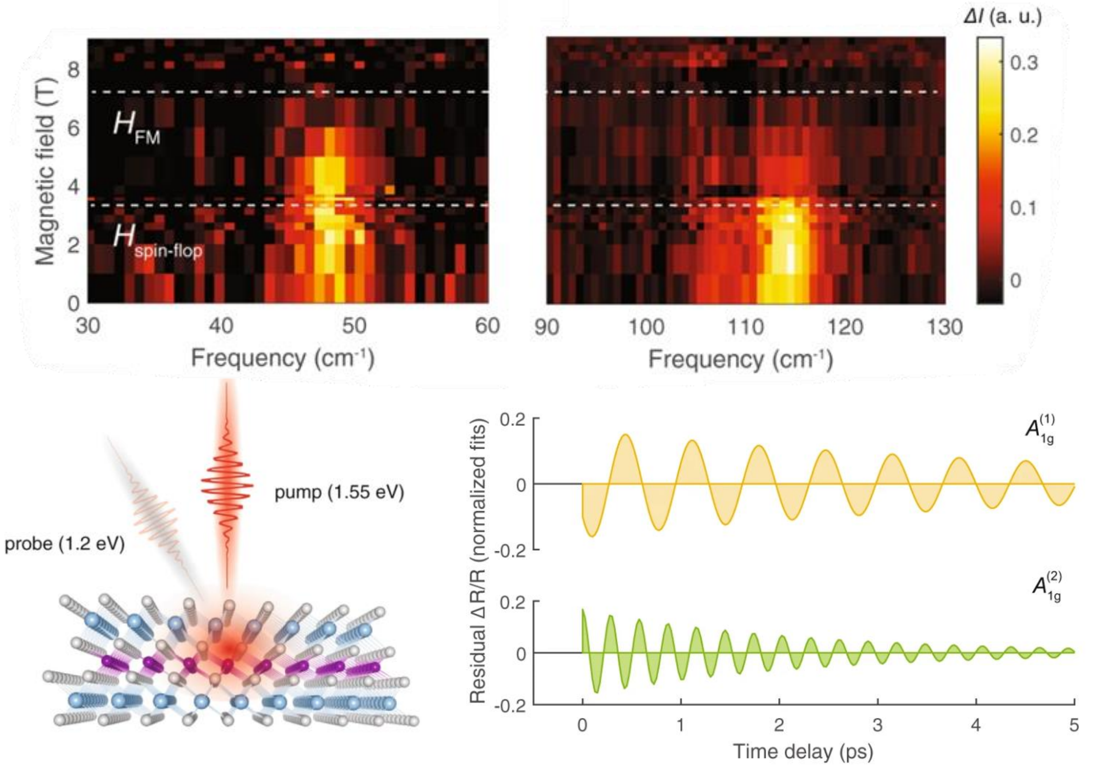
Project Summary: Magnetism in quantum materials creates a fertile ground for the exploration of fundamental spin-based phenomena, with applications in spintronics, magnetic memory, and quantum information technology. This is exemplified by the recent development of magnetic topological materials. As with other functional materials, the identification of accessible coupling pathways to the foremost degree of freedom is key to enabling new fundamental and technological breakthroughs. In $MnBi_2Te_4$, a layered intrinsic antiferromagnetic topological insulator, we found sub-picosecond coherent magnetophononic coupling. Specifically, light-induced displacive excitation of phonons coherently modulates magnetism at terahertz frequencies, orders of magnitude faster than traditional techniques such as strain or magnetic fields. Moreover, we also used Raman spectroscopy to reveal magnetic control derives from strong optical phonon exchange coupling. These results are buttressed with insights obtained from MeV ultrafast electron diffraction experiments. Interestingly, we found that the experimentally observed magnetic response speed belies expectations based on an equilibrium description of the exchange interaction. Thus, we demonstrated the first step towards achieving ultrafast control of magnetic topological phases and challenged the prevailing ideas of nonequilibrium magnetism at femtosecond timescales. The detailed findings are published in *Nature Communications* **13**, 1929 (2022).

2DCC Role: This research resulted from a close collaboration between 2DCC and a local user, Prof. Venkatraman Gopalan at Penn State. The high-quality single crystals of $MnBi_2Te_4$ used for this study were grown using a self-flux method at the 2DCC Bulk Growth facility.



Contour plots of the difference of Raman spectra upon subtracting the 9 T spectrum, as a function of magnetic field. The dotted lines denote the ferromagnetic and spin-flop critical fields (top panels). Schematic of pump-probe measurement (bottom left). Individual decaying sinusoidal components corresponding to the $A_{1g}^{(1)}$ and $A_{1g}^{(2)}$ phonons, respectively (bottom right).