Distinguishing surface and bulk electromagnetism via their dynamics in an intrinsic magnetic topological insulator*

Khanh Duy Nguyen[1], Woojoo Lee[1], Jianchen Dang[2], Tongyao Wu[2], Gabriele Berruto[1], Chenhui Yan[1], Chi Ian Jess Ip[1], Haoran Lin[1], Qiang Gao[1], Seng Huat Lee[3], Binghai Yan[4], Chaoxing Liu[3], Zhiqiang Mao[3], Xiao-Xiao Zhang[2], Shuolong Yang[1]

[1]Pritzker School of Molecular Engineering, the University of Chicago, Chicago, IL 60637, USA. [2]Department of Physics, University of Florida, Gainesville, FL 32611, USA. [3]Department of Physics, Pennsylvania State University, University Park, PA 16802, USA. [4]Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 7610001, Israel.

Bringing magnetism to the itinerant electronic states on the surface of three-dimensional (3D) topological insulators (TIs) is foundational to a variety of low-dimensional topological orders [1, 2]. The magnetism in 3D TIs can be established via various mechanisms. However, the unconventional Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction on the material surface is required for the time-reversal (T) symmetry breaking on the topological surface states (TSSs) in magnetic TIs (MTIs)[3]. This mechanism has been predicted to enhance the surface magnetism of 3D MTIs, where the itinerant Dirac fermions with vanishing Fermi momenta strongly favor ferromagnetic coupling [4]. Thus, the 2D RKKY interaction fundamentally determines the size of the T-symmetry–broken energy gap, and, consequently, the operating temperatures of low-dimensional topological orders, such as the quantum anomalous Hall effect.

Here, we combine time- and angle-resolved photoemission spectroscopy (trARPES) and time-resolved magneto-optical Kerr effect (trMOKE) to reveal this distinct mechanism contributing to the surface magnetism in $MnBi_2Te_4$ (MBT): A quasi-2D state mediates the surface 2D RKKY interaction via *p-d* coupling on the top MBT layer, Fig. 1(a). While trARPES resolves the dynamics of the exchange gap in the *q*-2DS with meV-scale precisions, Fig. 1(b), trMOKE observes the evolution of the magnetization. We construct a 2D RKKY model involving localized Mn 3*d* moments and itinerant *p* electrons, which accounts for the rapid dynamics of the magnetization and the exchange gap. Furthermore, it can reconcile several open problems in MBT. These include the vanishing gap at the Dirac point of the TSSs [5] and the nonzero residual magnetization in even-layer MBT flakes [6]. Our work highlights the special magnetic interactions on the surface of MBT and establishes the physics foundation for effective ultrafast manipulation of magnetism in tandem with topological orders.

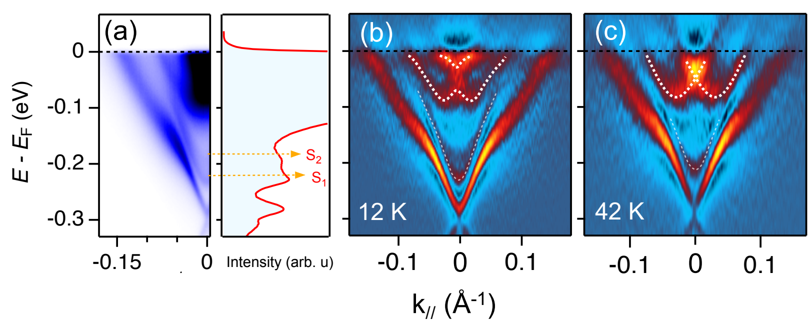

Figure 1: ARPES and trARPES spectra of $MnBi_2Te_4$. (a) Static ARPES spectrum shows a quasi-2D state splitting into S_1 and S_2 at 12 K. (b, c) trARPES taking at 360 fs at the base temperature of 12 K and 42 K respectively, under 1.5-eV pump fluence of 10 μJ/cm^2.

[1] C.-Z. Chang et al., *Science* **340**, 167170 (2013)
[2] C. Liu et al., *Nat. Mater.* **19**, 522–527 (2020)
[3] Q. Liu et al., *Phys. Rev. Lett.* **102**, 156603 (2009)
[4] J. Wang, B. Lian, S.-C. Zhang, *Phys. Scr.* **T164**, 014003 (2015)
[5] C. Yan et al., *Phys. Rev. B* **104**, l041102 (2021)
[6] S. Yang et al., *Phys. Rev. X* **11**, 011003 (2021)

*Publication reference: Nguyen *et al*., Sci. Adv. **10**, eadn5696 (2024)

Supplementary information

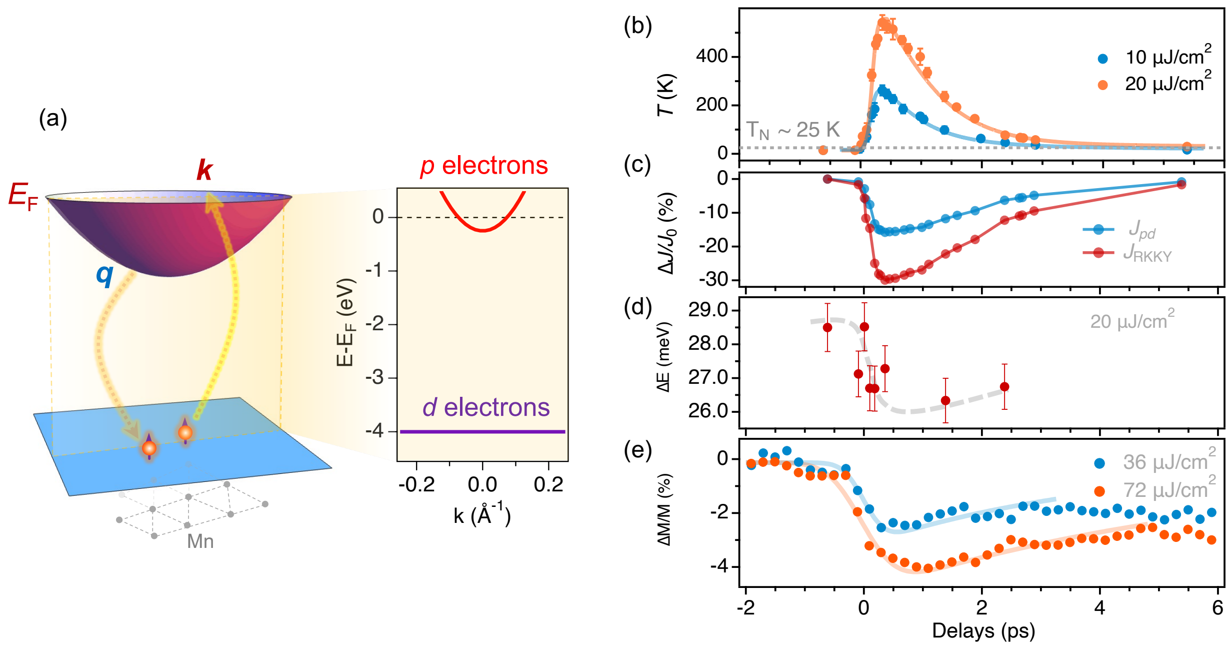

Figure 2. Magnetic interactions in a 2D system with itinerant *p* electrons and localized Mn *d* electrons. (a) Illustration of an RKKY model that assumes the indirect interaction between localized *d* electrons via itinerant *p* electrons in two dimensions. (b) Transient electronic temperatures extracted by fitting the momentum-integrated EDC at each delay to a modified Fermi-Dirac (FD) function. Error bars show SD of the fitting. Solid curves denote simulation results using a microscopic Boltzmann model. The resulting *p*-*d* and RKKY coupling strengths are calculated and summarized in (c) based on the transient electronic temperature using a pump fluence of 20 μJ/cm^2. (d) Time-dependent exchange gap in the *q*-2DS also using the pump fluence of 20 μJ/cm^2. (e) Photoinduced demagnetization measured by trMOKE using a pump fluence of 36 and 72 μJ/cm^2. Solid lines indicate the fitting curves using an exponential decay convolved with a Gaussian function.