Layer-by-layer engineering and deciphering of topological orders in magnetic topological insulators

Woojoo Lee,¹ Sebastian Fernandez-Mulligan,¹ Hengxin Tan,² Chenhui Yan,¹ Yingdong Guan,³ Seng Huat Lee,^{3,4} Ruobing Mei,³ Chaoxing Liu,³ Binghai Yan,² Zhiqiang Mao,³ and <u>Shuolong Yang</u>,^{1,+}

¹ Pritzker School of Molecular Engineering, The University of Chicago, Chicago, Illinois 60637, USA

² Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 7610001, Israel

³ Department of Physics, Pennsylvania State University, University Park, State College, Pennsylvania 16802, USA

⁴ 2D Crystal Consortium, Materials Research Institute, Pennsylvania State University, University Park, State College, Pennsylvania 16802, USA

The advent of intrinsic magnetic topological insulators enables us to envisage various lowdimensional topological orders, such as the quantum anomalous Hall insulators and the axion insulators, at realistic cryogenic temperatures. These materials are represented by MnBi₂Te₄ and its derived superlattices $MnBi_{2n}Te_{3n+1}$. However, it has been controversial whether these materials exhibit the key ingredient for magnetic topological phases: an energy gap due to the time-reversal symmetry breaking. Moreover, the construction of high-quality magnetic topological insulators at the ultrathin limit has met significant challenges. In this talk, I will present a new technique, layer-encoded frequency-domain photoemission spectroscopy, which allows us to decipher the layer origins of various electronic states. By encoding layer indices with intralayer phonon frequencies, we measure the strengths of coupling with layerspecific phonons. This experiment reveals that the topological surface states on antiferromagnetic MnBi₄Te₇ are partially relocated to the nonmagnetic layers, reconciling the mystery of vanishing broken-symmetry gaps [1]. Moreover, I will present our recent progress on the "carpet-growth" of Bi2Te3 ultrathin films and MnBi2Te4/Bi2Te3 heterostructures using molecular beam epitaxy. These thin films extend coherently across a millimeter spatial scale without disruptions by substrate step edges. Angle-resolved photoemission spectroscopy studies yield unprecedentedly sharp electronic structures in agreement with first-principles calculations layer-by-layer, and suggest opportunities to realize the quantum spin Hall effect and quantum anomalous Hall effect at near-ambient temperatures [2].

[1] W. Lee *et al.*, Nature Physics **19**, 950-955 (2023).

[2] W. Lee *et al.*, Submitted (2023).

⁺ Author for correspondence: yangsl@uchicago.edu

Supplementary Page 1

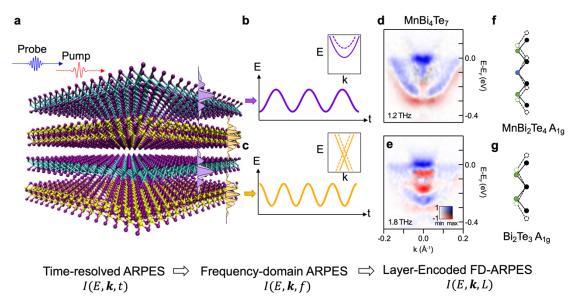


Figure 1. Resolving the layer origins of electronic states in MnBi₄Te₇ using layer-encoded, frequency-domain ARPES. (a) Schematic of pump-probe setup on a superlattice material. (b,c) Different electronic states are coupled to layer-specific coherent phonon oscillations. (d,e) Frequency-domain (FD) ARPES allows us to resolve the electronic states coupled to the MnBi₂Te₄ A_{1g} mode and the Bi₂Te₃ A_{1g} mode, respectively. The layer-frequency correspondence leads to layer-encoded FD-ARPES. The mode configurations are shown in panels (f) and (g).