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Report on the thesis

Electronic, spin structure and magnetic properties of intrinsic magnetic and magnetically doped topological insulators

by Dmitriy A. Estyunin (St. Petersburg State University)

Magnetic topological insulators (MTIs), combining the topological protection of electronic states with an intrinsic magnetic field, are presently one of the most active research fields. At low temperatures of a few tenths of K, these materials can transit into a quantum anomalous Hall (QAH) state where the topological protection of the edge states allows lossless spin-polarised current. This phenomenon might allow realisation of novel quantum devices for a wide range of applications, including power-saving electronics, spintronics and quantum computing. A deep fundamental understanding of the electronic and magnetic properties of the MTIs is required, however, before practical devices based on the QAH could be developed. In this regard, the thesis of D. A. Estyunin, devoted to investigation of MTIs with a handful of electron spectroscopy and magnetometric methods, is highly actual. The thesis focuses on two paradigm MTIs, where the internal magnetic field in the host topological insulator is created either by incorporation of magnetic impurities (Gd-doped Bi_2Se_3) or by insertion of an ordered magnetic layer (the $MnBi_2Te_4(Bi_2Te_3)_m$ family).

Chapter 1 presents a survey of the literature in the electronic and magnetic structure of MTIs. The survey is well organised, comprehensive, and a pleasure to read. I would only bring the author's attention to our work on V-doped Bi_2Se_3 [Krieger *et al*, Phys. Rev. B **99** (2019) 064423] where we used resonant photoemission (ResPE) to identify a significant concentration of V-derived defect states in the region of the Dirac point, which compete with the QAH state. From the muon spin spectroscopy, we explain the QAH effect in this system by embedding of pristine Bi_2Se_3 domains between highly-doped magnetic ones.

Chapter 2 describes the physical principles and instrumental realisation of the employed spectroscopic methods, including angle-resolved photoelectron spectroscopy (ARPES) and its spin-resolved version (SARPES), ResPE, and X-ray magnetic circular dichroism (XMCD). Here, one of my remarks is that the efficiency of the spin detectors is actually measured not merely with the Sherman function *S*, but rather with the so-called figure of merit (FOM) which combines it with

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the intensity loss as FOM = S^2 (I/I_0). Furthermore, I have noticed that whereas the ResPE is used as the elemental-specific probe throughout the work, the author explains the principles of resonant Auger spectroscopy. I wished that the author had highlighted the fundamental difference between the coherent (**k**-conserving) ResPE process and incoherent resonant Auger one, as well as their different dependence on the excitation energy (constant binding vs kinetic energy). The participator vs spectator schemes of the resonant Auger processes might have also been explained. The chapter also reviews the X-ray diffraction (XRD) and atomic-force microscopy (AFM) as the structural characterization methods, and the Superconducting Quantum Interference Device (SQUID) used for the magnetometric characterization, as well as the sample growth methods. The results obtained with these methods for the actual Gd-doped Bi_2Se_3 and $MnBi_2Te_4(Bi_2Te_3)_m$ samples are presented.

Chapter 3 unfolds the experimental band structure, spin texture and macroscopic magnetic properties of Bi_2Se_3 doped with magnetic Gd atoms. Additional doping with Sb shifts the Dirac point to the Fermi level, which is prerequisite for the QAH effect. Significant Gd 4*f* to Se 3*p* hybridization has been identified with ResPE. The sought-for band gap has indeed been observed in the Dirac point. However, the magnitude ~30 meV of this gap is almost independent of temperature, suggesting its chemical rather than magnetic origin. These results are extremely convincing. I would only add that the Gd *M*-edge resonant enhancement of the spectral intensity in the vicinity of the Dirac point is restricted to the dispersive Se 3*p* band without any traces of Gd impurity states which might compete with the QAH state. Though I have a question regarding the finite acceptance of the analyzer in the K_y direction (perpendicular to the analyzer slit). In view of the sharp band dispersion near the Dirac point, does it affect the obtained band gap values?

Chapters 4 and 5 (which are logically connected and, in my humble opinion, might have been joined) report the temperature-dependent spectroscopic and magnetometric studies to $MnBi_2Te_4$ the intrinsic MTI where the internal magnetic field is formed by incorporation of Mn layers into the Bi_2Se_3 crystal lattice. The crystallinity of the Mn sublattice, in contrast to the random Gd impurities, ensures the absence of defect states (at least theoretically) and the regularity of the internal magnetic field. A band gap of ~70 meV has been found in the Dirac point, reducing to ~40 meV above the Neel temperature (T_N). This fact strongly suggests a magnetic contribution to the gap, likely connected to the QAH state, on top of a temperature-independent chemical contribution. Interestingly, splitting of the conduction-band minimum has been found below T_N though its connection to the magnetic effects is yet unclear. All these results are interesting and convincing. My only remark here is that the calculations in Fig. 4.1f are actually the surface-projected band structure (including the topological and trivial surface states) rather than the bulk band structure. Finally, I was intrigued about the origin of the spectral intensity pile-up below T_N. It reminded me of the BCS theory of conventional superconductivity, where the spectral weight from the opening superconducting gap distributes to the surrounding spectral peaks. Could there be any analogy between the antiferromagnetic and superconducting transitions in terms of the spectral weight redistribution?

The experimental material in Chapter 6 is devoted to verification of the obtained results on $MnBi_2Te_4$ over different batches of samples grown in different labs. I deem such systematic studies, albeit time consuming, extremely important for drawing reliable conclusions on the physics of the studied systems.

Chapter 7 extends the studies to the whole $MnBi_2Te_4(Bi_2Te_3)_m$ family where the magnetic septuple $MnBi_2Te_4$ layers are sandwiched between the non-magnetic quintuple Bi_2Te_3 ones. The variation of m modulates the electronic coupling between the $MnBi_2Te_4$ layers and thus the electronic and magnetic properties of the system. Upon cleavage, the surfaces of $MnBi_2Te_4(Bi_2Te_3)_m$ are a statistical mixture of the $MnBi_2Te_4$ and Bi_2Te_3 terminations, whose electronic structure probed by ARPES appears different, with a weak dependence on m. Such studies require a small footprint of the incident light on the sample, in the present study about ~5 µm delivered from a laser source. I have no particular remarks on this chapter.

Chapter 8 is devoted to the effects of irradiation during the ARPES experiments. It is shown that adsorption of residual gases during exposure of cleaved surfaces in ultra-high vacuum ($\sim10^{-10}$ Torr) results, even for relatively short exposures (< 1 hour), in time-dependent energy shifting and broadening of the ARPES structures. Measurements on freshly cleaved surfaces ensure minimal irradiation effects though. Though these studies are extremely important for obtaining reliable experimental data, they are largely known and, in my humble opinion, might have been placed in an appendix to the thesis.

Finally, two general remarks: (1) The figures presented in the thesis often use different and sometimes opposite colorscales (for example, Figs. 3.6 and 3.7). It would have been helpful to show colorbars at all figures; (2) I wished the author had added a few detail of the DFT calculations used through the whole thesis. In particular, which exactly exchange-correlation potential has been used?

Overall, I am impressed by the amount and depth of the new scientific knowledge on the physics of MTIs achieved in this thesis work. My critique remarks can no way affect my overall assessment of the thesis as an excellent piece of scientific research. The experimental results are novel, extensive and reliable, and the interpretations are convincing. The main results of the thesis are reported in 7 publications in refereed journals, including 2 first-author publications. The thesis stands as a significant development in the field of condensed-matter physics, and Dmitriy A. Estyunin the author undoubtedly deserves awarding him the degree of candidate of physico-mathematical sciences on the scientific specialisation 1.3.8 (condensed matter physics).

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