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It is certified that the work contained in the thesis titled ***"Electronic and Magnetic Properties of Some Topological materials"*** by **Debarati Pal** has been carried out under my supervision and that this work has not been submitted elsewhere for a degree.

It is further certified that the student has fulfilled all the requirements of comprehensive examination, candidacy and SOTA for the award of Ph.D. Degree.

**Signature:**

**Supervisor**
**Dr. Swapnil Patil**
**(Assistant Professor)**
**Department of Physics**
**Indian Institute of Technology**
**(Banaras Hindu University)**
**Varanasi-221005 (UP)**

Assistant Professor
Department of Physics
Indian Institute of Technology
(Banaras Hindu University)
Varanasi-221005

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I, ***"Debarati Pal"***, certify that the work embodied in this thesis is my own bonafide work and carried out by me under the supervision of **"*Dr. Swapnil Patil"*** from **"*July 2017*"** to **"*July 2022*"**, at the **"*Department of Physics*"**, Indian Institute of Technology (BHU), Varanasi. The matter embodied in this thesis has not been submitted for the award of any other degree/diploma. I declare that I have faithfully acknowledged and given credits to the research workers wherever their works have been cited in my work in this thesis. I further declare that I have not willfully copied any other's work, paragraphs, text, data, results, *etc*., reported in journals, books, magazines, reports dissertations, theses, *etc*., or available at websites and have not included them in this thesis and have not cited as my own work.

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**Place:** IIT (BHU), Varanasi

Debarati Pal

**Signature of the student**

(*Debarati Pal*)

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It is certified that the above statement made by the student is correct to the best of my/our knowledge.

**Supervisor**
**Dr. Swapnil Patil**
**(Assistant Professor)**

Assistant Professor
Department of Physics
Indian Institute of Technology
(Banaras Hindu University)
Varanasi-221005

**Signature of Head of Department**

HEAD/विभागाध्यक्ष
भौतिकी विभाग/Deptt. of Physics
भा०प्रौ०सं०/(का०हि०वि०)/IIT (BHU)
वाराणसी/Varanasi-221005

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(Debarati Pal)

*I dedicate this thesis to my beloved parents*

# Contents

# List of Figures

# List of Tables

In this thesis, we have investigated the topological nature, Fermi surface properties, magnetic behaviour, electronic transitions, structural transitions, and electronic transport properties of several doped Topological compounds like Cu doped $Sb_2Te_3$, Fe and S co-doped $Bi_2Te_3$, Fe and S co-doped $Sb_2Te_3$, Dy doped $BiSbTe_3$ and P doped $MoTe_2$ under different conditions. These materials provide an ideal playground to tune the lattice dynamics, Fermiology, spin dynamics and create exceptional quantum phenomena due to the effect of dopant and its interaction with the host element. Each of these systems we studied, represent some exotic and new features with rich physics, thus make them extraordinary from the parent compound. The precise choice of dopant allows us to nurture the physics behind, and fundamentally interesting from Physics and Material science point of view. The motivation behind each work has been elaborated in the respective chapters.

In **Chapter 1**, we have revisited the theoretical and experimental discoveries that led to our current understanding of topological band theory. These breakthroughs spanned decades, and each of them proved to be a significant step in quantum condensed matter physics. We begin with the concept of quantum Hall state which is the onset of the historical developments of Topological Insulators (TIs). The integer quantum Hall effect (IQHE) is then observed, followed by the establishment of Quantum spin Hall insulator (QSHI) and topological order in real systems. The physics of TIs and Topological semimetals (TSMs), specifically Weyl Semimetals (WSMs) have been articulated in detail. Following that, in chronological sequence, we presented the fascinating properties of TIs and the effect of magnetism, pressure and temperature on the Topological property. The basics of various important quantum features that we observed in studying the materials are also explained to understand the physics behind them. This comprises the effect of magnetism- anomalous Hall effect, the intrinsic and extrinsic mechanism behind AHE, spin chirality induced skew scattering beyond the usual scattering effect, magnetic frustration including spin glass or

cluster glass and also the effect of high pressure like electronic topological transition (ETT), metal-semiconductor transition, structural transitions when we put different dopants into the TI or TSMs system. This chapter also deals with other issues like the concept of Berry phase, Shubnikov-de Haas (SdH), de Haas-van Alphen (dHvA) oscillation, weak antilocalization (WAL)-weak localization (WL) effect, the protection of Topological surface state etc. Finally, we highlighted the important findings of this thesis and emphasised how these works prompted us to investigate such unique quantum phases of matter.

In **Chapter 2** we discuss the synthesis and characterization techniques that are used throughout this thesis. This includes how the single crystal Topological insulators and Weyl semimetals are grown using advanced technology. Some specific choices that made in order to play with the electronic structures of those materials are also mentioned. The characterization specifications for operation are also discussed. The structural analysis was performed with single crystal and powder XRD ($CuK_\alpha$ $\lambda$~1.54Å), the structural transition or changes in electronic topology of the Fermi surface was captured with synchrotron angle dispersive X-Ray diffraction ADXRD ($\lambda$~0.7842Å), pressure dependent Raman (Diode pumped solid state laser ~532 nm), temperature dependent Raman (He-Ne laser ~633 nm). The electronic behaviour was analysed by transport studies including transverse and longitudinal resistivities and performed with the help of physical property measurement system (PPMS). To shed light on some particular phenomena, magnetic behaviour measured with magnetic properties measurement system (MPMS). To delve into the materials' complex band topology and relate that with Fermi surface, dHvAa, SdH and angle resolved photo emission spectroscopy (ARPES) are executed carefully.

In **Chapter 3** the electronic properties of a near-perfect compensated semimetal MoTeP were investigated using longitudinal and transverse resistivity (Hall) measurements. A

structural transition along with an electronic transition was confirmed with temperature dependent resistivity and Raman measurements. The crossover from low-field positive magnetoresistance (MR) to high field negative MR at room temperature is attributed to suppression of spin scattering effect. This spin scattering suppression is led by defect induced ferromagnetism as confirmed from the magnetic M-T, M-H data. This is further verified by the spin polarized density of states calculation by adding some defects into the crystal structure.

In **Chapter 4** we unfold a unique semiconductor to metal transition, an electronic topological transition (ETT) and 3 structural transitions (Rhombohedral→monoclinic C2/m→ another monoclinic C2/c → disordered cubic Im-3m phase) in Cu doped $Sb_2Te_3$ while high pressure measurements. The structural phases are analyzed by Reitveld/Le-Bail refinement at various pressure. Importantly the semiconductor to metal transition occurred at a pressure below 1 GPa which is lowest amongst the TI family of compounds ($Bi_2Te_3$, $Sb_2Te_3$, $Bi_2Se_3$). The unusual softening of two newly Raman modes combined with sudden collapse in bond length, angle of succession, intralayer distance and the direct observation of band gap closure from density function theory (DFT) validates this metallic transition. The ETT is found at a lower pressure compared to the parent one and confirmed by c/a minimum.

Additionally, we also reveal the high-pressure studies of Fe and S co-doped $Sb_2Te_3$ system that emerge one ETT and 3 structural transitions with increasing pressure. Our studies unravel how suitable choice of dopant can lower the ETT pressure. Importantly, the initial rhombohedral phase is not retrieved with removal of pressure, rather transformed into an alloy. These studies indicate that appropriate doping can indeed help us to trap unique states at ambient pressure and can thus help in synthesizing new materials.

**In Chapter 5** we deal with the characterization of the Dy doped $BiSbTe_3$ topological insulator. The resistivity measurements demonstrate nonlinearity in the Hall behaviour. Due to the widespread applicability in magnetic sensors, random access memory, and spin logic devices, AHE is gaining tremendous research interest in condensed matter physics. Our findings shed light on the TI $Bi_{0.95}Sb_{0.95}Dy_{0.1}Te_3$ with AHE at temperatures much above antiferromagnetic ordering (Neel temperature $T_N$~44K). Up to magnetic transition temperature it is the concomitant effect of magnetism and the frustrated lattice that realizes this extraordinary effect. The AHE above $T_N$ is attributed to thermal average of the spin chirality in the system. The present observation exhibits short range spin correlation and noncoplanar spin textures thus possess a large Hall angle compared to the usual limit of scattering. The nontriviality of the material was envisaged by the SdH oscillation study. From the magnetic AC susceptibility study, we aim to confirm the glassy nature at low temperature. Our studies demonstrates that Dy-doped BST is not only a topologically nontrivial material, but exhibit a frustrated cluster spin glass nature at low temperature.

**In Chapter 6** we confirm Topological magnetic structure in Fe and S co-doped Topological insulator $Sb_2Te_3$ and $Bi_2Te_3$. The systems are characterized with magneto-transport, magnetic AC susceptibility at different frequency, M-T, M-H, dHvA, SdH, temperature dependent Raman and ARPES techniques. A unique combination of disordered glassy phase, competitive FM-AFM interactions and nontrivial topology make the system distinct from other TIs. Topological frustrated magnets, which can host both magnetic frustrations and Dirac quasiparticles, are highly sought after class of compounds. Furthermore, as seen by the quantum oscillation study, the Fermiology changes with doping and produces multiple Fermi pockets, revealing a rich complexity in the underlying electronic structure. We have discussed the impact of those complicated magnetic phases upon the observed

AHE in $Sb_{1.9}Fe_{0.1}Te_{2.85}S_{0.15}$ and $Bi_{1.9}Fe_{0.1}Te_{2.85}S_{0.15}$ with magnetotransport studies. The in depth dHvA and SdH oscillation complements the ARPES study.

**In Chapter 7** we executed density functional theory (DFT) to address the electronic properties of various compounds under pressure. The idea is to tune electronic properties under extreme conditions and create rich physics and hold them as candidates for optoelectronics, spintronics and nanoelectronics. The electronic character of the band structure and density of states (DOS) exhibit a metallic transition near 3 GPa for both MoTeSe and WTeSe whereas they showed a semimetallic behaviour at ambient condition. The present work depicts a pressure studies up to 10 GPa to explore the lattice parameters, volume changes and c/a anomaly. Out of the two, WTeSe shows a Dirac like dispersion without any band crossings along Z-U symmetry path.

The density functional theory (DFT) calculation shows electronic topological transition (ETT) under the application of pressure in $Sb_2Te_3$ Topological insulator. When pressure is applied conduction band and valence shifts significantly which is indicated as electronic transition of the Fermi surfaces. The band gap between conduction band and valence band reduced at Z-high symmetry point whereas it is observed to enhance at Γ-point with increase in pressure indicating the presence of ETT. Interestingly, we found semiconductor to metal transition near 2 GPa with LDA approximation. The overall change in the Fermi surfaces is found to follow other topological insulators like $Bi_2Te_3$ and $Bi_2Se_3$. The topologically non-trivial surface state remains unchanged under pressure; however, the bulk band gap reduces.

**In Chapter 8** we briefly discuss the applications of these extraordinary class of materials in the field of optoelectronic devices, p-n junction, superconducting materials, field effect transistor, memory device and spintronics, ultrafast photodetection and quantum

computations. We have also presented the summary of the present thesis with a brief future scope of work.