Magnetic and Transport Properties of Spin Modulated Bulk and Heterostructured Quantum Materials



# THESIS SUBMITTED IN PARTIAL FULFILLMENT FOR THE AWARD OF DEGREE

# Doctor of Philosophy

By

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#### 7.1 Summary

This thesis is devoted to experimental studies of the structural, magnetic, electronic and thermoelectric properties of different spin-modulated quantum materials in the form of bulk and heterostructures. The high-quality single crystal samples of topological insulators, ferromagnetic Weyl semimetals were synthesized by following modified Bridgman technique and heterostructures of dichalcogenide topological insulator and ferromagnetic chalcogenides were grown by pulsed laser deposition technique. The crystalline nature and surface morphology of all grown systems were confirmed by XRD and HRSEM and AFM. Further, the extensive electrical, transport and magnetic measurements have been performed along with vibrational Raman spectroscopy.

In **Chapter 3**, the temperature dependency of the phonon dynamics in the  $Bi_{1.9}Dy_{0.1}Te_3$  was described by the anharmonicity of phonon-phonon interaction determining the lattice contribution of thermal conductivity as 1.1095 Wm<sup>-1</sup>K<sup>-1</sup> using Umklapp method, estimating anharmonic Gruneisen parameter. Further, the electronic contribution of thermal conductivity was estimated as 0.8709 Wm<sup>-1</sup>K<sup>-1</sup> employing the Wiedemann–Franz law. Hence the total thermal conductivity was used to calculate the figure of merit evaluated as 0.7764 using the values of Seebeck coefficient and electrical conductivity at room temperature which is even larger than that for pure  $Bi_2Te_3$  and have been supported by DFT calculation. Thus, the single crystalline  $Bi_{1.9}Dy_{0.1}Te_3$  demonstrates the possibility of being used as an excellent thermoelectric material.

In **Chapter 4**, prepared FSBS5, FSBS10 single crystals were compared with FS/BS/FS layered structure perusing the structural, electronic, physical and magnetic measurements. After confirming the structural and morphological transparency of both the systems, the XPS spectra were analyzed in depth comparing all the present elements Bi, Se, and Fe in both the bulk and low dimensional systems. The FS/BS/FS heterostructure

demonstrates a Kondo-like nature below 12 K justifying the manifestation of interfacial magnetic domain structures in the system. In addition, The WAL effect is dominating in the heterostructure at lower temperatures admitting the augmentation of the surface state at lower dimensions. Thus, irrespective of the fact that the interfacial magnetic impurity adequately causes disruption in TRS emerging a finite gap at the Dirac point, the confinement in dimensionality effectively enhances the surface state in such a 2-D layered system.

**Chapter 5** demonstrates the hetero-structure manifesting two different chalcogenide systems prepared using the PLD technique exhibits a metal-semiconductor transition below 150 K. This transition was further confirmed to be initiated by electron-phonon interaction, which was probed by investigating the Raman shift with varying temperatures. Magnetic measurements show the existence of anti-parallel spin alignment at very low, which is consistent with the well-established Kondo model fitted in the resistance data below 15 K. Further, the magnetization curve displays sudden drop adjacent to 475 K, signifying some local charge ordering at the vicinity of interface. This, local magnetic ordering evidencing the anomalous behavior in Raman shifts demonstrates the signature of spin-phonon coupling driven by charge ordering at the edges of the particular 75.3 nm thick hetero-structure. Thus, the proximity-induced local charge orientation at the interfaces of the tetragonal FeSe and rhombohedral Sb<sub>2</sub>Te<sub>3</sub> is modified enormously depending on the confinement of the sandwiched structure.

In **Chapter 6**, Systematic utilization of chemical pressure from the partial replacement of Sulphur by relatively larger Selenium atoms in the lattice of  $Co_3Sn_2S_2$  yields extended temperature stability of a Skyrmion-like magnetic phase at an exceptionally small applied magnetic field. We inspected  $Co_3Sn_2S_{2(1-x)}Se_{2x}$  systems demonstrating Skyrmion-

like phase by observing the magnetization curves and studying isothermal  $\chi'_{ac}(H)$  and  $\chi''_{ac}(H)$ . Interestingly, with a larger doping concentration of Se atoms the Skyrmionic phase space is expanding vibrantly. The enhancement in the temperature range of the complex magnetic structure is introduced by reorientation of the spins due to the tension created on the crystal chemically by the replacement of Se over S. This phenomenon unswervingly affects the stabilization of the Skyrmion phase in the Kagome ferromagnet positively even at such a small size, presenting such materials to become the information carriers in the forthcoming spintronics devices.

### 7.2 Future Perspectives

The interplay between topology and magnetism is emerging as the new frontier in fundamental quantum physics after the discovery of Topological materials. These materials have Dirac and Weyl quasi-particle excitations, exhibit extraordinary properties from the viewpoint of both fundamental and technological enthusiasm. The focus of the present work was to observe some interesting results exploring new quantum phases and modification of magnetic orders in the materials by manipulating internal and external parameters of some well-studied TIs and WSMs for the benefit of fundamental research and technological applications. Few given goals may be achieved in the future.

 We have succeeded to achieve the stability of the magnetic Skyrmion phase employing chemical pressure. Therefore, we have planned to prepare two dimensional layers of such Kagome magnet to match the stress induced mechanism, hence propose the further enhancement of Skyrmion stability conveying Co<sub>3</sub>Sn<sub>2</sub>S<sub>2(1-x)</sub>Se<sub>2x</sub> as a potential candidate to be applied in spintronics devices.

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2. Also, thermoelectric or magnetic field induce Nernst effects can be measured by the fabrication of suitable devices using such Kagome magnets in 2D and convey such materials in future energy storage and thermoelectric applications.