ABSTRACT

Topological crystalline insulators (TCIs) such as SnTe, with a rock-salt crystal structure, exhibit four Dirac cones—one each on the (001) and (110) surfaces and two on the (111) surface—centered at the Γ and M points. In contrast, conventional topological insulators (TIs) like Bi$_2$Te$_3$ and Bi_2Se_3 possess only a single Dirac cone. The presence of topological surface states (TSS) in SnTe has been confirmed through angle-resolved photoemission spectroscopy (ARPES) and scanning tunnelling microscopy (STM). Theoretically and experimentally, it is established that the formation energy of Sn vacancies is much lower than that of Te vacancies. Consequently, SnTe exhibits a high Sn-vacancy concentration ($10^{21}/cm^3$), which disrupts its crystal symmetry through Sn-vacancy migration (hole formation) and partially destroys the TSS. Enhancing TSS conduction, therefore, requires minimizing the bulk carrier contribution via nanostructuring and elemental doping. Techniques such as MBE and CVD for thin-film growth, and hydrothermal or solvothermal methods for chemical synthesis, are widely employed to produce nanostructured TCIs. Elemental doping increases the Sn-vacancy formation energy and substitutes for missing Sn atoms, thereby restoring crystal symmetry and improving the mirror-symmetry-protected TSS. When magnetic impurities are introduced into SnTe at concentrations comparable to its intrinsic Sn-vacancy level, they reduce hole conductivity through substitutional incorporation or charge-transfer-induced hole localization, leading to enhanced TSS conductance. This process further enables carrier-mediated anisotropic Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions, which are influenced by strain, band gap, and impurity separation across different sublattices. The origin of magnetism in TCIs and magnetic TCIs remains an open question. However, magnetic doping in TCIs offers a distinct advantage over TIs, as the TSS protection in TCIs arises from mirror symmetry rather than time-reversal symmetry. Consequently, magnetic moments of impurities do not destroy TSS protection but instead couple the TSS with spin-split impurity bands, producing proximity-induced magnetic anisotropy. In SnTe, magnetic doping not only reduces the Sn-vacancy concentration but also localizes itinerant holes through charge transfer, introducing defect- and impurity-induced strain. These effects create a scalar perturbative potential (u) that breaks electron–hole symmetry, enabling spin-independent electron scattering. Simultaneously, magnetic proximity coupling of impurity ions induces crystal-field splitting and orbital hybridization, described by the magnetic scattering vector (m). This interaction scatters the spins of itinerant carriers through localized magnetic moments, resulting in competing canted antiferromagnetic (AFM) and ferromagnetic (FM) RKKY interactions. This thesis aims to investigate how structural defects (such as Sn vacancies) and magnetic impurities influence anisotropic magnetic interactions in the presence of spin–orbit coupling (SOC), and to explore their potential applications in catalysis. (i) Proximity-coupled magnetic anisotropy and WAL response of disordered bare and transitional metal (V) doped SnTe nanoparticles. (ii) Topological spin dynamics in Eu-doped SnTe, akin to anisotropic spin-flop transition and WAL response. (iii) The role of defect and charge transfer induced localization on magnetic anisotropy and WAL response in MWCNT and RGO anchored SnTe.

Part 1: We synthesized bare and V-doped SnTe nanoparticles using the chemical reflux method. Their deformed disc-like morphology, with radii ranging from 5–50 nm, was characterized using HRTEM and FEGSEM. The optical properties were investigated through Raman, photoluminescence (PL), and FTIR spectroscopy, while XPS measurements confirmed the charge states and impurity configurations. DFT calculations provided both qualitative and quantitative insights into charge transfer effects in the presence of Sn vacancies and V impurities, and further revealed how interstitial and substitutional V incorporation influences the magnetic behavior of the system. DC magnetisation and ESR measurements at room and low temperatures demonstrated the presence of an anisotropic, competing itinerant carrier-mediated Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, evidenced by distinct

normal and parallel components of the internal magnetic fields. Additionally, the low-temperature ESR spectra, recorded while sweeping the magnetic field from negative to positive regimes, exhibited a differential magnetoconductance, whose integrated response revealed both negative magnetoconductance (NMC) and weak antilocalization (WAL) characteristics.

Part 2: We synthesized bare and Eu-doped SnTe nanoparticles using the chemical reflux method. Their deformed disc-like morphology, with radii ranging from 5–50 nm, was characterized using HRTEM and FEGSEM. The optical properties were investigated through Raman, photoluminescence (PL), and FTIR spectroscopy, while XPS measurements confirmed the charge states and impurity configurations. DFT calculations provided both qualitative and quantitative insights into charge transfer effects in the presence of Sn vacancies and Eu impurities, and further revealed the anisotropic strain and magnetic moment-dependent magneto-elastic energy and magnetic behavior of the system. DC magnetization and ESR measurements (performed at both room and low temperatures) demonstrated the presence of an anisotropic, competing itinerant carrier-mediated Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, evidenced by spin-independent electron scattering by defect and stress-induced scalar perturbative potentials (u) and spin-dependent scattering by proximity-coupled magnetic scattering vector (m). A magnetic field–dependent spin-flop transition of Eu^{+3} spins within host TCI SnTe is evident in the anomalous M–H curves and AC susceptibility at low temperatures (2.9 K). Additionally, the low-temperature ESR spectra, recorded while sweeping the magnetic field from negative to positive regimes, exhibited a differential magnetoconductance, whose integrated response revealed both negative magnetoconductance (NMC) and weak antilocalization (WAL) characteristics.

Part 3: We synthesized bare, MWCNT-anchored, and RGO-anchored SnTe nanoparticles using the Hummers' method and the chemical reflux technique. The deformed disc-like morphology of SnTe, with radii ranging from 5 to 50 nm, and the anchoring of SnTe on the surfaces of MWCNT and RGO were characterized using HRTEM and FEGSEM. The optical properties were examined using Raman, photoluminescence (PL), and FTIR spectroscopy, while XPS measurements confirmed the charge states and impurity configurations. DFT calculations provided qualitative and quantitative insights into charge transfer mechanisms involving Sn vacancies and surface defects on MWCNT and RGO during the reduction process. Furthermore, DFT revealed the influence of anisotropic strain and magnetic moment–dependent magnetoelastic energy on the overall magnetic behavior of the system. DC magnetization and ESR measurements, conducted at both room and low temperatures, confirmed the presence of an anisotropic, competing itinerant carrier-mediated Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, arising from spin-independent electron scattering due to defect- and stress-induced scalar perturbative potentials (u). At elevated temperatures and magnetic fields, the system exhibited a transition toward a superparamagnetic or paramagnetic state, attributed to the suppression of the RKKY interaction caused by loss of crystal symmetry and spin–orbit coupling (SOC) effects. Moreover, low-temperature ESR spectra, recorded while sweeping the magnetic field from negative to positive regimes, displayed differential magnetoconductance, whose integrated response revealed both negative magnetoconductance (NMC) and weak antilocalization (WAL) features.