Investigation of anomalous Hall transport and related phenomena in some Mn-based intermetallic compounds

Thesis submitted in partial fulfillment

for the Award of

DOCTOR OF PHILOSOPHY

in

MATERIALS SCIENCE

by

NISHA SHAHI

SCHOOL OF MATERIALS SCIENCE AND TECHNOLOGY

INDIAN INSTITUTE OF TECHNOLOGY

(BANARAS HINDU UNIVERSITY)

VARANASI – 221005

ROLL NUMBER 18111005

YEAR OF SUBMISSION 2023

Chapter 8

Summary and Suggestions for Future Work

8.1 Summary

In the present thesis, intriguing transverse transport properties such as anomalous Hall effect, topological Hall effect have been investigated in the Heusler Mn_2CoAl, Mn_2NiGa, pnictide MnSb and antiperovskite Mn_3GaN compounds. These transport phenomena have attracted a large amount of interest in recent years due to their potential in understanding the fundamental physics and applications in spintronics-based data storage devices and Hall sensors. Furthermore, interesting longitudinal transport properties such as the Kondo effect, and the correlation between magnetic and electronic transitions, including metal-to-insulator transitions, have been well established in the antiperovskite Mn_3GaN compound.

These studies have been carried out on polycrystalline bulk (prepared using arc melting technique) and thin film (prepared by magnetron sputtering technique) systems, which followed by vacuum annealing as per requirement. The structural characterization has been performed using laboratory x-ray diffraction and high resolution synchrotron x-ray diffraction technique (large-scale experiment facility at the P02.1 beamline of PETRA-III, DESY, Hamburg, Germany). The homogeneous growth and composition of the prepared compounds have been confirmed by collecting the back scattered image and energy dispersive x-ray data, respectively. The physical properties such as magnetization, AC-susceptibility, resistivity, magnetoresistance and Hall has been measured using in house experimental facilities like physical property measurement system and cryogen-free measurement system. The key findings of the present study are summarized below:

(1) In **Chapter 3**, we establish the evidence of anti-site disorder enhanced intrinsic AHC in the Mn_2CoAl Heusler compound by comprehensive analysis of the crystal structure and anomalous Hall effect using experimental and theoretical tools. The high-resolution SXRD data reveals 25% anti-site disorder between Mn_{4c} and Al atoms within inverse Heusler structure. The temperature-dependent resistivity shows semiconducting behavior and follows Mooij's criteria for disordered metal. Scaling behavior suggests that the intrinsic mechanism dominates over the extrinsic mechanism in the AHE. The experimental intrinsic AHC is found to be larger than the theoretically reported value for the ordered Mn_2CoAl. The first-principle calculations conclude that the anti-site disorder enhances the Berry curvature induced intrinsic AHC, which is in good agreement with the experimentally found

intrinsic AHC.

(2) In **Chapter 4**, we present a giant THE in the bulk Mn_2CoAl compound, which is generally observed due to the microscopic non-coplanar spin texture/mesoscopic skyrmion like topological spin texture. The AC-susceptibility measurements, which do not exhibit any anomaly, suggest the absence of skyrmion-like topological spin texture. The reported theoretical studies suggest the presence of ferromagnetic and antiferromagnetic exchange interaction between magnetic atoms (Mn and Co), which gives rise to non-collinear magnetic texture as a result of the strong magnetic frustration. The presence of the anti-site defect in the Mn_2CoAl compound may further enhance the magnetic frustration and result in non-collinear magnetic texture. The THE in the Mn_2CoAl compound may arise due to the formation of non-coplanar spin texture as a result of the competition between cubic anisotropy, and ferromagnetic and antiferromagnetic exchange interactions.

(3) In **Chapter 5**, we demonstrate that the AHE in the high-temperature cubic phase is mainly driven by extrinsic skew scattering and side jump mechanisms. The analysis of temperature-dependent resistivity and magnetoresistance reflects that electron magnon scattering dominates in the high-temperature cubic region of the Mn_2NiGa compound. The anomalous Hall resistivity due to side jump and intrinsic mechanism is in linear relationship with the magnetoresistance at high field, which indicates that the high temperature anomalous Hall due to side jump and intrinsic mechanism is primarily governed by the side jump mechanism originating from the electron-magnon scattering. Whereas, in the low-temperature martensite phase, the side jump contribution can be omitted and the intrinsic mechanism has a nearly equal contribution to the AHE as the skew scattering. The experimental findings suggest that the AHC in the low-temperature martensite phase is governed by the intrinsic mechanism to a greater extent as compared to the high-temperature phase. The theoretical calculation in the austenite cubic phase gives a smaller value of intrinsic AHC than the martensite tetragonal phase, which is in agreement with the experimental result that the intrinsic mechanism is being suppressed as we move toward the highly symmetric cubic phase from the less symmetric tetragonal phase. These results make one additional conclusion that there is no straightforward rule to connect the Berry curvature to the crystal symmetry; rather, it depends on the symmetry-induced change in the electronic band structure.

(4) In **Chapter 6**, we report a combined experimental and theoretical investigation of the AHE in the MnSb manganese pnictide. Temperature-dependent magnetization measurement reveals an anomalous drop in magnetization below $\sim$120 K, which is anticipated due to the SRT. Magneto-transport data demonstrates the non-monotonic temperature dependence of negative magnetoresistance, which increases from room temperature to the SRT temperature (T_{SR}) $\sim$120 K, but then decreases and becomes positive at very low temperatures. A similar non-monotonic temperature dependence is observed in the AHC, which is nearly constant in temperature range from room temperature to T_{SR}, followed by a drop and sign reversal at low temperatures. Detailed scaling analysis of anomalous Hall data suggests that the AHE above T_{SR} is primarily governed by the intrinsic Berry curvature and the obtained value of intrinsic AHC is about 310 S/cm. In contrast, below T_{SR} the extrinsic skew scattering becomes the dominant contributor to the AHE compared to the intrinsic Berry curvature and the obtained value of intrinsic AHC is about -28 S/cm. Our first-principles calculations reveal that changes in the sign and magnitude of the intrinsic AHC are attributed to modifications in the Berry curvature associated with band splitting near the Fermi level. The Berry curvature modification emerges from the reconstruction of electronic band structure, under the variation of spin orientation from the c-axis to the ab-plane of crystal system due to the SRT.

Besides the AHE, we report the topological Hall effect, which is independent of temperature in low-temperature region below T_{SR}, while indicating a large dependence on temperature above T_{SR}. A detailed micromagnetic simulation confirms that the emergence of skyrmionic bubbles due to competition of the uniaxial MCA with different energies is responsible for the THE below T_{SR}, which is also supported by the magnitude (in nΩ.cm) and temperature independent behavior of THE. In contrast, above T_{SR} the simulation findings, and the comparatively large ($\mu\Omega$.cm) and temperature-dependent behavior of the THE suggest that the THE could be attributed to the microscopic non-coplanar spin texture, rather than being solely driven by skyrmionic bubbles.

(5) In **Chapter 7**, we present evidence for complex interplay of lattice, electronic, and spin degrees of freedom in the temperature dependence of the electrical resistivity (ρ_{xx}-T) of polycrystalline thin film samples of antiperovskite compound Mn_3GaN in the 2-300 K range. The analysis of the ρ_{xx}-T in terms of the Bloch-Gruneisen model reveals signatures of the paramagnetic (PM) to the AFM Γ_{5g} as well as the Γ_{5g} to the ferrimagnetic M-1 transitions reported recently in a neutron diffraction

study [K. Shi et al., Adv. Mater. **28**, 3761 (2016)] with different Debye temperatures for the two phases. Interestingly, the ρ_{xx}-T data shows a resistivity minimum at 6 K, which provides the first evidence for Kondo spin-flip scattering in the family of antiperovskites. The Kondo effect in our sample has been confirmed by the observation of logarithmic upturn in ρ_{xx}-T below the Kondo temperature T_K ~5.5 K and negative magnetoresistance with quadratic magnetic field dependence below and slightly above the T_K at low fields. The analysis of the low field magnetoresistivity using the numerical renormalization group n-channel Kondo model at 2 K provides additional support for the presence of the Kondo effect in Mn3GaN thin film. Our results show that the ρ_{xx}-T below the resistivity minimum temperature $T_{\rho\text{-min}}$ can be modeled in terms of the dominant Kondo contribution with some contribution from the electron-electron interaction while above $T_{\rho\text{-min}}$ consideration of Fermi liquid and electron-phonon interactions are also required up to 15 K. In addition to the exotic features in resistivity data, the Hall data reflects the presence of AHE, which might be related to a small finite rotation from the Γ^{5g} spin configuration.

8.2 Suggestions for future work

In the present study, the fascinating structural, electronic and magnetic properties has been studied on the polycrystalline Mn_2CoAl, Mn_2NiGa, MnSb and Mn_3GaN compounds. The results open several possibilities for future experimental and theoretical investigations. In future, these studies on single crystal and thin film systems will be important from the point of view of fundamental science and technological application. To further investigate these properties, we can directly probe the modulation in electronic band structure using angle-resolved photo-emission spectroscopy technique, and can also check the magnetic spin texture using several direct tools such as neutron diffraction, Lorentz transmission electron microscopy, magnetic force microscopy, and spin-polarized scanning electron microscopy etc. Besides, we outline a few of them given below:

(1) The combined experimental and theoretical studies of structural and magneto-transport properties of the spin gapless semiconducting Mn_2CoAl compound, explicitly suggest that the inherent anti-site disorder affects the electronic band structure. The change in band structure actually enhances the momentum space Berry curvature and further enhances the AHC in the Mn_2CoAl compound. Hence,

we may modulate the AHC by introducing any kind of disorder externally such as compositional disorder or by doping with foreign elements, which may shift the Fermi level and/or can affect the Berry curvature linked with the electronic band structure. Nowadays, there is controversy in this field regarding whether disorder enhances or suppresses the AHC as a combined effect of crystal symmetry and spin orbit coupling. For that we have synthesized polycrystal system of the Mn_2CoAl compound doped with various amount of silicon at the place of Al. In future, we will perform magnetic and electrical transport measurements by which we can further establish a connection between disorder and AHC with the help of experimental as well as theoretical investigations. In addition, the enhanced AHC due to the anti-site defect in the Mn_2CoAl compound paves the way to explore other untouched Heusler compounds, which are expected to be enriched with antisite defects/disorder.

(2) The Hall measurement in the Mn_2CoAl compound suggests the presence of THE in addition to the ordinary and anomalous Hall effect. The THE might be originated due to the non-coplanar spin texture with non-vanishing spin chirality as a result of the competition between exchange interaction, and magnetocrystalline anisotropy, which is confirmed by performing the micromagnetic simulation. The additional real space measurements such as neutron diffraction or Lorentz-transmission electron microscopy (LTEM) is still needed to confirm the presence of non-trivial magnetic textures. There are many other cubic Heusler compounds, which are untouched and need to explore in this context. Such non-coplanar structure can be tuned into a topologically stable skyrmionic structure by changing physical parameters such as thickness and ordering of the system, which has the potential to replace the magnetic domain in the future magnetic storage devices.

(3) The magnetic shape memory (MSM) Mn_2NiGa compound, exhibits structural phase transition from cubic austenite phase (in high temperature region) to tetragonal martensite phase (in low-temperature region), is showing distinct origin of the observed AHE above and below the phase transition. In the austenite phase, the AHC is mainly originated by extrinsic mechanisms (skew scattering + side jump). Whereas, the AHC in martensite phase, is basically originated by intrinsic mechanism along with the skew scattering contribution. The theoretical study also suggest lowering of intrinsic AHC in austenite phase. Similar study can also be perform in other MSM Heusler compounds to insight the fundamental science behind the origin of AHE and its connection with

the crystal symmetry. In addition, the experimental realization of band structure [by performing angle-resolved photo-emission spectroscopy (ARPES) measurement] is needed to further confirm the phenomena actually happening in the Mn_2NiGa compound.

(4) The Mn-based pnictide MnSb compound, exhibits strong hybridization, have a number of linear band dispersion near the Fermi level, one of which is splitted around high symmetry point Γ as incorporated the spin orbit coupling. The band splitting gives rise to the intrinsic AHC depending on the magnetization orientation. The MnSb compound has been anticipated to show spin reorientation transition, around which we observed crossover from intrinsic to extrinsic AHC (less intrinsic contribution) with the change in sign. The theoretical calculation indicates that changes in the magnitude and sign of intrinsic AHC is related to the anisotropic band structure for different spin orientation around SRT. In addition, we have observed the presence of THE, possibly arising due to the emergence of skyrmionic bubble and/or microscopic non-coplanr spin texture. To probe the magnetic structure, neutron diffraction study and/or LTEM is needed for direct investigation. Additionally, this finding suggest to explore other pnictides in context of electronic band structure, AHE and THE.

(5) The resistivity data of the Mn_3GaN compound exhibit a number of transitions coinciding with the magnetic transitions based on the reported neutron diffraction study. We found metal to insulator and insulator to metal transition along with low temperature upturn due to Kondo effect. In the Mn_3GaN type compounds with antiperovskite structure we may tune the dilute Kondo system towards dense Kondo insulator behavior as a function of the anti-site disorder similar to that reported in the AFM Weyl semimetals like $Mn_{3+x}Sn_{1-x}$ but different from the way it appears in the heavy fermion systems. In addition, we found AHE by performing Hall measurement in the Mn_3GaN compound that might be originated from the spin canting due to the coexisting phase of ferromagnetic ordering. We believe that our findings open new possibilities for exploring the fascinating magnetotransport properties and electronic correlation effects in the family of geometrically frustrated antiferromagnetic X_3BA type compounds, containing other possible elements at the X (such as Co, Fe, Ni), A (such as C, O) and, B (such as Ge, Cu, Zn) sites with different concentration of anti-site disorder.