Preface

Shape memory alloys (SMAs) are a fascinating class of materials that have the remarkable ability to return to their original shape after undergoing deformation under external stimuli such as temperature or stress. This unique property is primarily driven by a reversible martensite phase transformation between the austenite and martensite phases. Recently, a unique class of SMAs has gained significant attention for its ability to undergo controlled shape transformation and generate substantial strain when exposed to an external magnetic field. These materials, known as magnetic shape memory alloys (MSMAs), are highly valued for their large, rapid, and reversible strain response to magnetic fields, making them ideal for applications in magnetic sensors and actuators. Ni-Mn-In-based MSMAs are notable for their exceptional technological properties, including the giant barocaloric effect, reversible magnetocaloric effect (MCE), elastocaloric effect, superelastic stability, colossal negative magnetoresistance, anomalous Hall effect (AHE), topological Hall effect (THE), Nernst effects, large exchange bias, and the zero-field skyrmionic phase. These key technological properties are linked to the composition and the magnetostructural transition between the austenite and martensite phases. Typically, Ni-Mn-In-based MSMAs undergo a direct transition from the austenite (high symmetry) phase to the martensite (low symmetry) phase upon cooling. The large MCE observed in these materials arises from the significant change in magnetization at their first-order magnetostructural (martensitic) phase transition (FOMST). However, while this FOMST enables a large MCE, it also results in substantial thermal hysteresis, leading to the irreversibility of the phase transition under repeated magnetic field cycles. This irreversibility presents a major challenge in the development of magnetic refrigeration devices. One approach to minimize hysteresis is through atomic substitution at specific lattice sites. Two key parameters, isothermal entropy change (ΔS_{iso}) and adiabatic temperature change (ΔT_{ad}), are used to estimate the MCE. Indirect measurement (i.e., measuring ΔS_{iso} or indirectly estimating ΔT_{ad}) remains a popular choice due to its practicality and ease of use. However, it is always subject to measurement errors. Direct measurement of ΔT_{ad} provides the true value of MCE in materials, which is crucial to determine whether the materials can be used in real-life cooling applications. Recent studies have demonstrated that certain Ni-Mn-In-based MSMAs (such as $Ni_2Mn_{1.4}In_{0.6}$) transition from the austenite phase to the martensite phase through an intermediate premartensite phase (PM-phase) during cooling. The PM-phase plays a crucial role in facilitating the formation of skyrmionic textures. Generally, the PM-phase exists within a narrow temperature range. However, a recent study suggests that chemical doping can stabilize the PM-phase over a wider temperature range. Since skyrmions are a source of the THE, studying the temperature-dependent magnetotransport properties in the PM-phase is essential for these MSMAs. There is another class of material MnPtGa is known to host Néel skyrmions, which are topologically protected spin structures. MnPtGa displays thermodynamically stable bulk-type Néel skyrmions within the temperature range of 210–220 K. Achieving stable skyrmions near room temperature is highly desirable for technological applications. In the MnPtGa system, the ferromagnetic transition temperature (T_C) increases with hydrostatic pressure at a rate of $dT_C/dP{\sim}1.7$ K/kbar. This suggests the potential to shift the stable skyrmion temperature range closer to, or even beyond, room temperature. Therefore, a thorough pressure-dependent study of the PtMnGa system is crucial. In the present thesis, a detailed study of the magnetic states, MCE, AHE, THE, and simulation of the skyrmions are performed in $Ni_2Mn_{1.4}In_{0.6}$, $Ni_{1.9}Pt_{0.1}Mn_{1.4}In_{0.6}$, $Ni_{50}Mn_{34}In_{15.2}Al_{0.8}$ MSMAs, and a related hexagonal MnPtGa system. The research conducted for this thesis is presented in eight chapters, with a brief summary of each chapter and its key findings provided below:

- **Chapter 1** provides a detailed introduction, offering comprehensive explanations of shape memory alloy, martensitic transformation, magnetic shape memory alloy, magnetocaloric effect, Heusler alloys, anomalous Hall effect, topological Hall effect, and other key terms relevant to this work. This is followed by a literature review focused on Ni-Mn-In-based MSMAs and the related MnPtGa hexagonal system.

- **Chapter 2** outlines the synthesis process, offering a detailed discussion of the various components and characterization techniques employed. These techniques include laboratory-based X-ray diffraction for phase purity assessment. Furthermore, the chapter describes the magnetic and magnetotransport measurements carried out using a physical properties measurement system. It also covers the synchrotron X-ray powder diffraction (SXRPD) measurements conducted at the P02.1 beamline of PETRA-III and the Xpress beamline of Elettra under varying temperatures and pressures. Finally, the chapter provides an overview of the muon spin relaxation experiments.

- **Chapter 3** introduces a simple prototype utilizing a permanent magnet to measure the direct MCE (ΔT_{ad}) at room temperature. Additionally, a measurement setup with a 1 T electromagnet is designed to measure the ΔT_{ad} over a temperature range from ambient temperature to 100 K. Both setups use a PT100 temperature sensor, selected for its precision in magnetic fields and minimal thermal losses.

- **Chapter 4** explores the enhanced crystallographic compatibility and reversible MCE in a 10% Pt-substituted $Ni_2Mn_{1.4}In_{0.6}$ magnetic shape memory Heusler alloy. The 10% Pt substitution reduces the thermal hysteresis from approximately $\sim$8 K to $\sim$4 K. This reduction is linked to the middle eigenvalue λ_2 of the phase transformation matrix U, which is calculated to be very close to 1 (0.9982), indicating excellent crystallographic compatibility between the austenite and martensite phases. Furthermore, the nearly identical ΔS_{iso} values obtained from three different measurement protocols (isothermal, loop, and isofield) confirm the reversible MCE in $Ni_{1.9}Pt_{0.1}Mn_{1.4}In_{0.6}$.

- **Chapter 5** investigates the magnetic state in the so-called paramagnetic (PM) gap regime of the martensite phase in $Ni_2Mn_{1.4}In_{0.6}$ MSMA using muon spin relaxation technique. Muon spin-polarization undergoes significant decoupling at $\sim$395 K, accompanied by a sharp decrease in asymmetry. Above this temperature, the temperature-independent and large asymmetry value indicates the PM nature of the high-temperature phase. The gradual reduction in asymmetry with decreasing temperature signifies a magnetic transition from a disordered (PM) to an ordered (ferromagnetic/antiferromagnetic) state. The asymmetry remains low until the second magnetic transition ($T_C^M \sim 190$ K) in the martensitic phase, indicating that the magnetic state within the so-called PM gap regime cannot be purely disordered (PM). Interestingly, the transition temperature observed in the asymmetry vs. temperature plot is higher than the Curie temperature of $\mathrm{Ni_2Mn_{1.4}In_{0.6}}$. Temperature-dependent pair distribution function analysis reveals the presence of a local martensitic phase (with a finite magnetic moment) within the high-temperature austenitic phase, which accounts for the reduced asymmetry even above the Curie temperature.

- **Chapter 6** explores the intrinsic AHE and THE in the thermodynamically stable PM-phase of the $\mathrm{Ni_{50}Mn_{34}In_{15.2}Al_{0.8}}$ MSMA. This alloy exhibits complex magnetic and structural phase behavior, with the PM-phase playing a crucial role in enabling topological phenomena. The magnetotransport measurements reveal that AHE is driven by the Berry curvature in momentum space. Moreover, the emergence of a strong THE in the PM-phase underscores the presence of nontrivial spin textures, such as skyrmions, stabilized by the interplay of structural and magnetic interactions. These findings shed light on the coupling between electronic transport, magnetic configurations, and structural transitions in multifunctional materials, offering a foundation for developing advanced spintronic systems with controllable topological properties.

- **Chapter 7** investigates the effect of pressure on the structural properties of the MnPtGa hexagonal system, a known host for Néel skyrmions. Using pressure-dependent SXRPD, an isostructural phase transition is identified at approximately 6 GPa. Evidence for this transition is further supported by the observation of two distinct Birch-Murnaghan (B-M) equations of state (EoS) in the pressure–unit cell volume relationship. Analysis of combined pressure- and temperature-dependent SXRPD data suggests that applying moderate pressure (0.8–1.09 GPa) shifts the thermodynamically stable skyrmion regime closer to room temperature. Theoretical calculations under pressure corroborate these experimental results, highlighting the potential of pressure as a tuning parameter. Together, the experimental and theoretical findings illustrate the feasibility of engineering materials to support stable skyrmions at higher temperatures and modest pressures, achievable through chemical substitution or by strain in epitaxial thin films.

- **Chapter 8** provides the summary and suggestions for future work.