Chapter 5

Disorder-influenced first-order phase transition and thermomagnetic history dependence of functional properties in the Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy

In earlier Chapters it has been discussed that the key to multifunctional properties observed in Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy is a temperature (\(T\)) and magnetic field induced first-order martensite (MST)-austenite (AST) phase transition. Traditionally, Arrott plots (isothermal \(M^2\) vs. \(H/M\) plots, where \(H\) is the applied magnetic field and \(M\) is the measured magnetization of a sample) are used to analyse the ferromagnetic character of a material [5.1]. It has also been shown theoretically [5.2] that the sign of the slope of the \(H/M\) vs. \(M^2\) isotherms may be used to distinguish between the first- and second-order magnetic phase transitions. It was found that a positive slope of the \(H/M\) vs. \(M^2\) isotherm is related to the second-order magnetic phase transition, while a negative slope corresponds to the first-order magnetic phase transition. This criterion [5.2] has been employed for the identification of the order of the magnetic transitions in MnAs [5.2], Ru-doped CeFe\textsubscript{2} [5.3] and various other manganite systems [5.4-5.8]. The same criterion has also been used to investigate the magnetic transitions in the ferromagnetic shape memory alloy system Ni-Mn-Ga [5.9]. In the present Chapter, we perform a detailed investigation of the magnetic field-induced MST to AST phase transition in Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} by
studying $H/M$ vs. $M^2$ isotherms. We show that, though the martensitic transition (MT) in this alloy is first-order in nature, the criterion of negative slope of the $H/M$ vs. $M^2$ isotherms is not strictly applicable here. It is found that, instead of a negative slope, a decrease of slope is observed in some of the isothermal $H/M$ vs. $M^2$ curves in the temperature regime of the magnetic field-induced MST to AST phase transition. This has been correlated with the disorder-influenced nature of the first-order magnetic transition. Phase coexistence and thermomagnetic hysteresis is a generic feature of a disorder-influenced first-order phase transition and has been observed in various classes of magnetic systems undergoing a first-order magneto-structural phase transition [5.10]. In this Chapter we also probe the magnetic field induced MST to AST phase transition in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy using scanning Hall probe imaging experiments. The scanning Hall probe images clearly show the coexistence of the MST and AST phases across this magnetic field induced MST to AST phase transition and provide visual evidence of thermomagnetic hysteresis, implying that the phase transition is indeed first-order in nature. The magnetic field dependence of the volume fraction of the AST phase fits with an equation representing the Kolmogorov–Johnson–Mehl–Avrami (KJMA) relation [5.11] (originally proposed for study of crystallization of solids from melts), which suggests an underlying nucleation and growth mechanism of the phase transition. The local $M(H)$ loops constructed from the Hall images indicate the presence of a landscape of critical magnetic fields (for the MST–AST phase transition) distributed over the sample volume and thus confirm the disorder-influenced nature of the phase transition. In addition, the present results suggest that the functional properties (magnetoresistance (MR) and magnetocaloric effect (MCE) etc.) of Ni$_{50}$Mn$_{34}$In$_{16}$ will depend on the thermomagnetic history of the sample. In the present Chapter we also show from experimental results that the magnitude and reversibility of the functional
properties under application of a magnetic field, depend on the thermomagnetic history of the sample.

5.1 Influence of disorder on the martensite-austenite phase transition in Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy

In the following discussion we explore the disorder broadened first-order nature of the temperature and magnetic field induced MST -AST phase transition in Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy.

5.1.1 Temperature dependence of magnetization in the Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy

In section 3.1.3 of Chapter 3 it was mentioned that in Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy the onset temperature of AST to MST phase transition (\(T_{MS}\)) while cooling is higher than the onset temperature of MST to AST phase transition (\(T_{AS}\)) while heating the sample. This can also be seen in Fig. 5.1, which presents the temperature (\(T\)) dependence of magnetization (\(M\)) of Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy measured in a magnetic field (\(H\)) of 10 kOe in the field-cooled cooling (FCC) and field-cooled warming (FCW) protocols. The temperatures \(T_{MF}\) and \(T_{AF}\) are respectively the

![Graph showing temperature dependence of magnetization](image)

Fig. 5.1: Temperature (\(T\)) dependence of magnetization (\(M\)) for the Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy in 10 kOe magnetic field.
limits of supercooling (of the AST phase) and superheating (of the MST phase) and are identified as the temperature where the thermal hysteresis ceases to exist in the cooling and warming temperature path. These characteristic temperatures of the MST-AST phase transition in the alloy are marked in Fig. 5.1. In $H = 10$ kOe clearly $T_{\text{MS}}>T_{\text{AS}}$. This is true for all magnetic field values (Fig. 3.4 of Chapter 3).

5.1.2 Magnetic field dependence of magnetization in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy

Fig. 5.2 presents selected isothermal $M$ vs. $H$ curves for the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy. For each isothermal $M(H)$ measurement, the respective temperature was approached from 300 K in zero field. As discussed earlier (section 3.1.5 of Chapter 3), away from the temperature regime of the MT, the isothermal $M(H)$ curves are that of a soft ferromagnet. But the isothermal $M(H)$ curves

![Fig. 5.2: Isothermal magnetization ($M$) vs. magnetic field ($H$) curves of the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy at representative temperatures.](image-url)
in the temperature regime of the MT show an additional rise in $M$ above a critical $H$. The distinct rise in $M$ with increasing $H$ and the associated magnetic field-hysteresis signify a magnetic field induced first-order phase transition from the MST phase to the AST phase. The signatures of the magnetic field induced MST to AST phase transition mentioned above are observed more clearly in the $M(H)$ curves in the T-regime 200-242K.

5.1.3 $H/M$ vs. $M^2$ plots in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy

Fig. 5.3 shows representative $H/M$ vs. $M^2$ curves of the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy across the AST to MST phase transition (200-244 K). The $H/M$ vs. $M^2$ curves were drawn using the increasing magnetic field portions of the isothermal $M(H)$ curves. The negative slope region in the $H/M$ vs. $M^2$ curves in the temperature range 200-238 K corresponds to the portion of $M(H)$

![Fig. 5.3: Isothermal $H/M$ vs. $M^2$ plots at representative temperatures across the martensitic transition in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy.](image)
curves that represent a magnetic field induced first-order MST to AST phase transition. But in the temperature range 240-244 K, though the isothermal $M(H)$ curves show the signatures (explained in the preceding sub-section) of a first-order magnetic field induced MST to AST phase transition, the $H/M$ vs. $M^2$ isotherms do not exhibit any negative slope. In this temperature regime, a decrease of slope of the $H/M$ vs. $M^2$ curves is found to correspond to the magnetic field induced first-order MST to AST phase transition.

It has been shown theoretically that in systems undergoing a magnetic field induced first-order paramagnetic to ferromagnetic phase transition, the slope of the $H/M$ vs. $M^2$ curves should be negative [5.2]. The negative slope of $H/M$ vs. $M^2$ isotherms in such systems is found to decrease gradually as $T$ approaches the zero-field transition temperature [5.4-5.8]. Such decrease in the magnitude of the negative slope of $H/M$ vs. $M^2$ isotherms is also observed across the magnetic field induced first-order antiferromagnetic to ferromagnetic phase transition [5.3]. On the other hand, the absence of negative slope of $H/M$ vs. $M^2$ isotherms across a magnetic field induced first-order magnetic transition, as observed here, is also reported in certain compositions of ferromagnetic shape memory alloy Ni-Mn-Ga [5.9], and in another composition of the Ni-Mn-In alloy system viz. Ni$_{50}$Mn$_{35}$In$_{15}$ [5.12]. In Ni-Mn-Ga, as the temperature approaches a characteristic temperature, the negative slope of the $H/M$ vs. $M^2$ curves changes to a decrease of slope with an overall positive value across a paramagnetic to ferromagnetic phase transition (see Ref. 5.9, and Fig. 5(a) therein) as well as a ferromagnetic to ferromagnetic phase transition (see Ref. 5.9, and Fig. 3(a) therein).
5.1.4 Characteristic magnetic fields and temperatures of the field-induced martensite to austenite phase transition in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy

Instead of using the $H/M$ vs. $M^2$ plots, the first-order AST-MST phase transition in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy may be investigated in another way: We examine the temperature and magnetic field dependence of various characteristic parameters related to the first-order AST-MST phase transition in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy. The width of the transition (see Fig. 5.1) suggests that the transition is broadened by quenched disorder (see section 1.3.4 of Chapter 1 for a discussion on this topic), probably created during alloy formation. The disorder-influenced broadening of a first-order phase transition leads to a distribution of transition temperatures over the sample volume [5.13]. As a result, the phase transition line then broadens into a band [5.14-5.16]. The AST to MST phase transition region in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy with the lowering of temperature (see Fig. 5.1) extends from the temperature of MST start ($T_{MS}$) to the MST finish temperature ($T_{MF}$). As $T$ decreases from $T_{MS}$ to $T_{MF}$, the MST phase fraction increases from 0 to 1 and the AST phase fraction decreases from 1 to 0 [5.17]. Similarly the MST to AST phase transition with increasing temperature extends from the temperature of AST start ($T_{AS}$) to the temperature of AST finish ($T_{AF}$). Coexistence of MST and AST phases is observed in the temperature ranges $T_{MS}$ - $T_{MF}$ and $T_{AS}$ - $T_{AF}$. The temperatures $T_{MF}$ and $T_{AF}$ are respectively the limits of supercooling (of the AST phase) and superheating (of the MST phase) [5.18]. In section 3.1.3 of Chapter 3 we have noted that in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy $T_{MS}$, $T_{MF}$, $T_{AS}$ and $T_{AF}$ decrease with increasing magnetic field.

Similar to the case of a temperature driven transition, the magnetic field driven transition will also have a band of transition fields in the presence of disorder leading to a region of phase
coexistence [5.16]. This is visible in Fig. 5.2 where the MST-AST phase transition occurs isothermally over a width of applied magnetic field. We therefore characterize the magnetic field induced disorder-broadened first-order magnetic transition in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy by defining the characteristic magnetic fields of the transition. Figs. 5.4(a) and 5.4(b) present the $M$ vs. $H$ curve and the corresponding $dM/dH$ vs. $H$ curve at a representative temperature 224 K. When 224 K is approached from 300 K in zero field, both the MST and AST phases are present in the sample as.
this temperature is lower than $T_{MS}$ but higher than $T_{MF}$ in zero field. Now, as the magnetic field increases $M$ increases towards saturation due to domain alignment (MST and AST phases are ferromagnetic in nature) and $dM/dH$ decreases. Further increase in magnetic field leads to the transformation of the initial MST phase to the AST phase and as a result $M$ as well as $dM/dH$ increases (it has been discussed earlier that the AST phase has higher $M$ value than the MST phase). At a certain magnetic field, the rate of the MST to AST phase transformation is maximum. This gives rise to a peak in $dM/dH$, and this magnetic field value is marked as $H_{PI}$. At sufficient high magnetic field, the MST to AST phase transition is complete and $M(H)$ reaches saturation. Above this field value, $M(H)$ and $dM/dH$ for the increasing magnetic field cycle are indistinguishable from those for the decreasing magnetic field cycle. This field value is marked as $H^*$ and is the limit of metastability (superheating [5.18]) of the MST phase at this temperature. As the field is now decreased, the AST phase is stable up to a field marked as $H_M$ where the start of the AST to MST phase transition is accompanied by a decrease in $M$ and a corresponding increase in $dM/dH$. At certain lower field value, the rate of transformation from the AST to MST phase is maximum. This produces a peak in $dM/dH$ and this field value is marked as $H_{PD}$. As the field decreases to zero we are back in the phase coexistence regime because the AST to MST phase transition is not completed in zero field at $T > 170K$ (this temperature has been estimated from low field $M$-$T$ results; see Fig. 3.4(f) of Chapter 3). It may be observed that Fig. 5.4(a) or 5.4(b) cannot give the estimate of the magnetic field $H_A$ corresponding to the start of MST to AST phase transition with increasing magnetic field because in zero field, in the beginning of the field increasing experiment, we are already in the phase coexistence region (see Fig. 3.4(f) of Chapter 3). Similarly at the end of the field decreasing experiment, in zero field, the sample is again in the phase coexistence region and we cannot reach the magnetic field $H^*$, the limit of
metastability (supercooling [5.18]) of the AST phase. $H_A$ and $H^*$ can be estimated from the $M(H)$ curves for temperatures below 170 K where only the MST phase exits in zero field. However, the characteristic magnetic fields $H_M$, $H^*$, $H_A$ and $H^{**}$ can also be estimated from the $M(T)$ results obtained in various constant applied magnetic fields as the curves $H_M(T)$, $H^*(T)$, $H_A(T)$ and $H^{**}(T)$ should be identical with the curves $T_{MS}(H)$, $T_{MF}(H)$, $T_{AS}(H)$ and $T_{AF}(H)$ respectively. The temperature dependence of the characteristic magnetic fields $H_M$, $H^*$, $H_A$ and $H^{**}$, or the magnetic field dependence of the characteristic temperatures $T_{MS}$, $T_{MF}$, $T_{AS}$ and $T_{AF}$, constitute the $H$-$T$ phase diagram of the alloy. In Fig. 5.4(c) we present the temperature dependence of the characteristic field values of the first-order phase transition in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy, determined from the isothermal $M(H)$ curves and the isofield $M(T)$ results. It is observed in Fig. 5.4(c) that while the $H_{PI}$ curve is closer to $H^{**}$ as compared to the $H_A$ curve, the $H_{PD}$ curve lies closer to $H_M$ as compared to $H^*$. This is related to the shape of $M(H)$ curves with increasing and decreasing magnetic fields across the transition (see Fig. 5.2).

We note in Fig. 5.4(c) that $H_{PI}$ and $H_{PD}$ have approximately linear $T$ dependence. Earlier, while analyzing the magnetic field induced first-order magnetic transitions with the help of the negative slope of the $H/M$ vs. $M^2$ curves [5.2], the characteristic magnetic field $H_{PI}$ was predicted to increase with temperature [5.8]. Such temperature dependence of the characteristic magnetic fields was observed in the first-order paramagnetic-ferromagnetic phase transitions in various systems like manganites [5.8], Ni-Mn-Ga [5.9], MnAs [5.19], Gd$_5$Si$_{1.7}$Ge$_{2.3}$[5.20] and also in the ferromagnetic-ferromagnetic phase transition in Ni-Mn-Ga [5.9]. But $H_{PI}$ as well as all the other characteristic magnetic fields of Ni$_{50}$Mn$_{34}$In$_{16}$ alloy exhibit a totally different $T$ dependence, e.g. they have a negative slope. The negative slope of the characteristic magnetic field in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy is related to the fact that transition temperature in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy shifts
towards lower $T$ with increasing magnetic field. Similar $T$ dependence of the characteristic magnetic fields has been observed across the antiferromagnetic-ferromagnetic transition in Ru doped CeFe$_2$ alloys [5.21].

5.1.5 Analysis of the slope of the $H/M$ vs. $M^2$ plots and the temperature dependence of the characteristic magnetic fields

The possible explanation for the above observations in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy is as follows. It is worthwhile to note that the criterion of negative slope in the $H/M$ vs. $M^2$ isotherms was inferred from the theory of first-order magnetic phase transitions, where spontaneous magnetization is the appropriate order parameter. In the case of Ni-Mn-Ga [5.9], spontaneous magnetization is the relevant order parameter for the first-order paramagnetic-ferromagnetic transition. But spontaneous magnetization is not the suitable order parameter for the first-order transition from one ferromagnetic to another ferromagnetic phase in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy. However, irrespective of this difference, an ideal first-order magnetic transition should exhibit a discontinuity in magnetization [5.22]. If $M$ increases discontinuously (or very sharply) across the transition with increasing $H$, the slope of $H/M$ vs. $M^2$ curves would become negative. In the case of a disorder broadened first-order transition this discontinuity smears out [5.13]. The present experimental data and similar results on Ni-Mn-Ga [5.9] suggest that even in the case of broadened first-order transition the increase of $M$ with increasing $H$ becomes relatively faster in the transition region. This results in a decrease in the slope of the $H/M$ vs. $M^2$ curves, and the decrease may or may not lead to a negative slope in the $H/M$ vs. $M^2$ isotherm. In Ni$_{50}$Mn$_{34}$In$_{16}$, starting from a temperature (300 K in the protocol adopted for measurement of isothermal $M$-$H$ curves) well above the martensitic transition region, as the temperature is decreased the phase
fraction of MST (AST) phase increases (decreases) from 0 to 1 (1 to 0) as the temperature decreases from \( T_{\text{MS}} \) to \( T_{\text{MF}} \) [5.17]. This MST phase transforms back to AST phase under the application of magnetic field. The increase in \( M \) with increasing \( H \) because of this magnetic field induced phase transition would depend upon the amount of initial MST phase (at \( H = 0 \)) as well as the difference in \( M \) between the MST and AST phases. We have noted earlier that the MST and AST phases in the Ni\(_{50}\)Mn\(_{34}\)In\(_{16}\) alloy have a large difference in \( M \). Also the amount of MST phase formed, in the temperature region \( T_{\text{MF}} - T_{\text{MS}} \), decreases with increasing temperature (see Fig. 5.4(c)). At lower temperatures (within the \( T \) range \( T_{\text{MF}} - T_{\text{MS}} \)), the initial amount of MST phase formed is larger and the magnetic field induced MST to AST phase transition gives larger change in \( M \) with increasing \( H \) and this effectively makes the slope of \( H/M \) vs. \( M^2 \) isotherm negative. Closer is the temperature to \( T_{\text{MS}} \), relatively smaller is the amount of MST phase formed initially. With smaller initial MST phase fraction, the magnetic field induced phase transition becomes less effective in changing the slope of the \( H/M \) vs. \( M^2 \) isotherms. Thus the absence of a negative slope in \( H/M \) vs. \( M^2 \) isotherms in the temperature regime where isothermal \( M(H) \) curves depict clear signatures of a first-order magnetic transition, is correlated with the disorder broadened nature of the first-order phase transition.

Now we investigate the possible cause of negative slope in temperature dependence of the characteristic magnetic fields \( H_M, H_{\text{PD}}, H^*, H_A, H_{\text{PI}} \) and \( H^{**} \) in Ni\(_{50}\)Mn\(_{34}\)In\(_{16}\). In systems where the characteristic magnetic field increases with temperature [5.8, 5.9, 5.19, 5.20], the higher temperature phase has a lower magnetization value. On the other hand in Ni\(_{50}\)Mn\(_{34}\)In\(_{16}\) the lower temperature phase (MST) has a lower magnetization. Similarly in the case of the first-order antiferromagnetic-ferromagnetic phase transition in Ru doped CeFe\(_2\) [5.3, 5.21, 5.23] the lower temperature antiferromagnetic phase has a lower magnetization, and therefore the
characteristic magnetic fields have a negative slope in the $T$ dependence. In fact these results are in accord with the Clausius-Clapeyron equation for a first-order magnetic transition [5.19]:

$$\left( \frac{dT}{dH} \right)_P = -\frac{T\Delta M}{L_Q} \quad (5.1)$$

Here $L_Q = T\Delta S$, $\Delta S$ being the entropy change during the transition. Applying eqn. (5.1) in the case of a magnetic field induced first-order magnetic transition we find that if the higher (lower) temperature phase has higher (lower) magnetization value, then the transition temperature will decrease with increasing magnetic field, and as a result the characteristic magnetic fields for the isothermal transition will have negative temperature dependence. Moreover, eqn. (5.1) gives only single value of $dT/dH$, while from Fig. 5.4 it is clear that the various characteristic temperatures of MST-AST phase transition in present alloy have different values of $dT/dH$. From present work it appears that in the case of the disorder broadened first-order phase transitions, though the Clausius–Clapeyron equation predicts the nature of the slope of the phase transition lines, an exact quantitative result is rather difficult to obtain.

5.2 Imaging the magnetic field induced martensite-austenite phase transition in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy

In the previous sections it has been discussed that the MST-AST phase transition in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy is a disorder-influenced first-order phase transition. Thermomagnetic hysteresis is a generic feature of a disorder-influenced first-order phase transition and has been observed in various classes of magnetic systems undergoing a first-order magneto-structural transition [5.10]. Scanning Hall probe imaging experiments have been very useful in the
investigation of such magnetic systems, revealing clear phase coexistence and metastability across the first-order phase transition [5.24, 5.25]. To get the visual evidence of phase coexistence across the MST-AST phase transition in \( \text{Ni}_{50}\text{Mn}_{34}\text{In}_{16} \) alloy implying that the transition is indeed disorder broadened first-order phase transition in nature, we have performed scanning Hall probe imaging of the magnetic field induced MST–AST phase transition in \( \text{Ni}_{50}\text{Mn}_{34}\text{In}_{16} \) alloy.

### 5.2.1 Scanning Hall probe imaging study of the magnetic field induced phase transition

We have chosen 236 K as the most appropriate temperature to capture images of the phase transition as this temperature is within the MST-AST phase coexistence regime (see Fig. 5.5 which is a part of H-T phase diagram of Fig. 5.4(c) and is presented separately for clarity). Also, the maximum magnetic field that can be applied in our scanning Hall probe imaging experimental set-up is 40 kOe. At 236 K, the magnetic field induced transition is nearly completed in 40 kOe magnetic field. Two different protocols were adopted to reach the temperature 236 K:

(i) In the protocol P1, the sample was cooled from 300 K in zero magnetic field down to 236 K without undershooting the target temperature. The scanning Hall probe imaging experiments were performed at constant temperature first in the increasing magnetic field cycle (up to 40 kOe) and then in the decreasing magnetic field cycle (down to zero).

(ii) In the protocol P2, the sample was cooled in zero magnetic field from 300 K to 30 K. Then the sample was warmed up to 236 K without overshooting the target temperature, and subsequently the scanning Hall probe imaging was performed at constant temperature in the increasing and decreasing magnetic field cycles.
Fig. 5.5: The magnetic field ($H$)–temperature ($T$) phase diagram of the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy. The dotted line in the $H$–$T$ phase diagram depicts the path on which the scanning Hall probe imaging experiments were performed in the increasing and decreasing magnetic field cycles at 236 K. The single arrowhead shows the increasing magnetic field path and the double arrowhead shows the decreasing magnetic field path.

We have already seen that the magnetization of the MST phase is lower than that of the AST phase (see Fig. 5.1). To differentiate between the MST and AST phases in Hall voltage profile, we have chosen a voltage threshold such that the voltages below this threshold represent the MST phase, and the voltages higher than this threshold represent the AST phase. Since the magnetic field induced MST to AST phase transition is nearly complete in 40 kOe magnetic field at 236 K (see Fig. 5.5), we have taken the whole sample to be in the AST phase in 40 kOe field at this temperature. We have taken the Hall voltage threshold at 50% of the maximum in Hall voltage profile in 40 kOe magnetic field. The same voltage threshold has been used in both the protocols P1 and P2. The colour figures thus obtained in both the protocols in the increasing ($H\uparrow$) and decreasing magnetic field ($H\downarrow$) cycles are shown in Fig. 5.6. Here, the yellow regions represent the AST phase, and the black regions represent the MST phase. The field of view for imaging was larger than the sample size. Once the background (due to the applied magnetic
field) was removed, this also produced a black region around the periphery of the sample which simply defines the edge of the sample.

As the magnetic field is increased from zero to 10 kOe at 236 K, some portions of the images show yellow colour indicating that the MST to AST phase transition has started in these regions of the sample. From Fig. 5.6 it is clearly seen that the amount of the AST phase in \( H = 10 \) kOe is larger in protocol P1. With increasing magnetic field, newer yellow patches appear and the existing yellow patches grow in area indicating the nucleation and growth of the AST phase. Finally the entire sample is yellow in 40 kOe magnetic field for both the protocols P1 and P2. However, the nucleation and growth kinetics of the MST to AST phase transformation is slower.
in the protocol P2 for magnetic fields up to 25 kOe. Above this field, the MST to AST phase transformation is much faster in the protocol P2 as compared to the protocol P1. In the decreasing field cycle, the MST phase appears as black spots and the nucleation and growth of the MST phase is clearly seen in the lower panel of Fig. 5.6. The nucleation and growth kinetics of the AST to MST phase transition in both the protocols P1 and P2 appear to be quite similar as the field is decreased from 40 kOe to zero. The transition becomes quite slow in the low fields and some AST phase remains un-transformed in fields down to 5 kOe in both the protocols. Thus Fig. 5.6 actually shows the snap-shots of the coexistence of MST and AST phases on the length scale of tens of micrometers, extended over a very wide regime of magnetic field. Comparison of the top and bottom panels of Fig. 5.6 also confirms the magnetic field hysteresis associated with this magnetic field induced phase transition (for example compare the Hall images taken in 20 kOe magnetic field, in the increasing and decreasing magnetic field cycles). It is also observed in Fig. 5.6 that the region of sample where the MST to AST phase transition starts first (last) in increasing magnetic field cycle, is also the region where AST to MST phase transition starts at last (first) in decreasing magnetic field cycle. This shows that the hysteresis associated with the magnetic field induced MST-AST phase transition is influenced by microscopic characteristics (disorder, strain, etc.) specific to particular microscopic regions of the sample.

To investigate further into the magnetic field induced MST–AST phase transition, we estimate the volume fractions of the AST and MST phases as a function of magnetic field in both the protocols. In Fig. 5.6, the area of the yellow region in the images is a measure of the volume of the AST phase (VF\textsubscript{AST}). To calculate VF\textsubscript{AST}, we normalize the area of yellow region in an image with the area of the yellow region in the scanning Hall probe image obtained in 40 kOe magnetic field. The volume fraction of the MST phase (VF\textsubscript{MST}) is related to the VF\textsubscript{AST} through
the relation: $\text{VF}_{\text{AST}}(\%) + \text{VF}_{\text{MST}}(\%) = 100$. The volume fraction thus calculated is not an exact quantitative estimate since the MST and AST micro-regions beneath the surface of the sample also contribute to the Hall voltage profile in varying amounts depending on their depth. Hence the $\text{VF}_{\text{AST}}$ estimated from Hall images is only representative of the actual phase distribution in the bulk sample. The contribution from the MST/AST regions to Hall voltage profile also depends on their length scale relative to the resolution of Hall probe. If the length scale of MST-AST phase coexistence is smaller than the resolution of scanning Hall probe (nearly 10 $\mu$m in present case), the probe will sense effective magnetization averaged over a pixel. This also contributes to the uncertainty in the estimated $\text{VF}_{\text{AST}}$. Moreover, at low magnetic fields the local magnetization may not be fully aligned with the field due to anisotropy and/or domain wall pinning, and our scanning Hall probe senses the component of local magnetic induction perpendicular to the sample surface. Nevertheless we are able to make qualitative measure of the behaviour across this magnetic field induced transition.

The magnetic field dependence of the volume fractions of AST and MST phases is presented in Fig. 5.7. As indicated by Hall images in 10 kOe magnetic field in the increasing magnetic field cycle, the amount of AST phase is larger in protocol P1. The difference in the kinetics of the MST to AST phase transition in the protocols P1 and P2 with increasing magnetic field is clearly seen in Fig. 5.7. With decreasing magnetic field there is very little change in $\text{VF}_{\text{AST}}$ till 20 kOe, and below this magnetic field the rate of change of $\text{VF}_{\text{AST}}$ is enhanced appreciably. The magnetic field dependence of $\text{VF}_{\text{AST}}$ with decreasing magnetic field for the two protocols P1 and P2 are qualitatively similar, which is in contrast with the increasing field cycle discussed above.
We will see in the next section (section 5.3) that the $M(H)$ behaviour of Ni$_{50}$Mn$_{34}$In$_{16}$ across the AST-MST phase transition and the functional properties exhibited by this alloy sample across this phase transition are strongly dependent on the experimental protocol. These observations suggest that the kinetics of the magnetic field induced MST to AST phase transition in the Ni$_{50}$Mn$_{34}$In$_{16}$ alloy is affected by the thermomagnetic history of the sample. The protocols P1 and P2 were adopted to investigate, the effects of such thermomagnetic history on the magnetic field induced transition in this alloy. In the protocol P2, 236 K is reached after crossing the $T_{AS}$ or $H_A$ line. As this line is crossed, the MST to AST phase transformation is initiated. At 236 K, the AST phase is the equilibrium configuration at $H = 0$, and the MST phase is metastable (superheated) [5.18]. As the field is raised above 30 kOe under the protocol P2, the system goes very close to the limit of superheating [5.18] $T^{**}$ or $H^{**}$. The rate of the MST to AST transformation is very high in this field regime and this is in harmony with our earlier finding.
that the rate of a first-order magnetic phase transformation increases rapidly when the system
goes close to the spinodal limit [5.16, 5.26]. This phenomenon has earlier been observed across
the first-order antiferromagnetic-ferromagnetic phase transition in 4% Ru doped CeFe$_2$ alloy
[5.16] and across a magnetic field induced first-order phase transition in vortex matter [5.26]. In
the protocol P1, on the other hand, 236 K is reached after crossing the $T_{\text{MS}}$ or $H_{\text{M}}$ line. At $H = 0$
and $T = 236$ K in the protocol P1, part of the sample is in the equilibrium MST phase and the
remaining part is in the supercooled (metastable) [5.18] AST phase. As the field is increased,
however, the situation is changed altogether. The system now moves towards the limit of
superheating [5.18] of the MST phase. In this route the AST phase is the equilibrium state, and
the MST phase is metastable. The field increasing path in protocol P1 causes a kind of reversal
of the kinetics of the phase transition, and thus probably introduces a large disturbance in the
system. This kinetics is different from that of protocol P2, and accordingly, the behaviour of the
system in protocol P1 is different from that of P2 in the field increasing cycle. In both the
protocols P1 and P2, the system is very close to the limit of superheating ($T^{*\ast}$ or $H^{*\ast}$) at $H = 40$
kOe. When the field is decreased from 40 kOe, there is no path difference (in the H-T phase
space) between the two protocols and accordingly the kinetics of the phase transition is similar in
both the protocols (Figs. 5.6 and 5.7). The AST to MST phase transition is initiated below 20
kOe when the system crosses the $T_{\text{MS}}$ or $H_{\text{M}}$ line in both the protocols. The present scanning Hall
probe microscopy images suggests that the functional properties of the alloy will depend on the
protocol used for changing the field and temperature because of the relative volume fractions of
the MST and AST phases present in the alloy. It is clear from Fig. 5.6 and 5.7 that these volume
fractions are strongly dependent on the path traversed by the system in the $H$-$T$ phase space.
It is observed in Fig. 5.7 that the magnetic field dependence of volume fractions of the AST and MST phases exhibits a distinct magnetic hysteresis with increasing and decreasing magnetic fields. To compare it with the hysteresis observed in the magnetic field dependence of global magnetization, $V_{\text{F}_{\text{AST}}}$ was plotted along with the normalised $M(H)$ curve obtained in the protocol P1 at $T = 236$ K. The $V_{\text{F}_{\text{AST}}}(H)$ curve is qualitatively similar to the $M(H)$ curve (see Fig. 5.8 for protocol P1; magnetization is normalized with the magnetization at 40 kOe magnetic field). The data for the protocol P2 is not presented here for conciseness. The observation that $V_{\text{F}_{\text{AST}}}$ and normalized M have similar magnetic field dependence confirms that the width of the magnetic field induced MST–AST phase transition observed in the global field dependence of magnetization (and other bulk observables) is due to the nucleation and growth kinetics of this disorder broadened phase transition. It also confirms that the defined threshold voltage used to construct the scanning Hall probe images is realistic.
5.2.2 Nucleation and growth mechanism of the martensite-austenite phase transition in the Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy

We further attempt a quantitative analysis of the evolution of volume fraction of the AST phase as a function of magnetic field. The “S” shape of the magnetic field dependence of volume fraction of the AST phase in Fig. 5.7 resembles the transformation-time curve predicted by Avrami model [5.27] proposed for crystallization of solids. The central assumption of the Avrami model is that the product phase is nucleated by the germ nuclei which are already present in the parent phase. The density of these germ nuclei decreases in two ways. In the first way some of them become active growth nuclei of the product phase. The second way is the swallowing of other germ nuclei by these growth nuclei in the process of growth. The nucleation events are taken to be totally random. The time dependence of phase fraction \((f)\) of the product phase is given by the Kolmogorov–Johnson–Mehl–Avrami (KJMA) relation

\[
\begin{align*}
    f &= 1 - \exp\left(-k t^n\right) \\
    \text{(5.2)}
\end{align*}
\]

where \(k\) is related to the activation energy and the Avrami exponent \(n\) depends on geometrical factors. Originally the model was proposed for isothermal time dependence of evolution of product phase in crystallization from liquid. The model has been extended to temperature and magnetic field dependence of the phase fraction of the product phase for first-order phase transition in doped Fe-Rh alloy [5.28] and following relationship was found suitable for description of the transition.

\[
\begin{align*}
    f &= 1 - \exp\left(-k\left(T - T_0\right)^n\right) \\
    \text{(5.3)}
\end{align*}
\]

where for temperature driven transition \(T_0\) is the onset temperature of transition. Similarly, for a magnetic field driven transition the observable \(T\) in equation (5.3) can be replaced by \(H\) [5.28]. We now check whether the KJMA behaviour is observed in the magnetic field dependence of
volume fraction of the AST phase (here the volume fraction is used as the phase fraction). In the original Avrami model or in that used in the Ref. 5.28, there is an onset point of transition i.e. a value of control variable (time/temperature/magnetic field) up to which the phase fraction of product phase is zero. However in the present case even at the minimum value of magnetic field the volume fraction of product AST phase is non-zero as the sample is already in the phase coexistence region at $T = 236$ K. So we can not apply the Avrami model directly. We have modified the KJMA relation so as to have a finite value of volume fraction of the product phase at zero magnetic field. The modified KJMA equation is:

$$f = 1 - A_1 \exp\left(-kH^n\right)$$  \hspace{1cm} (5.4)

Fig. 5.9: Fitting of the magnetic field ($H$) dependence of the volume fraction of the austenite phase ($VF_{AST}$) across the H induced martensite–austenite phase transition at 236 K in protocol P1, using equation (5.4).
where the factor $A_1$ is included to allow finite non-zero value of phase fraction of AST phase in zero magnetic field. The fitting of equation (5.4) with the magnetic field dependence of $\text{VF}_{\text{AST}}$ in protocol PI is presented in Fig. 5.9. Clearly, equation (5.4) fits well with the magnetic field dependence of the volume fraction of the AST phase. The values of $A_1$ and $n$ (obtained as fitting parameters) on the increasing field cycle are 0.767 and 6.018 respectively. Similarly the values of $A_1$ and $n$ obtained on the decreasing field cycle are 0.920 and 2.730 respectively. The value of $n$ in decreasing field cycle is comparable to that obtained for the structural transition in doped Fe-Rh alloy in increasing/decreasing field cycle [5.28], but the value of $n$ in increasing field cycle is larger as compared to that in doped Fe-Rh alloy. Recently a similar large value of $n$ (exponent of the control variable) in a KJMA type fit was reported for the temperature driven structural transition in uranium [5.29]. In the present case, the very different values of $n$ in increasing and decreasing field cycle are due to the different shapes of the $\text{VF}_{\text{AST}}(H)$ curves in increasing and decreasing magnetic field cycles. This may be related with the intrinsic asymmetry between supercooling and superheating in the first-order phase transitions [5.16]. The observation that the volume fraction of AST phase across the magnetic field induced phase transition in the present alloy can be fitted with a KJMA type relation confirms a nucleation and growth mechanism in this magnetic field induced phase transition.

5.2.3 Landscape of transition field across the magnetic field induced martensite-austenite phase transition

We now analyze the Hall images further to explore the local magnetic behaviour of the sample across the magnetic field driven MST-AST phase transition. This is done by tracking the Hall voltage signal coming from the individual pixels which corresponds to $10 \mu\text{m} \times 10 \mu\text{m}$ sample area approximately. The magnetic field dependence of such a voltage signal produces a
Fig. 5.10: (a) Local Hall voltage vs. magnetic field \((H)\) loops at representative sites \((L_1, L_2,\) and \(L_3)\) in Hall images at 236 K in protocol P1. The Hall voltage is normalized with its value in a magnetic field of 40 kOe at site \(L_1\). (b) Comparison of the \(H\) dependence of bulk magnetization \((M)\) measured using the VSM with the integrated moment calculated from Hall images at 236 K in protocol P1. The integrated moment is normalized with the value of \(M\) in a magnetic field of 40 kOe.

local Hall voltage loop. Fig. 5.10(a) presents the local Hall voltage loops corresponding to three representative sites (pixels, named as \(L_1, L_2,\) and \(L_3)\) on the sample. The Hall voltages in the local loops are normalized to the Hall voltage at the site \(L_1\) in 40 kOe magnetic field. It is interesting to observe in Fig. 5.10(a) that while the magnetic field induced MST to AST phase transition is completed well below 40 kOe at the site \(L_1\), the transition is probably not completed
at the site \( L_2 \) at \( H = 40 \) kOe. On the other hand, the transition is very near to completion at the site \( L_3 \) in the same field value. Further it can be seen that among the three chosen sites, the MST to AST phase transition is completed first in increasing magnetic field cycle for the site \( L_1 \). But the AST to MST transition at this site starts at the last in the decreasing magnetic field cycle. This sequence is just the reverse for the site \( L_3 \). The characteristics exhibited by the local Hall voltage loops indicate the presence of a landscape of critical magnetic field for MST-AST phase transition across the sample. This kind of disorder induced landscape of transition temperatures or magnetic fields had been envisaged earlier theoretically by Imry and Wortis [5.13], and observed experimentally in vortex matter [5.30] and various magnetic systems [5.24, 5.25]. Thus it confirms the disorder-influenced nature of this magnetic field induced MST-AST phase transition in \( \text{Ni}_{50}\text{Mn}_{34}\text{In}_{16} \).

The integrated moment is the summation of the Hall voltage signals from all the pixels across the sample. Fig. 5.10(b) shows the comparison of the bulk magnetization measured in the vibrating sample magnetometer (VSM) and the integrated moments calculated from the Hall images. Though the integrated moments calculated from the Hall images follow the bulk magnetization behaviour, there is a visible difference between the two curves. We attribute this difference to the demagnetization factor [5.31]. While the sample used for the measurements in the VSM is distorted needle shaped of length approximately 1 mm, the sample used for scanning Hall probe imaging experiments is of the shape of a distorted circular plate of diameter approximately 2 mm. The shape of the field dependence of the integrated moments seems to be somewhat different from the individual local loops.
5.3 Thermomagnetic history dependence of the functional properties

We have seen that the interesting functional properties observed in Ni₅₀Mn₃₄In₁₆ alloy arise from the first-order MST-AST phase transition. In the previous discussion it is established that the MST-AST phase transition in Ni₅₀Mn₃₄In₁₆ alloy is a disorder-influenced first-order phase transition. Thermomagnetic history dependence of the volume fractions of AST and MST phases has been verified from Hall images of the sample across the transition. The functionalities in the alloy obviously are expected to depend on the volume fractions and so are expected to be dependent on the thermomagnetic history of the sample. Such history dependence is explored in the present section.

5.3.1 Thermomagnetic history dependence of magnetoresistance in the Ni₅₀Mn₃₄In₁₆ alloy

Fig. 5.11 shows isothermal magnetic field dependence of resistivity ($\rho$) of the Ni₅₀Mn₃₄In₁₆ alloy at 250 K measured in two protocols. In the first protocol the sample was cooled monotonically from 300 K, without undershooting, in zero field down to the target temperature. In the second protocol the sample was first cooled from 300 K in zero field down to 5 K. It was then warmed up monotonically without overshooting to the target temperature (in zero field). These protocols are equivalent to the protocols P1 and P2 respectively discussed in section 5.2.1.

In both the protocols the MR is negative. However, there is a substantial difference between the MR measured in the two protocols. In first protocol the MR is comparatively smaller and totally reversible. On the other hand MR is not only relatively larger but also irreversible in the second protocol. It is clear that the magnitude as well as the reversibility of MR depend on the history of the sample.
5.3.2 Thermomagnetic history dependence of magnetocaloric effect in the Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy

We have estimated isothermal magnetic entropy change ($\Delta S_M$) in Ni\textsubscript{50}Mn\textsubscript{34}In\textsubscript{16} alloy using isothermal M(H) curves measured in three different protocols P1, P2 and P3. The $M(H)$ thus measured at 242 K is shown in Fig. 5.12(a) for example. In P1 the sample is first zero field cooled to 150 K and then gradually warmed up to 242 K; in P2 protocol $T = 242$ K is reached by zero field cooling the sample from 305 K; in P3 the sample is first cooled down to 30 K before warming up to 242 K.

The magnitude of $\Delta S_M$ as a function of temperature estimated for a magnetic field excursion from zero to $H_{\text{max}} = 80$ kOe is shown in Fig. 5.12(b). It is clearly seen that the maximum obtainable value of MCE depends on the protocol of the measurement. The highest
value of $\Delta S_M = 24$ J/kg-K is obtained under protocol P1. We have also calculated the effective refrigerant capacity ($R_{\text{EFF}}$) for the three protocols. Table 5.1 summarizes the results concerning the refrigerant capacity obtained under the three different experimental protocols P1, P2, and P3. We find that the $R_{\text{EFF}}$ obtained under the protocol P2 has the largest value of 226 J/kg.

![Diagram](image)

**Fig. 5.12:** (a) Isothermal magnetization ($M$) vs. magnetic field ($H$) plots at $T = 242$ K in Ni$_{50}$Mn$_{34}$In$_{16}$, measured under three different experimental protocols P1, P2, and P3. (b) Isothermal change in magnetic entropy ($\Delta S_M$) as a function of temperature ($T$) across the austenite-martensite phase transition in Ni$_{50}$Mn$_{34}$In$_{16}$, measured under these three protocols.

**Table 5.1:** Useful temperature range, refrigerant capacity, average hysteresis loss, and effective refrigerant capacity $R_{\text{EFF}}$ estimated for Ni$_{50}$Mn$_{34}$In$_{16}$ measured under the three different protocols.

<table>
<thead>
<tr>
<th>Experimental protocol</th>
<th>$T_{\text{cold}}$ (K)</th>
<th>$T_{\text{hot}}$ (K)</th>
<th>Refrigerant capacity (J/kg)</th>
<th>Average hysteresis loss (J/kg)</th>
<th>Effective refrigerant capacity $R_{\text{EFF}}$ (J/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1</td>
<td>233.6</td>
<td>244.1</td>
<td>163</td>
<td>103</td>
<td>60</td>
</tr>
<tr>
<td>P2</td>
<td>208.6</td>
<td>240</td>
<td>367</td>
<td>141</td>
<td>226</td>
</tr>
<tr>
<td>P3</td>
<td>230.7</td>
<td>247.4</td>
<td>156</td>
<td>119</td>
<td>37</td>
</tr>
</tbody>
</table>
5.3.3 Thermomagnetic history dependence of the magnetic field induced strain in the \( \text{Ni}_{50}\text{Mn}_{34}\text{In}_{16} \) alloy

We have measured the magnetic field induced strain in the temperature regime of the MST–AST phase transition using two different experimental protocols. For example, the isothermal magnetic field dependence of relative length change \( \Delta L/L \) measured at 234 K, is presented in Fig. 5.13. In protocol P1 the sample was cooled from 300 K in a zero magnetic field without undershooting the target temperature 234 K. The subsequent isothermal \( \Delta L/L(H) \) measurements were performed at 234 K by increasing the magnetic field and decreasing it back to zero (see Fig 5.13(a)). In protocol P2 the sample was first cooled to 50 K in a zero magnetic field before warming it up to 234 K without overshotting the target temperature. The isothermal \( \Delta L/L(H) \) measurements were performed at this temperature (see Fig 5.13(b)). With the increase in the magnetic field, in both the protocols \( \Delta L/L(H) \) shows a sharp rise close to 20 kOe and then
saturates around the 40 kOe magnetic field. However, before reaching saturation $\Delta L/L(H)$ shows a hump like feature similar to that seen in the temperature induced transition (see Fig. 4.8 of Chapter 4). Further the complete $H$ cycle reveals a large magnetic field hysteresis in the $\Delta L/L(H)$ curve in both the protocols. All these features in isothermal $\Delta L/L(H)$ are related to the magnetic field induced transition from the MST to the AST phase. The magnetic field induced strain is larger in protocol P2. In decreasing the magnetic field cycle $\Delta L/L(H)$ remains nearly constant down to $H \approx 20$ kOe and decreases after exhibiting the hump like feature (Figs. 5.13(a) and 5.13(b)). Further, on reducing the magnetic field isothermally from the state with saturated $H$-induced strain, the zero-field strain value is completely recoverable in protocol P1 while it is only partially recoverable in protocol P2. Fig. 5.13(c) presents isothermal $\Delta L/L(H)$ curves in protocol P2, at various temperatures within the temperature regime of MST–AST phase transition. At all these temperatures, $\Delta L/L(H)$ exhibits features of magnetic field induced transition similar to that at 234 K, i.e. it shows a sharp rise at certain critical magnetic field and exhibits a field hysteresis. The magnetic field value, at which a sharp rise in $\Delta L/L(H)$ occurs, increases with the decrease in temperature. Further, as the temperature decreases, the residual strain in zero magnetic field decreases and at some temperature the magnetic field induced strain can be totally recoverable (with zero residual value) even in protocol P2 (see the $\Delta L/L(H)$ curve at $T = 222$ K in Fig. 5.13(c)). The observations in Figs. 5.13(a)–(c) signify that in the present alloy the $H$-induced strain can be recoverable or non-recoverable depending on the temperature and the experimental protocol.

As mentioned earlier thermomagnetic history dependence of the functional properties of Ni$_{50}$Mn$_{34}$In$_{16}$ alloy arises from the history dependence of the AST and MST volume fractions. In the temperature regime of phase-coexistence, these volume fractions depend on the path
followed in $H$-$T$ phase space to reach a specific point $(H,T)$. For example, in Fig. 5.7 $VF_{\text{AST}}$ at $(236 \text{ K}, H = 0)$ at start in protocol P2 not similar to that in P1. Application of field of 40 kOe causes a larger change in $VF_{\text{AST}}$ in protocol P2. Further, in the $H$-$T$ phase diagram (in Fig. 5.5) increasing temperature and increasing field paths are equivalent as both cause an MST to AST phase transition. Also a decreasing field path is equivalent to a decreasing temperature path. The point $(236 \text{ K}, H = 0)$ at the start and the point $(236 \text{ K}, H = 0)$ after field cycling are expected to have similar $VF_{\text{AST}}$ in protocol P1 but not in P2. Hence, while in protocol P1 no hysteresis is expected, P2 is expected to have a field hysteresis as has been observed in isothermal magnetic field dependence of resistivity (Fig. 5.11(b)) and strain (Fig. 5.13(b)).

5.4 Conclusion

Summarizing, the isothermal $M$ vs. $H$ curves of Ni$_{50}$Mn$_{34}$In$_{16}$ show the signatures of a magnetic field-induced first-order martensite to austenite phase transition in the temperature range 200–250 K. The isothermal $H/M$ vs. $M^2$ curves, however, do not exhibit a negative slope at certain temperatures within this temperature range. This suggests that the criterion of relating the negative slope of the $H/M$ vs. $M^2$ isotherms to the first-order magnetic transitions is not applicable for all the magnetic transitions. On the other hand, a decrease in slope of the $H/M$ vs. $M^2$ isotherm is found to characterize the first-order magnetic transition in the present case. Such a decrease in slope might lead to a negative slope of the $H/M$ vs. $M^2$ isotherms if there is a large change of magnetization because of the magnetic field-induced magnetic phase transition. The isothermal $M(H)$ curves and the temperature dependence of the characteristic fields of the first-order martensite–austenite phase transition can be explained within the framework of disorder broadened first-order transition. The nature of the temperature dependence of the characteristic
magnetic fields is found to follow the Clausius–Clapeyron relation for a first-order phase transition. Further, the scanning Hall probe imaging experiments on Ni\(_{50}\)Mn\(_{34}\)In\(_{16}\) alloy provide clear visual evidence of the coexistence of the martensite and austenite phases across the magnetic field induced martensite–austenite phase transition in this alloy. This highlights the disorder broadened first-order nature of the phase transition. Thermomagnetic history effects are found to play an important role in the evolution of this phase transition. The relative volume fractions of the martensite and austenite phases in the phase coexistence regime are found to depend strongly on the path chosen in the field–temperature phase space for varying the temperature and magnetic field. Quantitative analysis of the Hall images suggests nucleation and growth dynamics of phases across the magnetic field induced phase transition. Also the local Hall voltage loops constructed from the Hall images suggest a landscape of the critical magnetic field of the MST–AST phase transition, thus confirming the disorder-influenced nature of the transition. The magnitude and reversibility of the functional properties of the alloy under application of magnetic field, have been found to depend on the experimental path traversed in the \(H–T\) phase space to prepare the starting point of measurements.

**Publications based on the present Chapter:**

1. Large magnetoresistance in Ni\(_{50}\)Mn\(_{34}\)In\(_{16}\) alloy.


2. Studies on the field-induced martensite to austenite phase transition in Ni\(_{50}\)Mn\(_{34}\)In\(_{16}\).


   **Sharma V. K.,** Chattopadhyay M. K., and Roy S. B.
3. Thermomagnetic history dependence of magnetocaloric effect in Ni$_{50}$Mn$_{34}$In$_{16}$.

   Chattopadhyay M. K., **Sharma V.K.**, and Roy S. B.

4. A scanning Hall probe imaging study of the field induced martensite-austenite phase transition in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy.

   **Sharma V.K.**, Moore J. D., Chattopadhyay M.K., Morrison K., Cohen L. F., and Roy S.B.

5. Temperature and magnetic field induced strain in Ni$_{50}$Mn$_{34}$In$_{16}$ alloy.

   **Sharma V.K.**, Chattopadhyay M.K., Chouhan A., Roy S.B.