Anomalous Magnetic Properties in $Ni_{50}Mn_{35}In_{15}$

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Abstract. We present here a comprehensive investigation of the magnetic ordering in $Ni_{50}Mn_{35}In_{15}$ composition. A concomitant first order martensitic transition and the magnetic ordering occurring in this off-stoichiometric Heusler compound at room temperature signifies the multifunctional character of this magnetic shape memory alloy. Unusual features are observed in the dependence of the magnetization on temperature that can be ascribed to a frustrated magnetic order. It is compelling to ascribe these features to the cluster type description that may arise due to inhomogeneity in the distribution of magnetic atoms. However, evidences are presented from our ac susceptibility, electrical resistivity and dc magnetization studies that there exists a competing ferromagnetic and antiferromagnetic order within crystal structure of this system. We show that excess Mn atoms that substitute the In atoms have a crucial bearing on the magnetic order of this compound. These excess Mn atoms are antiferromagnetically aligned to the other Mn, which explains the peculiar dependence of magnetization on temperature.

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1. Introduction

Ni-Mn based Heusler alloys of the type $Ni_{50}Mn_{50-y}X_y$ (X = In, Sn and Sb) have recently been identified by Sotou *et.al* [1] as systems undergoing martensitic transition in the ferromagnetic state. Such compounds have the tendency to display huge strains with application of moderate magnetic fields. This unique magnetoelastic property has numerous technological implications leading to wide spread applied research like the magnetic shape memory, magnetocaloric and magnetoresisitive effect. These unique properties are an outcome of the *Martensitic Phase Transformation* that takes place in a magnetically ordered state. The classic example of this class of materials is the Heusler alloy, Ni_2MnGa [2, 3, 4, 5].

Martensitic transformation is a first-order structural phase change wherein the constituent atoms get displaced from its crystallographic positions over varied amplitude of displacement in a highly correlated fashion. Upon transformation, the crystal structure changes from a highly symmetric cubic phase to a low symmetry structure. Some ferromagnetic Heusler compounds are known to undergo a martensitic transition that is highly reversible and the compounds can be cycled several times through the transformation temperature. The generic formula for Heusler compounds is X_2YZ. In the high temperature phase, the structure can be viewed as four interpenetrating fcc lattices with X atoms occupying the (0,0,0) and ($\frac{1}{2}$,0,0) sites; Y atoms occupying the ($\frac{1}{4}$,$\frac{1}{4}$,$\frac{1}{4}$) sites and Z atoms occupying the ($\frac{3}{4}$,$\frac{1}{4}$,$\frac{1}{4}$) sites. A tetragonal or orthorhombic structure is observed in the low temperature martensitic phase with the displacement of atoms giving rise to modulations that extend over several crystal planes.

Recently found $Ni_{50}Mn_{50-x}In_x$ compounds that belong to the class of Heusler alloys have been in focus, especially for x having martensitic transition temperatures near room temperature. In particular, the stoichiometric composition $Ni_{50}Mn_{25}In_{25}$ is known to order ferromagnetically at $T_C \sim 315$ K [6], does not undergo a martensitic transition. While, the Mn-rich off-stoichiometric composition $Ni_{50}Mn_{50-x}In_x$ exemplifies a martensitic transition. This transition temperature (T_M) is highly sensitive to the Mn:In ratio and a value between 260 K to 302 K have been reported for In content varying over a small value of 13 to 16% atomic concentration [1, 7]. However, the T_C is not very influenced by the Mn:In ratio and varies only slightly with the composition. A large negative magnetoresistance over 50% is attainable in the Ni-Mn-In compound at moderate field strengths [8]. A giant isothermal entropy change takes place when the structural and the magnetic transition temperatures coincide or are in close vicinity to each other resulting in an inverse magnetocaloric effect (MCE) near room temperature [9, 10, 11]. It may be noted that an *inverse* MCE is generally displayed by materials with antiferromagnetic order and the magnitude obtained is generally quite low. It is therefore fundamentally important to gain thorough insight into the structural and magnetic aspects of the technologically important compound. With this aim, we propose to study the nature of magnetic interactions in the composition $Ni_{50}Mn_{35}In_{15}$ where a concomitant structural and magnetic transition is obtained near room temperature.

The key question to be addressed in the present study is the character of the various magnetic interactions that develop with varying distances between magnetic atoms in the two crystallographic phases. We have carried out the measurement of magnetic and transport properties of $Ni_{50}Mn_{35}In_{15}$ Heusler alloy and the results are presented here.

2. Experimental

$Ni_{50}Mn_{35}In_{15}$ was prepared by arc-melting the constituent elements of 4N purity under argon atmosphere. The button so obtained, was annealed at 1000 K for 48 hours followed by quenching in ice water. Subsequent energy dispersive x-ray (EDX) analysis confirmed the composition to be close to nominal with Ni = 49.26, Mn = 35.13 and In = 15.49. The ac susceptibility (χ_{ac}) and four probe resistivity measurements were performed using Quantum Design Physical Properties Measurement System. While for the dc magnetization measurements the Quantum Design SQUID magnetometers (MPMS-XL and MPMS SQUID-VSM) were employed. The χ_{ac} Vs. temperature was measured in the presence of different dc fields and with the excitation frequency varied over three decades. The sample was cooled to lowest measurement temperature (5 K) in zero field state and the data was recorded while warming up to $\sim$ 330 K. Magnetization as a function of field was measured under sweep magnetic fields up to $\pm$ 5 T at various temperatures. Before each measurement, the sample was heated to 330 K and cooled in zero field state to the desired temperature. For the resistivity measurements the data was recorded in the region 5 K to 380 K in the presence of 5 T and 9 T magnetic fields.

3. Results and Discussion

The temperature dependence of the magnetization from 5 to 380 K, recorded while cooling in the zero field state (nominal field 5 Oe), for $Ni_{50}Mn_{35}In_{15}$ is presented in figure 1. It can be clearly seen that with decreasing temperature the magnetization rises abruptly at the ferromagnetic ordering temperature $T_C = 306$ K. While the abrupt decrease in magnetization occurs at the martensitic phase transformation with $T_M = 302$ K. The inset shows the enlarged view of these magnetic and martensitic transformations. χ_{ac} measured at a frequency $f = 13$ Hz in an ac field of 1 Oe is presented in figure 2. The data shows a very sharp peak at about 300 K which is an outcome of concomitant martensitic and magnetic transformation taking place in the compound.

As mentioned earlier, the martensitic transformation is a first order structural transformation, taking place from a high symmetry cubic phase to a low-symmetry structure. It thus involves a start temperature, when the structure starts deforming and a finish temperature, when the transformation is complete. The variants of the new crystallographic phase that are formed in this region of the start and finish temperatures, re-establish the magnetic interactions that had been present in the parent cubic phase. The difference in anisotropy strongly modifies the field dependence of the magnetization in these two phases [12]. For the present sample, the structure starts deforming at 302

K and the transformation completes by $\sim$ 270 K. However, it is interesting to note that magnetization or χ_{ac} attains an almost zero value after the structural transition is complete. This is an uncommon feature and implies that the magnetic interactions of this compound should be quite complex.

The χ_{ac} measured during the warm-up cycle displays yet another broad peak at $T^* = 170$ K, while the dc magnetization (measured while cooling) shows a constantly increasing magnetization with a hump-like feature at the same temperature. However, the magnetization keeps increasing with the subsequent fall in temperature below T^*. The exact nature of T^* is not clear at the moment. The first report on this family of compounds by reference [1] claims the occurrence of an additional martensite-to-martensite transformation at low temperatures. They observe some intricate changes similar to T^* in their low temperature magnetization data (M(T) with H = 500 Oe) that are taken as signatures for the occurrence of second structural transformation. However, feature similar to T^* observed in the magnetization measurements by reference [7] has been ascribed to the magnetic ordering of the martensitic phase by them. Thus, it becomes essential to verify the nature of T^* and also to investigate the reason for the drastic fall in χ_{ac} to almost zero value upon martensitic transformation. Hence $\chi_{ac}(T)$ was measured in a constant dc field varying from 0 to 500 Oe and the plots are presented in figure 3. With the increase in dc field the peak at T^* broadens and decreases in magnitude. At 500 Oe, the peak smears out completely and a small hump begins to appear at a lower temperature, as can be seen in the inset of figure 3. Such dependence of the peak at T^* on dc magnetic field implies that there exists other competing magnetic interactions in this system that compete with the long range ferromagnetic order. Such complicated behaviour with competing ferromagnetic (FM) and antiferromagnetic (AFM) interactions generally results in a frustrated magnetic order.

A possibility that may give rise to competing FM/ AFM interactions in $Ni_{50}Mn_{35}In_{15}$ is the inhomogeneous distribution of the constituent elements forming regions of varied stoichiometries. Since the amount of In is the least in this compound, regions rich in Ni_2MnIn and NiMn can form. It is well established that Ni_2MnIn is ferromagnetic in nature [2] while NiMn displays antiferromagnetism [13]. In case such a possibility exists in the present compound, the random distribution of such magnetic entities would lead to cluster formation and eventually result in the freezing of the resultant magnetic moment. It is important mention here that the present compound shows robust martensitic phase transformation at room temperature while neither Ni_2MnIn or NiMn display this property. Hence little doubt exists about $Ni_{50}Mn_{35}In_{15}$ lacking in compositional uniformity. Nonetheless, to further investigate the cause for FM/AFM interactions due to possibility of inhomogeneous mixing, χ_{ac} was measured with varying frequencies. Temperature dependence of χ_{ac} at different frequencies f (13 to 1333 Hz) is presented at figure 4. It is evident from this plots that the peak position at T^* does not show a shift in temperature with changing frequencies. This observation rules out the possibility of T^* being a time dependent phenomenon and hence cannot

be related to freezing of the magnetic moment like in a spin-glass state.

The shape of the hysteresis loop generally provides a better understanding of the magnetic ground state of a material. Hence M(H) loops at select temperatures were obtained by heating the sample to the paramagnetic region (350 K) and cooling to the desired temperature in a zero field state. As can be witnessed from figure 5, the M(H) loop at 300 K (and 290 K) displays a very complex behaviour. The initial steep rise in M(H) for small field values demonstrates the ferromagnetic character of the sample. With the increase in field to intermediate values, a metamagnetic transition is observed as depicted in the inset. In the case of ferromagnetic shape memory alloys, M(H) measurements in the martensitic transformation region does display a metamagnetic transition due to reorientation of the already formed martensite variants. However, the type of metamagnetism seen in the present case is quite unusual. Also, there is a considerable increase in the saturation magnetization upon cycling the sample though magnetic field. The mechanism driving the transition between the two metastable states cannot be explained by a simple reorientation of an existing martensite component, but may be connected to the nucleation of an additional magnetic phase. It is rather difficult to attribute the exact cause for the metamagnetic transition. Further, the width of the hysteresis almost goes to zero at H = 0. Such hysteresis have been considered earlier as possible signatures of a field-induced firstorder metamagnetic transition from AFM to FM in cubic laves phase Co-doped $CeFe_2$ alloys [14]. Also, with the drop in temperature, there is a decrease in the overall saturation magnetization seen for the M(H) at 290 K. This observation is in agreement with the $\chi(T)$ plot where the susceptibility decreases with decreasing temperature in this temperature region and starts building up below 240 K with a peak at T^*. Thus M(H) was measured in the vicinity of T^*.

Initially the magnetization rises sharply at small field values indicating a ferromagnetic order. However, it does not saturate at high fields as expected for a typical ferromagnet. Also, interesting features are seen in the low field region of M(H) as displayed in figure 7. Small hysteresis is observed at 170 K and 160 K. For the M(H) at 100 K and below, the virgin curve initially shows a linear rise in magnetization with field up to $\sim$ 500 Oe and then the slope of the curve changes and it lies outside the hysteresis loop. Such a feature resembles to that of a field-induced transition. We can then define $H_{cric} \sim$ 500 Oe as a crossover field. This value of crossover field remains roughly the same for all the subsequent M(H) curves. It is coincidental that this value of crossover field is the same as the dc field applied in the χ_{ac}(T) measurements at which the peak at T^* smears out completely. The overall features observed in the M(H) curves clearly demonstrate that the Ni-Mn-In system does not show a pure ferromagnetic order. Hence T^* cannot be assigned to be the FM ordering temperature of the martensitic phase. The competing magnetic interactions present in the system do not allow the ferromagnetic state to stabilize. Moreover, the magnitude of the coercive field is too small and does not increase with the fall in temperature. This suggests that there is no clustering of regions having ferromagnetic character. It thus implies that the possibility of segregation of magnetic entities forming regions of varied stoichiometry should be ruled out.

M(H) at 10 K and 5 K show shifted loops with nearly zero coercive magnetization at H = 0. Typically, pinching of the hysteresis loop of this nature taking place along the magnetic field axis has been observed in systems like small coated particles, inhomogeneous materials, thin films and bilayer [15]. This effect is usually observed when the FM/AFM system is either zero field cooled in a demagnetized state or field cooled from above the Neel temperature of the antiferromagnet [16]. It is related to a FM/AFM exchange coupling interaction across the interface and believed to be due to the formation and pinning of domains either in the FM or in the AFM [17, 18]. The observation of such a feature in the low temperature magnetization for the present sample suggests a canted spin structure with ferromagnetic and antiferromagnetic spin components in zero magnetic field, and the ferromagnetism is field-induced. Hence the reduction in magnetization to almost zero upon martensitic transformation and the T^* signature is apparently due to incipient antiferromagnetic coupling inherent in the unit cell of the compound. The two interactions compete with each other throughout the measurement temperature range giving rise to complex behaviour in the magnetic properties of the Ni-Mn-In system.

Figure 8(a) represents the temperature dependence of resistivity measured in the region 5 K to 380 K. The most striking feature here is the large jump in the resistivity of $Ni_{50}Mn_{35}In_{15}$ at room temperature. When viewed from the high temperature side, a change in slope of $\rho(T)$ is seen at about $\sim$ 310 K. This is the signature of ferromagnetic ordering. At about 302 K, the resistivity sharply increases resulting in a large step-like feature at the start of the martensitic transition. With further fall in temperature the overall resistivity does not seem to change much down to the lowest measurement temperature.

A small step or kink in resistivity has routinely been observed for several other martensitic materials. Infact, such a feature along with the associated thermal hysteresis has traditionally been considered as the signature for martensitic transition (see for example reference [19]). The reason for the step /kink is believed to be the trapping of electrons in the nested regions of the Fermi surface due to long range structural ordering formed as a consequence of martensitic phase change. And the nesting vector corresponds to the modulation of martensite formed [20, 21, 22]. However, after the transformation is complete, ρ in the martensitic phase can be extrapolated to match with the curve obtained before the transition had occurred. What makes the observed step like feature special in $Ni_{50}Mn_{35}In_{15}$ is the accompanying change in magnitude of resistivity. It may be noted that the change in $\rho(T)$ is about $\sim$40% in this case.

The anomalous feature in the $\rho(T)$ of $Ni_{50}Mn_{35}In_{15}$ resembles to that observed in the intermetallic compounds undergoing a transition to AFM state. A typical example being $CeFe_2$ and its substitutional derivatives [14, 23]. In such cases, a rise in resistivity after the AFM transition is said to be due to the formation of super-zone gap. Due to the establishment of the AFM sublattice, the underlying zone boundaries get re-defined giving rise to a gap at the Fermi level. The conduction electrons thus have to overcome this gap resulting in a large anomaly in the transport property of the AFM state. The

magnetic properties of $Ni_{50}Mn_{35}In_{15}$ already reveals a possibility of AFM interactions being present in the system along with the underlying FM ones. Thus the anomalous feature in the $\rho(T)$ and all other aforementioned measurements indicate that the AFM interactions develops when the system heads towards the structural transition. To ascertain this further, we measured resistivity as a function of temperature in constant magnetic fields. The $\rho(T)$ curves in the presence of different magnetic fields show a very similar behavior to that at zero field. However, the martensitic transition temperature shifts to lower value with the increase in the magnetic field. The $\rho(T)$ curves at a magnetic field of 5 T and 9 T are also shown in figure 8(a). This trend implies that the magnetic field suppresses the structural phase transition temperature and stabilizes the FM phase. This observation is in congruence to that observed in figure 8(b) where a decrease in the T_M is observed in the field cooled data recorded at 1 T field. Also, the hysteresis observed in the field cooled and field warmed magnetization data further indicates the first order nature of the martensitic transformation. The antiferromagnetic interaction is seen to couple strongly with the martensitic phase transformation.

Magnetism in Heusler alloys has always been fascinating and continues to attract a lot of research interests till date [24]. As follows from the magnetic properties of $Ni_{50}Mn_{35}In_{15}$, the system exhibits complex interplay between ferro- and antiferromagnetic order. The competition between these two magnetic interactions exists through the entire temperature range of measurement up to T_C. Microscopically, the formation of the competing phases can be related to the interatomic exchange interactions that are dominated by separation between atoms and the change in conduction electron density. In the Mn- based Heusler systems, the spatial separation between the neighbouring Mn atoms being comparatively large ($\sim$ 4Å), a considerable direct overlap of Mn *3d* states is not observed here. Consequently, an indirect RKKY type exchange mediated via the conduction electrons of the system is often revoked to describe the magnetic ordering in these materials [25]. In addition, if such systems undergo a martensitic transition, the change in interatomic distances upon transformation are expected to highly manipulate the magnetic interactions.

Amongst the stoichiometric Ni_2MnZ (Z = Ga, In, Sn, Sb), only Ni_2MnGa undergoes a martensitic transformation. Experimentally, all of them are ferromagnetic and have similar values of the Curie temperature. In the case of Ni_2MnIn, the ferromagnetic ordering takes place at $T_C = 315$ [6] and the martensitic transformation is observed only in the non-stoichiometric, Mn rich compositions. The ordered crystal structure in such Mn rich composition contains excess Mn atoms at the In site in addition to its regular $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ sites. We have previously studied the local crystal structure of these systems in the cubic and martensitic phase and obtained the exact interatomic separation between constituent atoms in both the phases [26]. In the cubic phase, $Ni_{50}Mn_{35}In_{15}$ has a lattice parameters of $\sim$ 6.04 Å . Accordingly, the Mn atoms which substitute In atoms develop an additional Mn-Mn interaction at $\sim$ 2.911 Å with a 4.2 coordination number. While the separation between Mn atoms present at its own site is 4.27Å with coordination number 12. Since the magnetic coupling between atoms

is governed by the interatomic separation, the Mn-Mn interactions at 4.27Å is FM in nature. While the coupling between Mn atoms at ~ 2.91 Å apart, must be AFM. The AFM nature of such correlations have previously been anticipated from high resolution neutron powder diffraction measurements on similar composition, $Ni_{50}Mn_{34}Sn_{16}$ [27]. However it is important to note that no additional diffraction peaks corresponding to an antiferromagnetic sublattice was observed in this measurements.

Moreover, it is also known from the local structure study of $Ni_{50}Mn_{35}In_{15}$ that the cubic structure gets highly unstable with Mn atoms moving with largest amplitude of displacement as the T_M is approached. Such vibration of Mn atoms from its crystallographic position weakens the FM sublattice leading to its collapse at the structural phase transformation. The subsequent change in interatomic separation further uncovers the AFM sublattice interactions. These AFM interactions drive the system to nearly zero magnetic moment.

Once the structural transformation is complete and the martensitic phase is fully established, the constituent atoms cease to move vigorously. This is reflected in the low temperature bond distance and the associated thermal mean square variation. The change in crystal symmetry generates Mn-Mn bond at 2.89 Å with coordination number 4.2, while the Mn-Mn bonds at 4.27 Å in the cubic phase split into two correlations: 4.19 Å with coordination number 8 and 4.4Å with coordination number 4. The underlying magnetic interactions also get re-established with such a change and the associated FM/AFM interactions start competing resulting in an anomaly like at T^* in the temperature dependent magnetization measurements. The relative strength of the two magnetic interactions depends on the magnitude of the applied field. The average Mn-Mn distance for ferromagnetic interaction (i.e. 4.19 Å and 4.4Å) in the martensitic phase equals the Mn-Mn distance in the cubic phase. Thus though AFM interactions intensify during the process of martensitic transition, the FM phase emerges upon completion of the structural phase transformation and continues to dominate. Small magnetic field values are sufficient to strengthen the FM order further and help restore it. The two magnetic interactions compete for dominance and continue to co-exist.

4. Conclusion

In conclusion, for the non-stoichiometric Ni-Mn-In compounds the substituent has a crucial bearing on its magnetic properties. Based on our ac susceptibility and dc magnetization study it is clear that the origin of the anomalous magnetization behaviour of $Ni_{50}Mn_{35}In_{15}$ and the exotic properties associated with it are an outcome of competing FM/AFM magnetic interactions. The AFM interactions manifests when the system heads towards structural instability. In the low temperature region, the short-range antiferromagnetic correlations are easily suppressed by magnetic fields exceeding few hundred Oesterds once the structural transformation is complete.

References

[1] Sutou Y, Imano Y, Koeda N, Omori T, Kainuma R, Ishida K and Oikawa K, 2004 *Appl. Phys. Lett.* **85**, 4358

[2] Webster P J, Zeibeck K R A, Town S L and Peak M S, 1984 *Philos. Mag* **49**, 295

[3] Ullakko K, Huang J K, Kanter C, O'Handley R C and Kokorin V V, 1996 *Appl. Phys. Lett.* **69**, 1966

[4] Tickle R and James R D, 1999 *J. Magn. Magn. Mater.* **195**, 627

[5] Sozinov A, Likhachev A A, Lanska N and Ullakko K, 2002 *Appl. Phys. Lett.* **80**, 1746

[6] Webster P J and Ziebeck K R A, 1988 *Alloys and Compounds of d-Elements with Main Group Elements*, P. 2, edited by Wijn H R J, Landolt-Börnstein, New Series, Group III, **19/c** (Springer, Berlin) 75184

[7] Krenke T, Acet M, Wassermann E F, Moya X, Manosa L and Planes A, 2006 *Phys. Rev.* B **73**, 174413

[8] Yu S Y, Liu Z H, Liu G D, Chen J L, Cao Z X, Wu G H, Zhang B and Zhang X X, 2006 *Appl. Phys. Lett.* **89**, 162503

[9] Pathak A K, Khan M, Dubenko I, Stadler S and Ali N, 2007 *Appl. Phys. Lett.* **90**, 262504

[10] Bhobe P A, Priolkar K R and Nigam A K, 2007 *Appl. Phys. Lett.* **91**, 242503

[11] Sharma V K, Chattopadhyay M K and Roy S B, 2007 *J. Phys. D: Appl. Phys.* **40**, 1869

[12] Albertini F, Morellon L, Algarabel P A, Ibarra M R, Pareti L, Arnold Z and Calestani G, 2001 *J. Appl. Phys.* **89**, 5614

[13] Kasper J S and Kouvel J S, 1959 *J. Phys. Chem. Solids* **11**, 231

[14] Ali N and Zhang X, 1992 *J. Phys.: Condens. Matter* **4**, L351

[15] Nogues J and Schuller I K, 1999 *J. Magn. Magn. Mater.* **192**, 203

[16] Brück S, Sort J, Baltz V, Suriãch S, Muñoz J S, Dieny B, Baró M D and Nogués J, 2005 *Adv. Mater.* **17**, 2978

[17] Zhang S and Li Z, 2001 *Phys. Rev.* B **65**, 054406

[18] Miltényi P, Gierlings M, Keller J, Beschoten B, Güntherodt G, Nowak U and Usadel K D, 2000 *Phys. Rev. Lett.* **84**, 4224

[19] Vasil'ev A N, Buchel'nikov V D, Takagi T, Khovailo V V, and Estrin E I, 2003 *Phys. Usp.* **46**, 559 and references therein.

[20] Zhao G L, Leung T C, Harmon B N, Keil M, Müllner M and Weber W, 1989 *Phys. Rev.* B **40**, 7999

[21] Wilkinson I, Hughes R J, Major Zs, Dugdale S B, Alam M A, Bruno E, Ginatempo B and Giuliano E S, 2001 *Phys. Rev. Lett.* **87**, 216401

[22] Velikokhatnyi O I and Naumov I I, 1999 *Phys. Solid State.* **41**, 617

[23] Garde C S, Ray J and Chandra G, 1990 *Phys. Rev.* B **42**, 8643

[24] Sasioglu E, Sandratskii L M and Bruno P, 2008 *Phys. Rev.* B **77**, 064417

[25] Kübler J, Williams A R and Sommers C B, 1983 *Phys. Rev.* B **28**,1745

[26] Bhobe P A, Priolkar K R and Sarode P R, 2008 *J. Phys. D: Appl. Phys.* **41**, 045004

[27] Brown P J, Grandy A P, Ishida K, Kainuma R, Kanomata T, Neumann K-U, Oikawa K, Ouladdiaf B and Ziebeck K R A, 2006 *J. Phys.:Condens. Mater.* **18**, 2249

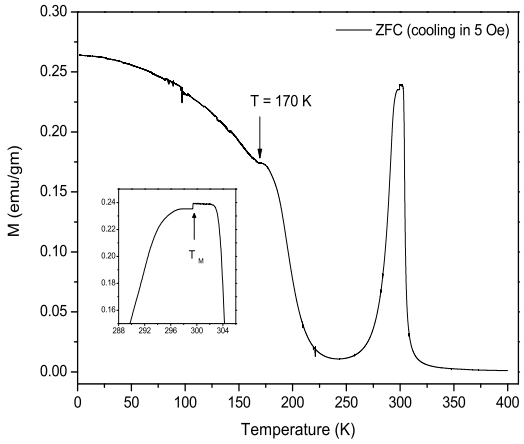

Figure 1. Magnetization as a function of temperature measured while cooling in zero field state with nominal field of 5 Oe. Inset shows the drop in magnetization that occurs upon martensitic transition.

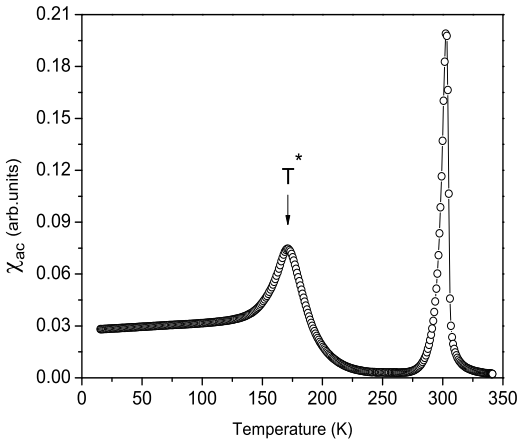

Figure 2. The real component of χ_{ac} for $Ni_{50}Mn_{35}In_{15}$ measured after cooling the the sample from room temperature to 5 K in zero field.

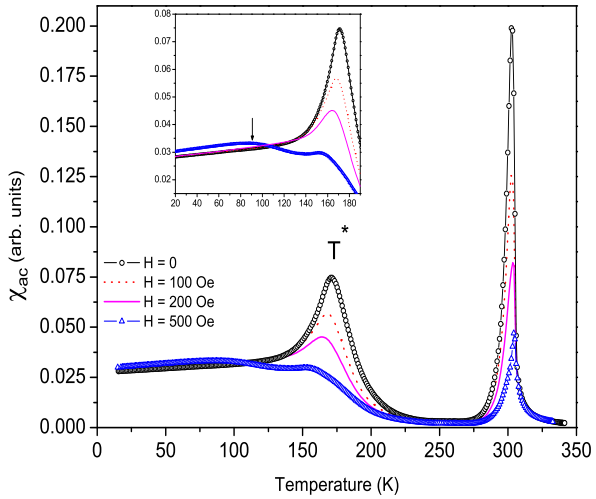

Figure 3. (Colour online) χ_{ac}(T) measured at $f = 133$ Hz in different dc fields. The intensity of the peak-like anomaly at T* decreases with increasing field. The inset shows the appearance of a hump like feature at a lower temperature in the H = 500 Oe data after the peak at T* has considerably been suppressed.

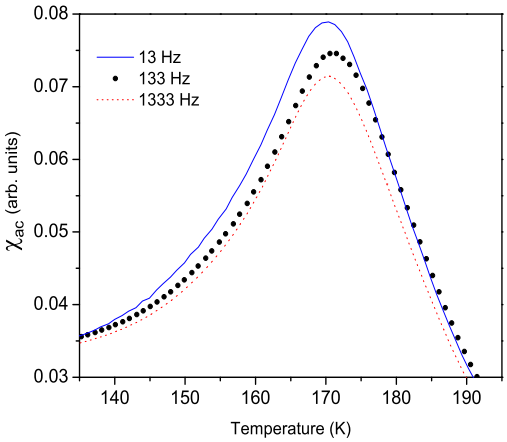

Figure 4. (Colour online) The region around T* is shown for the real component of χ_{ac} vs T measured at different frequencies. T* does not show any systematic dependence on *f*.

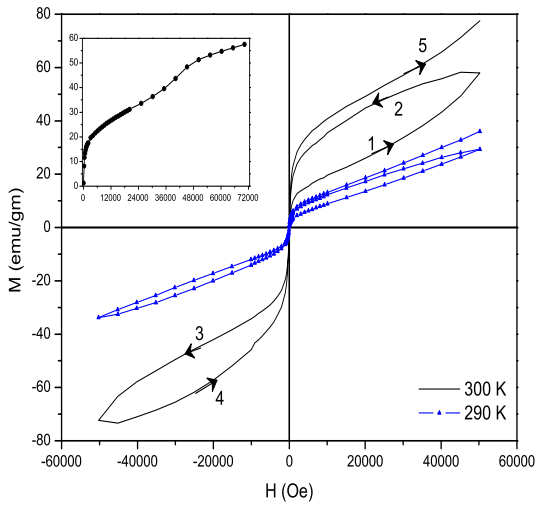

Figure 5. (Colour online) Magnetization as a function of applied field recorded in the vicinity of the martensitic transition. The metamagnetic character is seen at intermediate field values as shown in the inset.

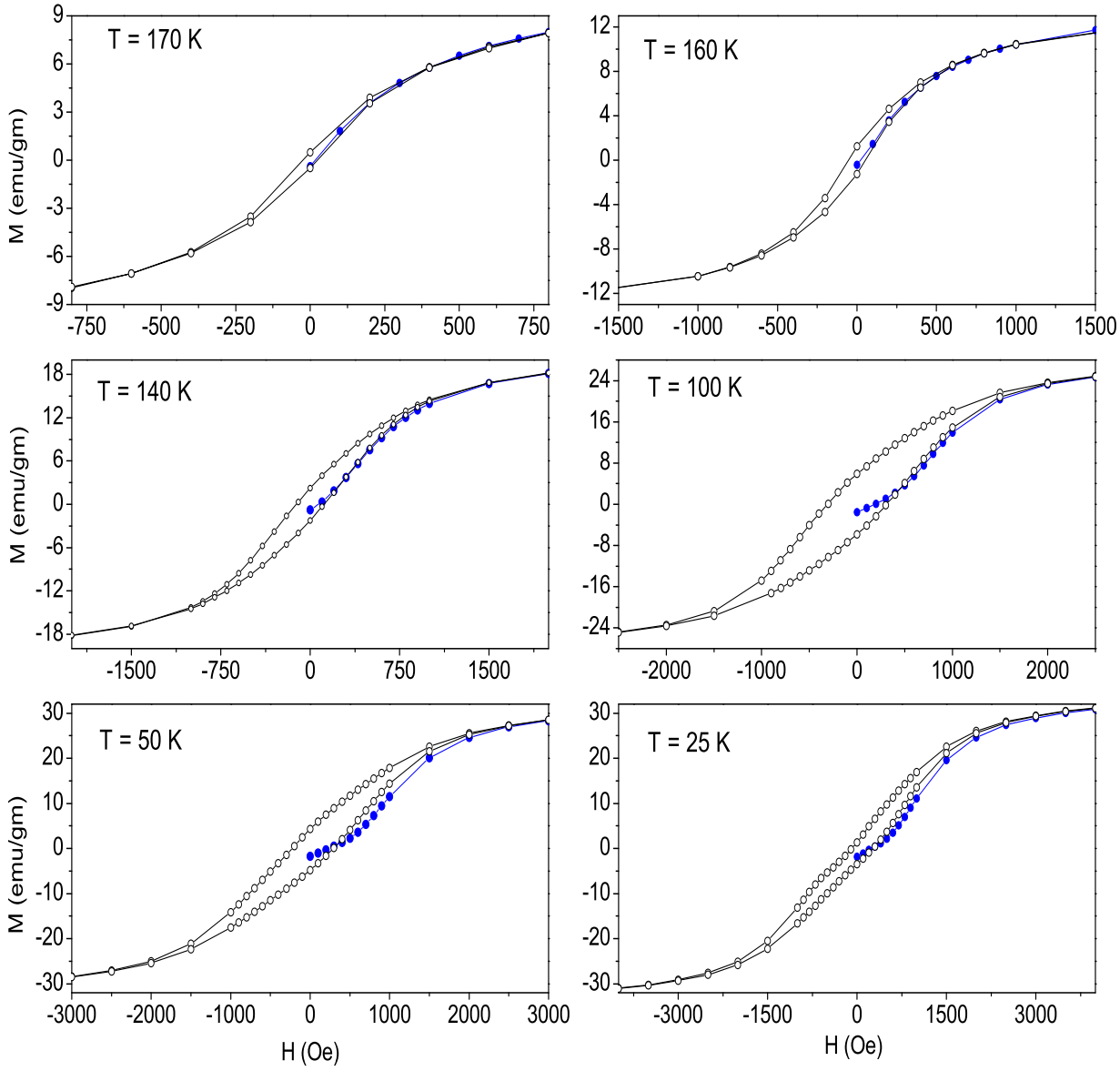

Figure 6. (Colour online) M(H) loops at different temperatures below T^*. Measurements were carried out by sweeping the field from +5 T to -5 T. A magnified view for a smaller field interval is shown here for clarity. Data points for the virgin curve is shown as solid circles while for the rest of the loop it is shown with empty circles.

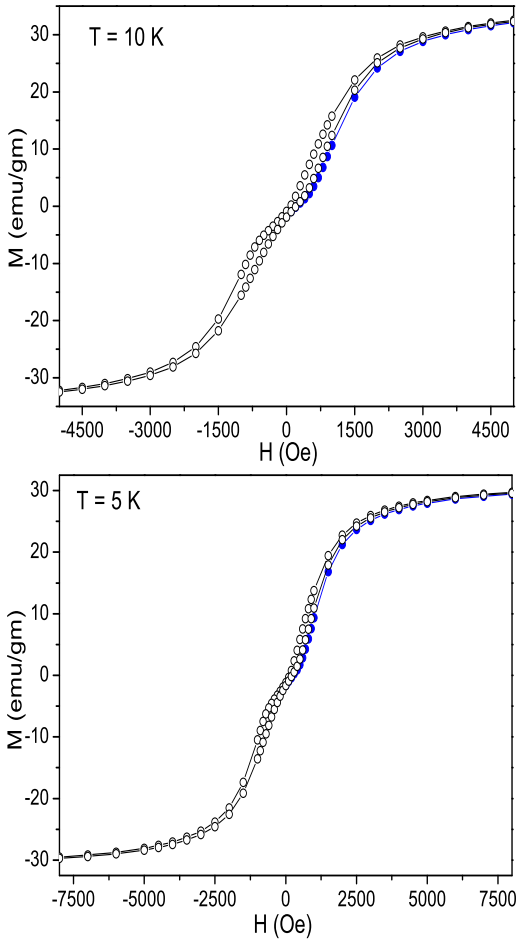

Figure 7. (Colour online) The double shifted loops obtained at 10 K and 5K are represented. The virgin curve is shown as solid circles while for the rest of the loop it is shown with empty circles. Measurements were carried out from +5T to -5T. A magnified view is shown here.

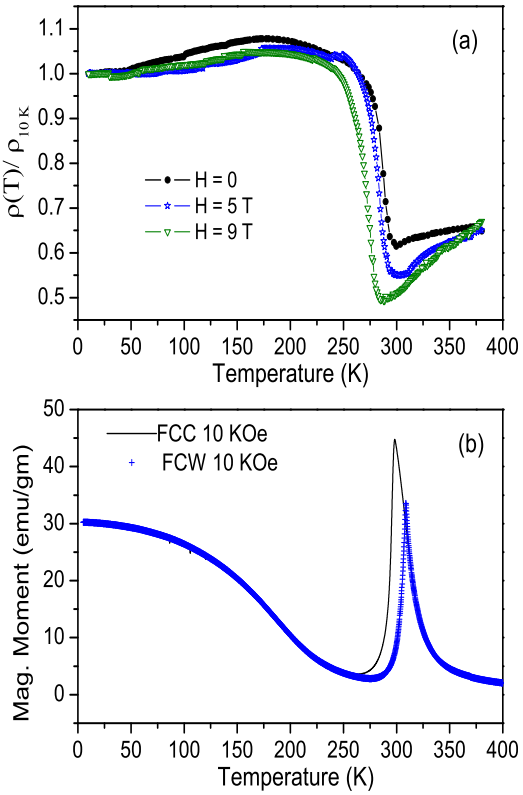

Figure 8. (Colour online)(a) Normalized resistivity measured at different magnetic fields in the temperature range of 10 K to 380 K. (b) Temperature dependent magnetization measured in the magnetic field of 10 KOe. The data was collected during the field cooled state (FCC) and the subsequent warming (FCW) in the same field.