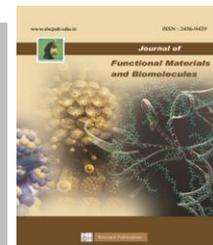




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## Structural, Magneto-transport and Strain induced Anomalous Hall effect properties of Ni-Mn-Ga Nanocrystalline Thin Films

K. Kamala Bharathi\*

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### Abstract

Nanocrystalline Ni-Mn-Ga films were prepared employing DC sputtering method and their structural, electrical and magneto-transport properties were investigated. Structural investigation indicates that the film grown at the substrate temperature ( $T_s$ ) of 350°C crystallizes in  $L2_1$  structure. Temperature variation of electrical resistance showed an abrupt change at 240 K and at 170 K while cooling and heating respectively, which can be attributed to the martensitic transformation. Magnetoresistance of -0.03% (at 80 K) has been observed for the film grown at 350°C. Transport properties of amorphous film indicate the typical disorder nature of the alloy. The negative extraordinary Hall coefficient ( $R_s$ ) is observed for the film grown at 350°C. Presence of hysteresis at room temperature reveals that the Ni-Mn-Ga film possesses the perpendicular anisotropy. Hysteresis loop become contracted at 80 K because of the strain mediated anisotropy at low temperature.

**Keywords:** Ni-Mn-Ga; Hall effect; Structural transformation; Magnetoresistance.

### 1 Introduction

Co-Ni-Ga, Co-Ni-Al and Ni-Mn-Ga [1-3] based alloys possess shape memory (SM) effect with the application of magnetic field at room temperature (RT) superior than any, complex oxides piezoelectric and magnetostrictive strains. Several research groups have been exploring the prospect of utilizing these SM metals as transducers and actuators [4,5].

$Ni_2MnGa$  and Ni-Mn-Ga are the magnetic SM alloys which are explored by many research groups, which show a first or second order phase transition (structural transition from cubic to tetragonal) at low temperatures [1-5]. In addition to that, with the application of magnetic field, strains in the structure have been shown in these alloys upto 10% [6]. The macroscopic shape change occurs due to the movement of double variants with the application of external strain (magnetic field) [7]. Off-stoichiometry, overload of any metal (Mn/Ni) in Ni-Mn-Ga alloy can elevate the structural transition close to RT [1-7].

SM properties can be accurately tuned for particular applications by controlling the metal ion ratio, growth conditions and preparation methods. High brittle nature of  $Ni_2MnGa$  in bulk form makes it unsuitable for device fabrication. These disadvantage can be surpassed by preparing these alloys in high quality films or in oriented crystal form [8,9]. Fabrication SM alloy thin films are the more hopeful way out to the difficulty of brittleness and utilizing for miniaturization. Recently few authors have demonstrated that thin films of Ni-Mn-Ga exhibit temperature dependent magnetoresistance (decrease in resistance) properties [10,11]. At 10 kOe ( $T=23$  K), -1.2% changes were observed for Ni-Mn-Ga thin films prepared on Si substrates employing pulse laser deposition method [12]. In addition to that, Ni-Mn-In,  $Co_2ZrSn$  Heusler alloys thin films are reported to exhibit anomalous Hall effect [13,14] and the massive value of the Hall angle ( $\tan^{-1}0.5$  for Ni-Mn-In), which is the interesting feature for potential usages of the Hall effect insensors (magnetic) and spintronics field.

In the present work an attempt has been made to investigate the structural transformation through electrical resistivity studies, magnetoresistance and the anomalous Hall effect of Ni-Mn-Ga thin film. Interestingly, from the studies carried out in this work, Ni-Mn-Ga film shows anomalous Hall effect at various temperatures. The structural transformation through resistivity studies, magnetoresistance properties and the conduction mechanisms of Ni-Mn-Ga films are reported in this paper.

### 2 Experimental

Ni-Mn-Ga films were grown onto glass and  $SiO_2/Si$  substrates by sputtering (DC magnetron). Chamber was evacuated to  $\sim 10^{-6}$  Torr before the deposition. Two inch Ni-Mn-Ga target (99.95 % purity) was used for thin film sputter deposition process. Distance between the cathode and the substrate was kept as 8 cm. Thin films were deposited at the 300 W electrical power. The samples were deposited at room temperature (RT) and at 350°C.

Structural, transport and Hall effect properties of Ni-Mn-Ga thin films were carried out. X-ray diffraction (XRD) at RT was carried out to explore the structural properties

\*Corresponding author e-mail: [kkamalabharathi@gmail.com](mailto:kkamalabharathi@gmail.com)

Department of Physics, Research Institute, SRM Institute of Science and Technology, Kattankulathur, Chennai

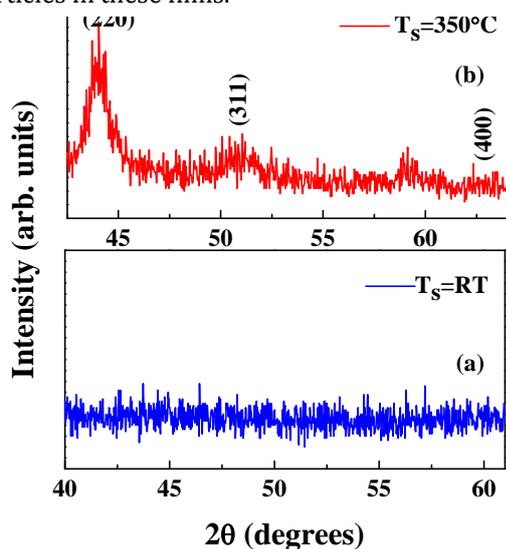
of on Ni-Mn-Ga films, employing PANalytical (X'pert PRO) instrument. The grain size ( $D_{hkl}$ ) was calculated employing the following equation [15]

$$D_{hkl} = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

where  $D_{hkl}$ ,  $\lambda$ ,  $\beta$  and  $\theta$  are the size ( $\sim 16$  nm), wavelength X-ray, peak width, and angle of the chosen diffraction peak. Surface morphology of the Ni-Mn-Ga thin films were studied using scanning electron microscope (Hitachi S-4800). Elements present in these thin films were investigated employing EDS (energy-dispersive x-ray spectrometry). Resistivity (DC) characteristics were carried out using Van der Paw technique at various temperatures (45-300 K) in a Helium refrigerator (closed cycle). Hall resistivity experiments were carried out RT and at 80 K by applying the magnetic field perpendicular to the thin film surface.

### 3 Results and Discussion

Figure 1 depicts the X-ray diffraction patterns of Ni-Mn-Ga films grown at RT and at 350°C. The XRD curve (Fig. 1 (a)) of Ni-Mn-Ga film grown at RT is observed to be amorphous in nature. The XRD curve (Figure 1 (b)) of Ni-Mn-Ga film grown at 350°C exhibits diffraction peaks corresponding to  $L2_1$  crystal structure. Broad peak observed at 44° indicates the existence of very small nanoparticles in these films.

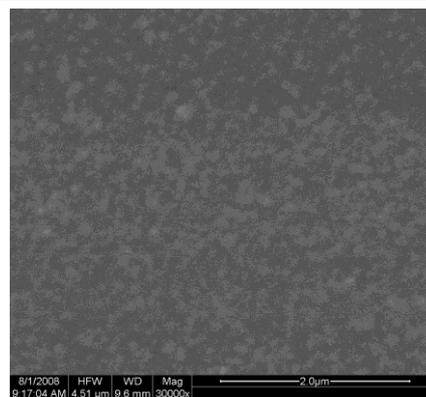


**Figure 1** XRD patterns of Ni-Mn-Ga film grown at RT (a); XRD patterns of Ni-Mn-Ga film grown at 350°C (b)

The lattice mismatch (strain factor  $\Gamma$ ) is estimated from the formula:

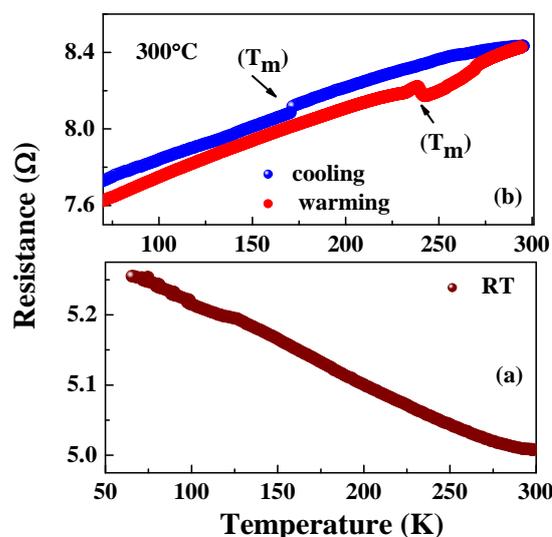
$$\Gamma = \frac{d_{(100)Si} - d_{(220)sample}}{d_{(220)sample}} \% \quad (2)$$

The value of  $\Gamma(\%)$  is seen to be 24% in the present case due to the lattice mismatch between the substrate and the Ni-Mn-Ga thin films.



**Figure 2** SEM image of Ni-Mn-Ga film grown at 350°C

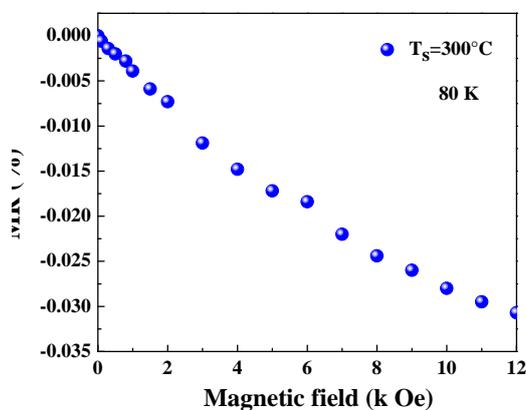
The high resolution SEM image of Ni-Mn-Ga film grown at 350°C is shown in Figure 2. Excellent microstructure and identical allocation of intense spherical particles are very clearly seen. The composition determined using EDAX was found to be Ni-51.7, Mn-23.1 and Ga-25.2.



**Figure 3** Temperature variation of resistance of Ni-Mn-Ga film grown at RT (a); Temperature variation of resistance of Ni-Mn-Ga film grown at 350°C (b)

Electrical resistance of Ni-Mn-Ga films was measured using Van der Paw method from 300 K to 50 K. Figure 3 (a) shows the temperature variation of resistance for the film grown at RT. The resistance was found to increase with decreasing temperature. The  $R(65\text{ K})/R(300\text{ K})$  is found to be  $\sim 1$ . Figure 3 (b) shows the resistance curve for the film grown at 350°C. The resistance was found to decrease with decreasing temperature, which is the typical metallic behaviour. The  $R(42\text{ K})/R(300\text{ K})$  is found to be  $\sim 0.90$ . The electrical resistance showed an abrupt change at  $T_m=240$  K and at 170 K while cooling and heating respectively. The observed peculiarities at 240 K and at 170 K can be attributed to the martensitic transformation. However, the obtained martensitic conversion shows great temperature hysteresis of about 70 K. The sudden change in electrical resistance upon structural transformation is due to the variation of the density of states (DOS) near the

Fermi level. The increase in resistance with decreasing temperature for the film grown at RT is typical for disordered alloys [16]. In addition to that, the structural transition (martensite–austenite) did not contribute to any anomalies of  $R$  vs  $T$  measurements, as seen in film grown at  $T_s=350^\circ\text{C}$ . The observed behaviour indicates the significant structural dependence of the electrical and transport properties of Ni-Mn-Ga alloys [17].

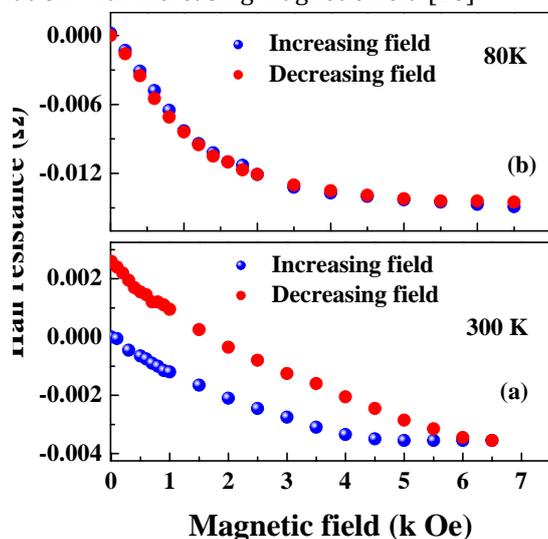


**Figure 4** Magnetoresistance curve (at 80 K) of Ni-Mn-Ga film grown at 350°C

Figure 4 shows the magnetoresistance (MR) curve (at 80 K) of Ni-Mn-Ga film grown at 350°C. The resistance was found to decrease from 7.676  $\Omega$  to 7.646  $\Omega$  (-0.03%) with increasing the magnetic field from 0 to 1.2 k Oe. Magnetoresistance (MR) was calculated using the formula

$$MR = \frac{R(H) - R(0)}{R(0)} \% \quad (3)$$

where,  $R(H)$  and  $R(0)$  represents the resistance at particular field and zero field respectively. The observed MR can be attributed to the movement of the conducting electrons among areas with dissimilar orientations of magnetic moments and inter-domain spin transport. The resistance of the film grown at RT did not show any variation with increasing magnetic field [18].



**Figure 5** Hall resistance curves at 300 K (a) and at 80 K (b) of Ni-Mn-Ga film grown at 350°C

Hall resistance curves for Ni-Mn-Ga film are shown in Fig. 5. Negative  $R_s$  is observed at 300 K as well as at 80 K because of the negative Hall resistance properties of Ni [19].  $R_s$  in the present case appears from side jump of charge carriers and skew-scattering [20,21]. Skew scattering is linked to the coupling between spin and orbit moments and estimated by a stable impulsive Hall angle:  $\theta_H = \rho_H/\rho$  where,  $\rho_H$  and  $\rho$  represents the Hall resistivity and ordinary resistivity respectively.

The magnetic field variation of Hall resistance shows feature comparable to the magnetization measurements. Field variation of  $R_s$  at 300 K is seen to almost saturate at 5k Oe. Nonlinear Hall resistivity in the present case arises from the anomalous Hall effect and reveals the presence of electron scattering with magnetic field. Huge hysteresis is seen at 300 K compared to 80 K. The subsistence of huge hysteresis at 300 K shows that the Ni-Mn-Ga films possess perpendicular anisotropy. Out of plane slanting anisotropy in a few Ni-Mn-Ga films has been reported by Chernenko *et. al* [22]. Therefore, the observed hysteresis at RT in the present case could be due to the perpendicular anisotropy. The anisotropy with decreasing temperatures in Ni-Mn-Ga films might be planar because of the enlarged strain enforced anisotropy due to the immense magnetostriction at temperatures below RT [23]. In-plane anisotropy might be forcing the moments align in to the in-plane, lead to the near absence of remanence.

#### 4 Conclusions

Nanocrystalline Ni-Mn-Ga films were prepared employing sputter-deposition and their structural, electrical and magneto-transport characteristics were explored. Structural investigation indicates that the film grown at room temperature is amorphous, whereas, the film grown at the substrate temperature ( $T_s$ ) of 350°C crystallizes in  $L2_1$  structure. Temperature variation of electrical resistance showed an abrupt change at 240 K and at 170 K while cooling and heating respectively, which can be attributed to the martensitic transformation. Martensitic transformation exhibits large thermal hysteresis of about 70 K. Magnetoresistance of -0.03% (at 80 K) has been observed for the film grown at 350°C. The negative extraordinary Hall coefficient ( $R_s$ ) is observed for the film grown at 350°C. Hysteresis nature at RT shows that the Ni-Mn-Ga film possesses the perpendicular anisotropy. Narrow loop at 80 K is because of the strain induced anisotropy at those temperatures.

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