Magnetotransport properties of Ni-Mn-In Heusler Alloys: giant Hall angle

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Abstract. We report experimental results on phase transitions, magnetic properties, resistivity, and Hall effect in Ni_{50-Mn_{50-x}}In_x (15<x<16.2) Heusler alloys. Several distinguishing features of magnetotransport properties were clearly observed in the vicinity of the first order structural martensitic transition at \( T_M \) and the ferromagnetic-paramagnetic transition at the Curie temperature \( T_C \) of the austenitic phase. It was found that the Hall resistivity \( \rho_H \) at 15 kOe is positive in martensitic and negative in austenitic phase, sharply increases in the vicinity of \( T_M \) up to \( \rho_H \approx 50 \mu \Omega\cdot\text{cm} \). This value is almost two orders of magnitude larger than that observed at high temperature \((T\approx200K)\) for any common magnetic materials, and comparable to the giant Hall effect resistivity in magnetic nanogranular alloys. The Hall angle \( \theta_H = \tan^{-1}(\rho_H/\rho) \) close to \( T_M \) reaches \( \tan^{-1}(0.5) \) which is the highest value ever reported for a magnetic material.

1. Introduction

It has been discovered recently that off-stoichiometric Ni-Mn-In Heusler alloys demonstrate the field and temperature induced first order phase transitions (FOPT) known as metamagnetic and structural martensitic transitions, respectively ([1, 2] and references therein). The martensitic and austenitic crystal phase modifications of the compounds are characterized by the large difference in magnetic moment. Therefore Ni-Mn-In based Heusler alloys can be considered as a magnetically disordered system in the vicinity of FOPT where the different magnetic and crystal phase co-exists.

In this paper we report the recent experimental results on magnetization \( M(T,H) \), resistivity \( \rho(T,H) \) and Hall resistivity \( \rho_H(T,H) \) in Ni_{50-Mn_{50-x}}In_x alloys focusing on the Hall resistivity. We observed very unusual field dependences of \( \rho_H \) and a giant value of Hall effect angle, \( \theta_H = \tan^{-1}(\rho_H/\rho) = \tan^{-1}0.5 \), which is the highest value ever reported for a magnetic material. It is shown that these features are due to the cluster-like microstructure of alloys under the investigation and electronic band structure changes in the vicinity of FOPT.

The Hall resistivity \( \rho_H \) in ferromagnets can be written as a sum of two terms:

\[
\rho_H = R_A B_z + R_s 4\pi M_z
\]

(1)

where the first term describes the ordinary Hall effect, which is related to the Lorentz force, and the second one is the anomalous Hall effect (AHE) resistivity. \( R_A \) is the AHE coefficient, \( M_z \) and \( B_z \) are magnetization and magnetic induction components respectively. It is well known that AHE originates from the spin-orbit interaction but the main mechanisms of AHE are still under debate (see [3, 4] for...
Since AHE is a relativistic effect and there is a correlation between AHE resistivity and ordinary resistivity, the Hall angle in ferromagnetic metals and alloys \( \theta_H = \tan^{-1}(\frac{\rho_H}{\rho}) \) is rather small. For example, it is about \( \theta_H = \tan^{-1}(2 \times 10^{-3}) \) in stoichiometric Ni$_2$MnSn Heusler alloys [5]. Even in the case of giant Hall effect (GHE) [6-9], which was discovered in nanogranular metal-insulator alloys near percolation threshold [6-8] and in all-metallic nanogranular alloys [9], the Hall angle does not exceed \( \theta_H = \tan^{-1}(10^{-2}) \) because of their very high resistivity. Several models, such as enhanced spin-orbit interaction, enhanced scattering by interfaces [10], and quantum percolation [11] were proposed for GHE explanation, but each of these hypotheses has its own drawback. GHE appears only if a metal volume fraction in the heterogeneous nanogranular systems is close to the percolation threshold, and if the granular size is less than several nanometers [7-10]. Such heterogeneity can be also expected in the Ni-Mn-In based Heusler alloys in the vicinity of FOPT and therefore it is reasonable to assume that this system also exhibit GHE.

2. Results and Discussion

The Ni$_{50}$Mn$_{50-x}$In$_x$ with x= 15.0, 15.05, 15.2, and 16 samples have been fabricated using the conventional arc-melting method described in Ref. [12]. Thermomagnetic curves M(T,H) have been measurement using VSM (LakeShore 7400 System) and SQUID (Quantum Design) magnetometers in the temperature interval 4-400K and magnetic field up to 50 kOe. Magnetotransport measurements were carried out by standard four-probe method using a fully automated system in a temperature interval 77-400K at magnetic fields in the range 0.005 - 15 kOe. The transition temperatures have been determined using the local maximums of dM/dT curves. All measurements were carried out during heating after the samples were cooled from 400 K to 5 K at \( H = 0 \).

As one can see from Fig. 1 and 2, where M(T,H) curves are shown, Ni$_{50}$Mn$_{50-x}$In$_x$ system passes

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**Figure 1.** Typical for Ni$_{50}$Mn$_{50-x}$In$_x$ system low and high field cooled M(T) curves (shown for x=15.05 and x=15.2). \( T_C \) and \( T_{CM} \) are the Curie temperature of austenitic and martensitic phases respectively. \( T_M \) is the temperature of martensite to austenite transition. Inset: (T-x) phase diagram of Ni$_{50}$Mn$_{50-x}$In$_x$ obtained at \( H=100 \) Oe.

**Figure 2.** M(H) isotherms of Ni$_{50}$Mn$_{50-x}$In$_x$ system (shown for x=15.2) typical for \( T<T_{CM} \) (shown at T=5K), \( T_{CM}<T<T_M \) (198K), \( T_M<T<T_C \) (340K) and \( T_C<T \) (314K) temperature intervals.
one field (metamagnetic), and at least three temperature induced phase transitions. The ferromagnetic type of \( M(H) \) is observed after and before structural transition in \( T<T_{CM} \) and \( T_{M}<T<T_{C} \) intervals respectively. The field induced metamagnetic transition from the magnetic state with low magnetic moment to ferromagnetic state is clearly detected in \( T_{CM}<T<T_{M} \) interval. Magnetic state below \( T_{CM} \) is also characterized by much low magnetic moment compared to that after metamagnetic transition. Sharp change of magnetization at \( T_{M} \) associated with martensitic transition is found to be extremely sensitive to magnetic field and shifted about 50 K to low temperature region in \( H=50 \) kOe. All compounds under investigation show the similar behaviour (see inset in Fig. 1). Hall effect resistivity \( (\rho_{H}) \) as a function of magnetic field and temperature is presented in Fig. 3. If \( T>T_{M} \), \( \rho_{H} \) increases with magnetization and the curve changes its slope at some field because the second term in Eq. (1), corresponding to the AEH, tends to saturate with magnetization, but the first term continues to grow with magnetic field (Fig. 3d). This is a typical behaviour for \( \rho_{H} \) of most common magnetic materials. However, below \( T_{M} \) (see Fig.3 (a) and (b)) \( \rho_{H} \) shows abnormal field dependences. Namely, \( \rho_{H}(H) \) continues to increase significantly even at 15 kOe, it changes its slope when there is no evidence of the saturation of magnetization (see \( M(H) \) at \( T=198K \) in Fig.2 (b)), and changes its sign (see Fig.3 (b)). Moreover, in vicinity of \( T_{M} \), \( \rho_{H}(H=15kOe) \) reaches a very large value of \( 50 \mu\Omega\cdot\text{cm} \) (see Fig.3 (c)), which is about two to three orders of magnitude greater than that observed for common magnetic materials, and it is comparable to the GHE resistivity in magnetic nanogranular alloys NiFe-SiO\(_2\) [6-8], and in all-metallic high-resistivity nanogranular alloys [9]. However, the compound with \( x=15.2 \) has a much smaller ordinary resistivity compared with nanogranular alloys, and therefore the Hall angle \( \theta_{H}=\tan^{-1}(\rho_{H}/\rho) \) reaches a giant value of \( \theta_{H}=\tan^{-1}(0.5) \) (see Fig.4).

\[ \text{Figure 3. Hall resistivity of } \text{Ni}_{50}\text{Mn}_{38.4}\text{In}_{15.2} \text{ sample at different temperatures.} \]

\[ \text{Figure 4. Temperature dependencies of } \tan\theta_{H} \text{ and } \rho(T) \text{ (inset) of } \text{Ni}_{50}\text{Mn}_{38.4}\text{In}_{15.2}. \]

It should be underlined that the Hall resistivity does not tend to saturate at 15 kOe and therefore we did not reach the maximum value of GHE. Such experiments are in progress.
To explain the experimental data we should take into account that the giant effect was observed only for fields corresponding to metamagnetic behaviour and only in a very narrow interval of temperatures. Besides, the Hall resistivity changes its sign immediately after reaching the maximum value. As far as metamagnetic transitions in metallic systems are associated with electronic band structure changes, and since the Hall resistivity changes its sign, we can conclude that the main reason for the GHE in our case is an electronic band structure transformation accompanied by an increase of the total density of states at the Fermi level, mostly from the $d$-component, when the Fermi level crosses the degenerated $d$-states. Therefore both the influence of degenerate states and strong disorder due to mixture of austenitic and martensitic phases provide favourable conditions for GHE. The details of proposed mechanism will be discussed elsewhere.

Finally, it should be strongly emphasized here that the value of the Hall angle is the most important characteristic for possible applications of the Hall effect in spintronics, and therefore our results open a new avenue for the study and application of these alloys.

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