

# Hall effect in Taylor-phase and decagonal Al<sub>3</sub>(Mn,Fe) complex intermetallics

Denis Stanić<sup>I,1</sup>, Jovica Ivkov<sup>I</sup>, Ana Smontara<sup>\*,1</sup>, Zvonko Jagličić<sup>II</sup>, Janez Dolinšek<sup>III</sup>, Marc Heggen<sup>IV</sup> and Michael Feuerbacher<sup>IV</sup>

<sup>I</sup> Institute of Physics, Laboratory for the Study of Transport Problems, Bijenička 46, POB 304, 10001 Zagreb, Croatia

<sup>II</sup> Institute of Mathematics, Physics and Mechanics & Faculty of Civil and Geodetic Engineering, University of Ljubljana, Jadranska 19, 1000 Ljubljana, Slovenia

<sup>III</sup> J. Štefan Institute, University of Ljubljana, Jamova 39, 1000 Ljubljana, Slovenia

<sup>IV</sup> Institut für Festkörperforschung, Forschungszentrum Jülich, Jülich 52425, Germany

Received June 12, 2008; accepted August 20, 2008

## Complex metallic alloys / T-phases / Hall effect

**Abstract.** The Hall coefficient ( $R_H$ ) of Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub> ( $x = 0, 2, 4$  and  $6$ ) complex metallic alloys has been measured in the temperature interval from 90 to 400 K. All the alloys are  $T$  (Taylor) phase except Al<sub>73</sub>Mn<sub>21</sub>Fe<sub>6</sub> that is a decagonal ( $d$ ) quasicrystal. The Hall coefficients of all the samples are positive and they decrease strongly with the increase of temperature according to the Curie-Weiss [ $C/(T-\theta)$ ] law. Therefore, for the separation of the normal ( $R_0$ ) and anomalous ( $R_S$ ) Hall coefficients, the results for the paramagnetic susceptibility ( $\chi$ ) in the corresponding temperature interval have been used. The values deduced from  $R_H(\chi)$  plots are about  $-2 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$  for  $R_0$ , and about  $5 \times 10^{-7} \text{ m}^3 \text{ C}^{-1}$  for  $R_S$ . When the possible dependence of  $R_S$  on temperature, that is due to the temperature dependence of the electrical resistivity  $\rho(T)$ , is taken into account the values are about zero for  $R_0$ , and about  $3 \times 10^{-7} \text{ m}^3 \text{ C}^{-1}$  for  $R_S$  (295 K). No significant composition dependence of  $R_0$  has been detected.

## Introduction

The Al–Mn–Fe system contains several complex metallic alloy phases which recently attract increasing interest. Among them is the orthorhombic Taylor ( $T$ ) phase, the structure of which is built of atomic layers stacked along the [010]-direction. Along this axis pentagonal columnar clusters are formed [1]. Therefore, they are considered to be approximants of the decagonal ( $d$ ) Al–Mn phases. The unit cell of the  $T$ -phase contains 156 atoms with many of the sites having either fractional occupation or mixed Al/Mn occupation, so that a great inherent chemical disorder exists on the lattice. As a part of the systematic investigation of the transport and magnetic properties of  $T$ -Al–Mn–Fe, here we present the results of the Hall-effect

measurement on Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub> complex metallic alloys. Al<sub>73</sub>Mn<sub>27</sub> and its ternary extensions Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub> ( $x = 2$  and  $4$ ) are  $T$  phases, whereas Al<sub>73</sub>Mn<sub>21</sub>Fe<sub>6</sub> is a quasicrystalline decagonal phase [2].

## Experimental procedure

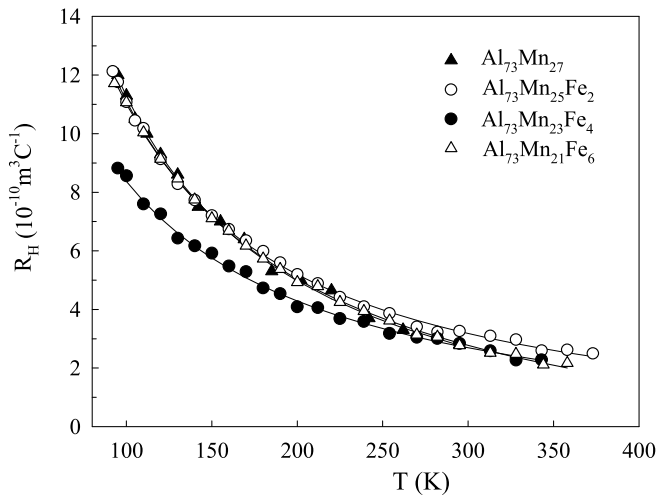
Polycrystalline samples were produced from the constituent elements by levitation induction melting in a water-cooled copper crucible under argon atmosphere. The Hall-effect measurements were performed by a standard AC technique and by a five point method in magnetic fields up to  $1T$ . The measurements were performed in the temperature interval from 90 to 370 K. The average of five data sets was taken to calculate  $R_H$  at each temperature. As we intend to use the same samples for the measurements of other physical properties they were not cut into the appropriate shape for the Hall-effect measurements. The samples were bar-shaped with dimensions of  $1 \times 1 \times 6 \text{ mm}^3$ . The small influence of the inappropriate shape on the demagnetizing field is discussed with the susceptibility results. The temperature-dependent magnetic susceptibility  $\chi$  was investigated in the temperature interval between 2 and 300 K, using a Quantum Design SQUID magnetometer, equipped with a  $5T$  magnet. Electrical resistivity was measured between 1.5 and 300 K using the standard four-probe AC technique.

## Results and discussion

In the temperature interval explored and for the magnetic fields up to  $1T$  the Hall resistivity  $\rho_H$  of all samples is a linear function of the magnetic field ( $B$ ) and therefore we present the results for the Hall coefficient  $R_H = \rho_H/B$  only. In Fig. 1 the Hall coefficient of Al<sub>73</sub>Mn<sub>27</sub> and Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub> is presented as a function of temperature. The shape of the  $R_H(T)$  curves indicates that the samples are paramagnetic, and that the anomalous (spontaneous) magnetic contribution to the Hall effect is dominant.

<sup>1</sup> Permanent address: Department of Physics, University of Osijek, Gajev trg 6, 31000 Osijek, Croatia

\* Correspondence author (e-mail: ana@ifs.hr)



**Fig. 1.** Temperature-dependence of the Hall coefficient of  $\text{Al}_{73}\text{Mn}_{27-x}\text{Fe}_x$ .

In magnetic materials the Hall resistivity follows the empirical relation [3]

$$\rho_H = R_0 B + \mu_0 M R_S, \quad (1)$$

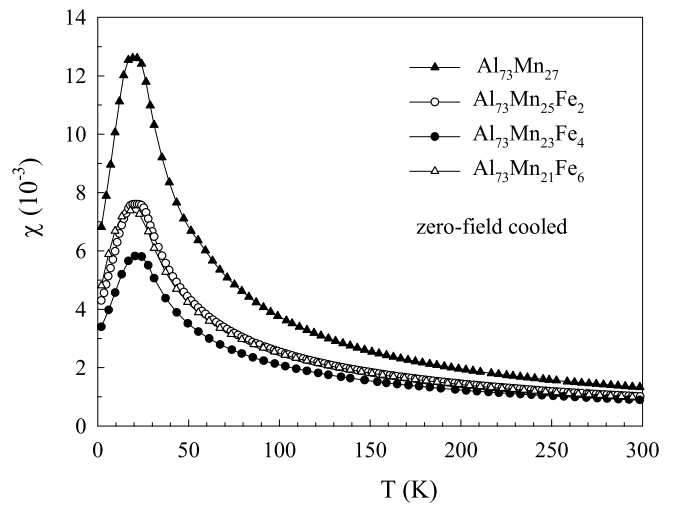
where  $R_0$  and  $R_S$  are the ordinary and anomalous Hall coefficients respectively and  $M$  is the magnetization. The normal and anomalous contributions have different origins. The normal Hall effect comes from the Lorentz force acting on the electrons that conduct electrical current. In crystalline solids it is directly connected with the properties of the Fermi surface and the relaxation times of the electrons. The anomalous or spontaneous Hall effect, on the other hand, is not due to the Lorentz force, but is a consequence of asymmetric scattering which stems from spin-orbit interactions [3] or is due to the influence of the spin-orbit interaction on the electronic wave functions [4]. Asymmetric scattering is still present in the unmagnetized sample and a magnetic field brings this asymmetry up to the macroscopic level by introducing the net magnetization of the sample. In Fig. 2 we present the magnetic susceptibility of our  $\text{Al}_{73}\text{Mn}_{27-x}\text{Fe}_x$  samples as a function of temperature.

These materials belong to the class of magnetically-frustrated spin system [5]. Spin-freezing temperatures of all the samples are well below those of interest for the separation of  $R_0$  and  $R_S$ . Paramagnetic susceptibility follows the Curie-Weiss ( $\chi \sim (T - \theta)^{-1}$ ) law with negative Curie-Weiss paramagnetic temperatures and negligible temperature independent part ( $< 1 \times 10^{-5}$ ). What is very important is that the susceptibility is small and of the order  $10^{-3}$ . Therefore, in spite of the inconvenient geometry of the samples, we can completely neglect the effects of demagnetizing fields and the difference between the inner and applied field. Therefore we can replace  $\mu_0 \cdot M$  with the  $\chi \cdot B$  so that Eq. (1) yields

$$R_H = R_0 + \chi \cdot R_S. \quad (2)$$

If  $R_0$  and  $R_S$  do not depend on temperature,  $R_H$  is linear in  $\chi$  and the normal Hall coefficient is given by the intercept on the  $R_H$  axis and  $R_S$  by the slope of the straight line.

The Hall coefficient  $R_H$  of  $\text{Al}_{73}\text{Mn}_{27}$  and  $\text{Al}_{73}\text{Mn}_{27-x}\text{Fe}_x$  samples as a function of paramagnetic sus-

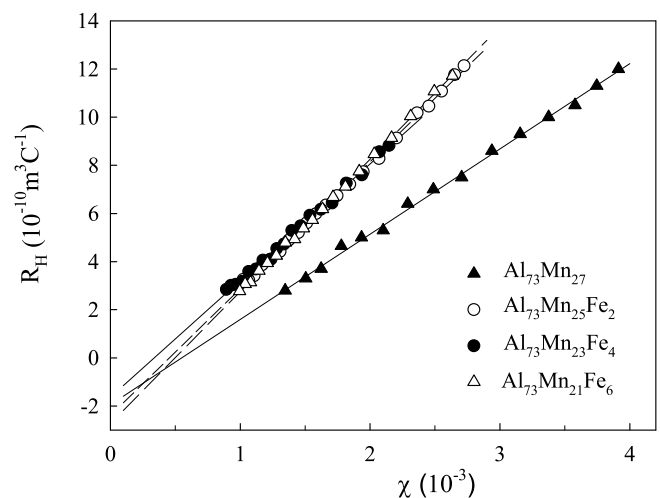


**Fig. 2.** Zero-field-cooled magnetic susceptibility of  $\text{Al}_{73}\text{Mn}_{27-x}\text{Fe}_x$  as a function of temperature.

ceptibility is presented in Fig. 3. We can conclude that  $R_H$  is almost linear in  $\chi$  and this enables us to estimate the values for  $R_0$  and  $R_S$  which are listed in Table 1. We expect the temperature dependence of  $R_0$  to be negligible for the materials with the metallic conductivity.

It is generally accepted [6] that in systems with a high resistivity the anomalous Hall effect is dominated by a side-jump mechanism *i.e.* by the lateral displacement which electrons undergo during scattering in the presence of the spin-orbit interaction. In this case the anomalous Hall coefficient  $R_S$  should be proportional to the square of the resistivity. Skew scattering yields  $R_S \propto \rho$  and is expected to be dominant in materials with small resistivity. We suppose that in our materials  $R_S \propto \rho^2$  holds. The Electrical resistivity of  $\text{Al}_{73}\text{Mn}_{27-x}\text{Fe}_x$  samples as a function of temperature is shown in Fig. 4. The values of the resistivity are rather high and the temperature dependence of the resistivity, although small, is not negligible.

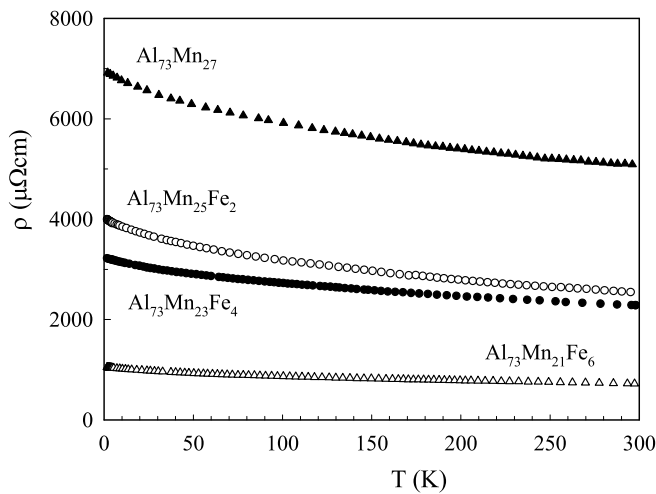
In Fig. 5 we plotted  $R_H (\chi \cdot \rho^2)$  (arbitrarily normalized to room-temperature values  $\rho_{RT}$ ). The  $R_H (\chi)$  and  $R_H (\chi \cdot \rho^2)$  plots are similar. Here it may be argued that, with the exception of the “pure”  $\text{Al}_{73}\text{Mn}_{23}$  alloy, the  $R_H(\chi)$  plots exhibit a small concave curvature while the  $R_H$



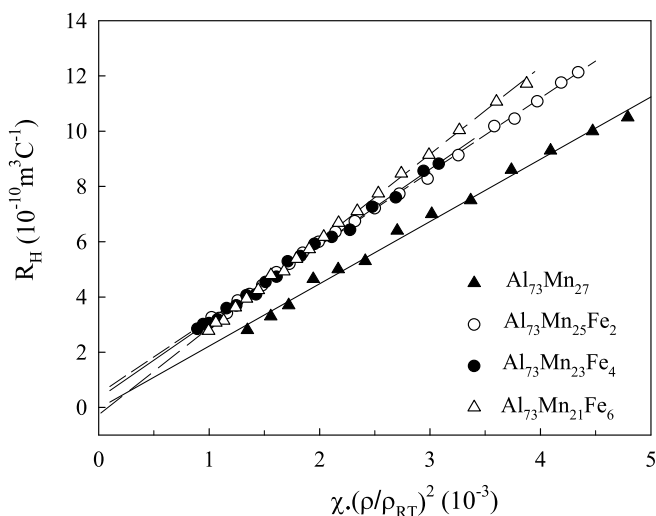
**Fig. 3.** The Hall coefficient of  $\text{Al}_{73}\text{Mn}_{27-x}\text{Fe}_x$  as a function of paramagnetic susceptibility.

**Table 1.** Normal Hall coefficient  $R_0$  and the anomalous Hall coefficient  $R_S$  deduced from  $R_H(\chi)$  and  $R_H(\chi \cdot (\rho/\rho_{RT})^2)$  plots.

Alloy composition	$R_0$ ( $10^{-10}$ m <sup>3</sup> C <sup>-1</sup> )		$R_S$ ( $10^{-7}$ m <sup>3</sup> C <sup>-1</sup> )	
	from $\chi$	from $\chi\rho^2$	from $\chi$	from $\chi\rho^2$
Al <sub>73</sub> Mn <sub>27</sub>	-1.9	0.0	3.5	2.3
Al <sub>73</sub> Mn <sub>25</sub> Fe <sub>2</sub>	-2.4	0.5	5.3	2.7
Al <sub>73</sub> Mn <sub>23</sub> Fe <sub>4</sub>	-1.6	0.3	4.8	2.7
Al <sub>73</sub> Mn <sub>21</sub> Fe <sub>6</sub>	-2.7	-0.3	5.5	3.1

**Fig. 4.** Temperature-dependent electrical resistivity of Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub>.

$(\chi \cdot \rho^2)$  plots, exhibit a better linearity. However, the most important difference between Figs. 3 and 5, is the shift of the intercept on the  $R_H$  axis from negative values of about  $-2 \times 10^{-10}$  m<sup>3</sup> C<sup>-1</sup> to positive values or zero. The results deduced for  $R_0$  from both  $R_H(\chi)$  and  $R_H(\chi \cdot \rho^2)$  plots are summarized in Table 1. We recall that the effective number of electrons for the case of a single band that correspond to  $R_0 = -1 \times 10^{-10}$  m<sup>3</sup> C<sup>-1</sup> is equal to  $0.6 \times 10^{23}$  cm<sup>-3</sup> and is characteristic for metals. However, the absolute values of  $R_0$  lower than  $-1 \times 10^{-10}$  m<sup>3</sup> C<sup>-1</sup> do not imply a higher carrier concentration but rather a mutual cancellation of the contributions of the electron-

**Fig. 5.** The Hall coefficient of Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub> as a function of  $\chi \cdot (\rho/\rho_{RT})^2$ .

like and hole-like regions of the Fermi surface. Table 1 also summarizes the values found for the anomalous Hall coefficient  $R_S$ . The values deduced from the  $R_H$  vs.  $\chi \cdot \rho^2$  plots are the room temperature values. We emphasize that the anomalous Hall coefficient of all samples is rather large, which is certainly due to their high resistivity. The  $R_S$  values are close to those determined for  $T$ -Al<sub>80</sub>Mn<sub>20</sub> and  $T$ -Al<sub>78</sub>Mn<sub>22</sub> phases ( $3.2$  and  $4.8 \times 10^{-7}$  m<sup>3</sup> C<sup>-1</sup>) [7]. Recently, for quasicrystalline icosahedral Al<sub>70.4</sub>Pd<sub>20.8</sub>Mn<sub>8.8</sub> a high value  $R_S = 1.8 \times 10^{-5}$  m<sup>3</sup> C<sup>-1</sup> has been reported [8]. For comparison, in amorphous ferromagnetic alloys with resistivities around  $150 \mu\Omega$  cm,  $R_S$  is usually of the order  $10^{-8}$  m<sup>3</sup> C<sup>-1</sup> [6]. However, the fact that in our alloys  $R_S$  practically remains the same in the samples whose resistivity differs almost for an order of magnitude may contain meaningful informations on the simultaneous changes in the electronic structure that cancel out the impact of the resistivity onto the anomalous Hall coefficient. The above discussion does not contradict our proposition about the temperature dependence of  $R_S$ .

## Conclusion

The lower bound of the normal Hall coefficient in Mn rich Al<sub>73</sub>Mn<sub>27-x</sub>Fe<sub>x</sub> alloys is determined to be  $-2 \times 10^{-10}$  m<sup>3</sup> C<sup>-1</sup>. This value corresponds to the high metallic conduction electron density of the order  $10^{23}$  cm<sup>-3</sup>. When the possible dependence of  $R_S$  on temperature, due to the temperature dependence of the electrical resistivity, is taken into account, the values for  $R_0$  are about zero. This would imply the mutual cancellation of contributions from the electron-like and hole-like regions of the Fermi surface. In either case no significant composition dependence of  $R_0$  was deduced. The anomalous Hall coefficient of these alloys is very large, due to the high resistivity, and of the order of  $10^{-7}$  m<sup>3</sup> C<sup>-1</sup>. However, while the resistivity of these alloys significantly depends on the composition, the anomalous Hall coefficient does not. This indicates that, upon the alloying, a number of changes in the electronic properties are taking place simultaneously which cancel out the impact of the resistivity.

*Acknowledgments.* This work was done within the activities of the 6th Framework EU Network of Excellence “Complex Metallic Alloys” (Contract No. NMP3-CT-2005-500140), and as been supported in part by the Ministry of Science, Education and Sports of Republic Croatia through the Research Project No. 035-0352826-2848.

## References

- [1] Klein, H.; Boudard, M.; Audier, M.; de Boissieu M.; Vincent, H.; Beraha, L.; Duneau, M.: The T-Al<sub>3</sub>(Mn,Pd) quasicrystalline approximant: chemical order and phason defects. *Philos. Mag. Lett.* **75** (1997) 197–208.
- [2] Balanetsky, S.; Meisterernst, G.; Heggen, M.; Feuerbacher, M.: Reinvestigation of the Al–Mn–Pd alloy system in the vicinity of T- and R-phases. *Intermetallics* **16** (2008) 71–87.
- [3] Hurd, C. M.: *The Hall Effect in Metals and Alloys*. Plenum, New York 1972.
- [4] Sinitsyn, N. A.: Semiclassical theories of the anomalous Hall effect. *J. Phys.: Condens. Matter* **20** (2008) 023201.
- [5] Dolinšek, J.; Slanovec, J.; Jagličić, Z.; Heggen, M.; Balanetsky, S.; Feuerbacher, M.; Urban, K.: Broken ergodicity, memory

- effect, and rejuvenation in Taylor-phase and decagonal  $\text{Al}_3(\text{Mn}, \text{Pd}, \text{Fe})$  complex intermetallics. *Phys. Rev. B* **77** (2008) 064430.
- [6] McGuire, T. R.; Gambino, R. J.; O'Handley, R. C.: In: *The Hall Effect and Its Applications*, (Eds. C. L. Chien, C. R. Westgate), pp. 137–200. Plenum, New York 1980.
- [7] Gozlan, A.; Berger, C.; Fourcaudot G.; Omari R.; Lasjaunias, J. C.; Préjean, J. J.: Anomalous Hall effect related to the magnetization in pure decagonal AlMn phases. *Phys. Rev. B* **44** (1991) 575–583.
- [8] Poddar, A.; Das, S.; Plachke, D.; Carstanjen, H. D.: Electrical transport, magnetic and thermal properties of icosahedral AlPdMn quasicrystals. *J. Magn. Magn. Mater.* **300** (2006) 263–272.