



Hall Effect of the Triclinic $\text{Al}_{73}\text{Mn}_{27}$ and T- $\text{Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ ($0 \leq x \leq 6$) Complex Metallic Alloys*

Jovica Ivkov,^{a,**} Denis Stanić,^a Zvonko Jagličić,^b
 Janez Dolinšek,^c Marc Heggen,^d and Michael Feuerbacher^d

^aLaboratory for the Physics of Transport Phenomena, Institute of Physics, Bijenička c. 46,
 P. O. Box 304, HR-10001 Zagreb, Croatia

^bInstitute of Mathematics, Physics and Mechanics & Faculty of Civil and Geodetic Engineering,
 University of Ljubljana, Jadranska 19, SI-1000 Ljubljana, Slovenia

^cJožef Stefan Institute, University of Ljubljana, Jamova 39, SI-1000 Ljubljana, Slovenia

^dInstitut für Festkörperforschung, Forschungszentrum Jülich, Jülich D-52425, Germany

RECEIVED JULY 23, 2008; ACCEPTED SEPTEMBER 4, 2009

Abstract. The Hall coefficient, R_H , of the triclinic $\text{Al}_{73}\text{Mn}_{27}$ and Taylor-phase $\text{Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ ($x = 0, 2, 4$ and 6) complex metallic alloys has been measured from 90 to 400 K. The Hall coefficients of all the samples are positive and they decrease strongly with the increase of temperature, T . For the separation of the normal, R_0 , and anomalous, R_S , Hall coefficient the results for the paramagnetic susceptibility, $\chi(T)$, and electrical resistivity, $\rho(T)$, have been used. The well defined linearity of the R_H vs. $\chi(T) \cdot \rho^2(T)$ plots confirms the assumption that in these materials R_H is dominated by spin-orbit interaction. The values deduced from the R_H vs. χ and R_H vs. $\chi \cdot \rho^2$ plots in T-AlMnPd phases, fall between $-2 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ and 0 for R_0 , and are about $5 \times 10^{-7} \text{ m}^3 \text{ C}^{-1}$ for R_S . The values deduced from the R_H vs. $\chi \cdot \rho^2$ plots in the triclinic $\text{Al}_{73}\text{Mn}_{27}$ alloy are about $-15 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ for R_0 , and about $1.5 \times 10^{-5} \text{ m}^3 \text{ C}^{-1}$ for R_S .

Keywords: complex metallic alloys, Hall effect

INTRODUCTION

Al-Mn based systems contain 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 [0 1 0] direction. Along this axis pentagonal columnar clusters are formed.¹ The unit cell of the T phase contains 156 atoms with many of the sites with either fractional occupation or mixed Al/Mn occupation, so that a considerable inherent chemical disorder exists on the lattice. As a part of the systematic investigation of the transport and magnetic properties of T-Al,Mn based alloys and related quasicrystals, here we present the results of the Hall-effect measurement on T- $\text{Al}_{73}\text{Mn}_{27}$ and its solid solutions T- $\text{Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ ($x = 2, 4$ and 6) alloys, and on the triclinic $\text{Al}_{73}\text{Mn}_{27}$ phase.

EXPERIMENTAL

Polycrystalline samples were produced from the consti-

tuent elements by levitation induction melting in a water-cooled copper crucible under argon atmosphere. Annealing at 900 °C for 312 h and subsequent quenching into water yields triclinic $\text{Al}_{73}\text{Mn}_{27}$ and T- $\text{Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ ($x = 2, 4$ and 6) solid solutions. Pure binary T- $\text{Al}_{73}\text{Mn}_{27}$ phase was prepared by: annealing at 900 °C for 312 h, additional annealing at 930 °C for 3 h, and quenching into water.² The Hall-effect measurements were performed by a standard AC technique and by a five-point method in magnetic fields, B , up to 1 T in the temperature interval from 90 to 370 K. The temperature-dependent magnetic susceptibility, χ , was investigated in the temperature interval between 2 and 300 K, using a Quantum Design SQUID magnetometer, equipped with a 5 T magnet. Electrical resistivity was measured between 1.5 and 300 K using the standard four-probe DC technique.

RESULTS AND DISCUSSION

In the temperature interval explored and for the magnetic

* Presented at the EU Workshop "Frontiers in Complex Metallic Alloys", Zagreb, October 2008.

Dedicated to Professor Boran Leontić on the occasion of his 80th birthday.

** Author to whom correspondence should be addressed. (E-mail: ivkov@ifs.hr)

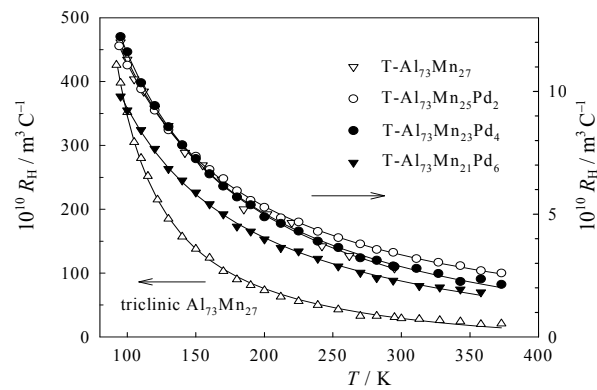


Figure 1. The Hall coefficient of triclinic $\text{Al}_{73}\text{Mn}_{27}$ and $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ as a function of temperature.

fields up to 1 T the Hall resistivity, ρ_{H} , of all the samples is a linear function of the magnetic field and therefore we present the results for the Hall coefficient, $R_{\text{H}} = \rho_{\text{H}}/B$, only. The Hall coefficients of the triclinic $\text{Al}_{73}\text{Mn}_{27}$ and $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ samples as a function of temperature are displayed in Figure 1. The shape of R_{H} vs. T curves indicates that the samples are paramagnetic, and that the anomalous magnetic contribution to the Hall effect is dominant.

Generally, in magnetic materials the Hall resistivity follows the empirical relation³

$$\rho_{\text{H}} = R_0 \cdot B + \mu_0 M \cdot R_S, \quad (1)$$

where R_0 and R_S are the ordinary and anomalous (spontaneous) 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. The anomalous Hall effect, on the other hand, 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

In Figure 2 we present the zero-field-cooled magnetic susceptibility of the $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ and triclinic $\text{Al}_{73}\text{Mn}_{27}$ samples as a function of temperature. The magnetic properties of the $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ alloys have been investigated in more detail.⁵ They belong to the class of magnetically frustrated spin systems with freezing temperatures well below those over which the results were used in this work for the separation of R_0 and R_S .

The high-temperature susceptibility of all alloys follows the Curie-Weiss ($\chi = \chi_0 + A/(T - \theta)$) law with negative Curie-Weiss temperature θ , and temperature independent $\chi_0 < 1 \times 10^{-5}$ which we neglect in the further discussion. As χ is small and (for $T > 90$ K) of the order 10^{-3} we ignore the effects of demagnetizing fields

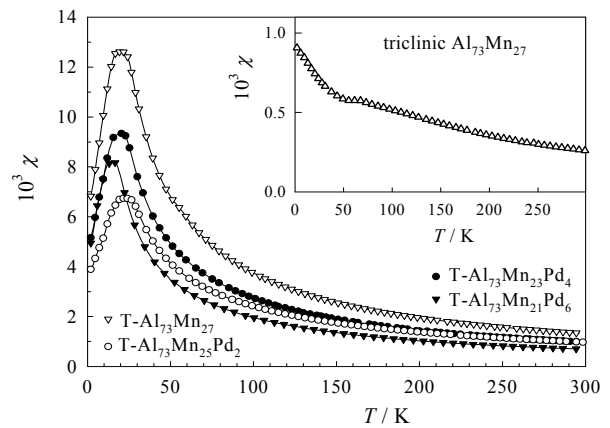


Figure 2. Zero-field-cooled magnetic susceptibility of $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ and triclinic $\text{Al}_{73}\text{Mn}_{27}$ as a function of temperature.

and replace $\mu_0 M$ with $\chi \cdot B$. Equation (1) then yields

$$R_{\text{H}} = R_0 + \chi \cdot R_S. \quad (2)$$

Before we present R_{H} as a function of $\chi(T)$ we briefly discuss the temperature dependence of R_0 and R_S . The temperature dependence of R_0 is expected to be negligible, as in materials with metallic conductivity. For the anomalous Hall coefficient, it is generally accepted⁶ that in systems with a high resistivity, ρ , R_S is dominated by the side-jump mechanism *i.e.* by the lateral displacement which electrons undergo during the scattering in the presence of spin-orbit interaction.⁷ In this case R_S is proportional to ρ^2 . The electrical resistivity of the triclinic $\text{Al}_{73}\text{Mn}_{27}$ and $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ alloys is shown in Figure 3 as a function of temperature. The values of ρ are rather high and its temperature dependence is not negligible, especially in the triclinic $\text{Al}_{73}\text{Mn}_{27}$ alloy.

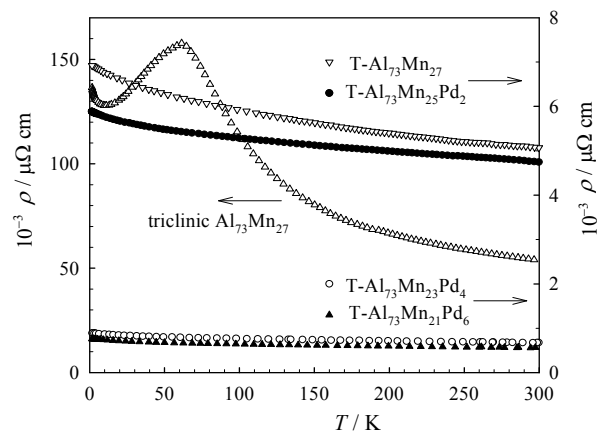


Figure 3. The electrical resistivity of triclinic $\text{Al}_{73}\text{Mn}_{27}$ and $\text{T-Al}_{73}\text{Mn}_{27-x}\text{Pd}_x$ as a function of temperature.

In Figure 4 we plotted R_H vs. χ and R_H vs. $\chi \cdot \rho^2$ (arbitrarily normalized to the room temperature values $\rho_{r.t.}$) for the triclinic $Al_{73}Mn_{27}$, while in Figure 5 we plotted the same for two $T-Al_{73}Mn_{27-x}Pd_x$ alloys. We see that $R_H(T)$ is a linear function of $\chi(T) \cdot \rho^2(T)$ and this enables the separation of the coefficients R_0 and R_S (295 K) as the intercepts and the slopes of the straight lines.³ Because of the small temperature dependence of ρ in the $T-Al_{73}Mn_{27-x}Pd_x$ alloys, the R_H vs. χ plots for these alloys seem (within the experimental error) linear too. A similar dependence of $R_H(T)$ on χ and $\chi \cdot \rho^2$ was already found in the $T-Al_{73}Mn_{27-x}Fe_x$ ($x \leq 6$) alloys.⁸

The results deduced for R_0 and R_S are summarized in Table 1. We recall that the number of electrons in a single band that yields $R_0 = -1 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ is equal to $0.6 \times 10^{23} \text{ cm}^{-3}$ and is characteristic for metals.

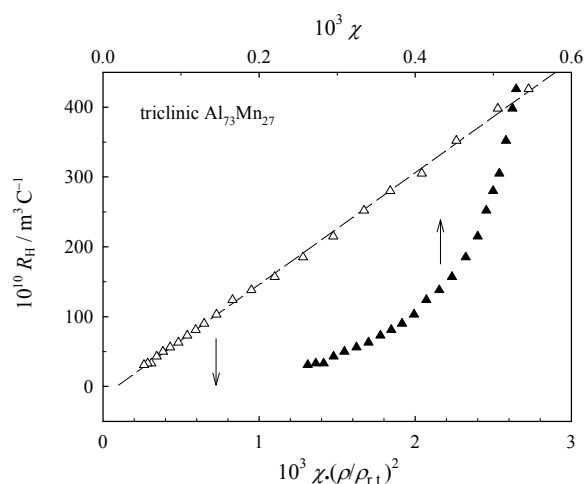


Figure 4. The Hall coefficient of triclinic $Al_{73}Mn_{27}$ as a function of paramagnetic susceptibility χ , and as a function of $\chi \cdot (\rho/\rho_{r.t.})^2$.

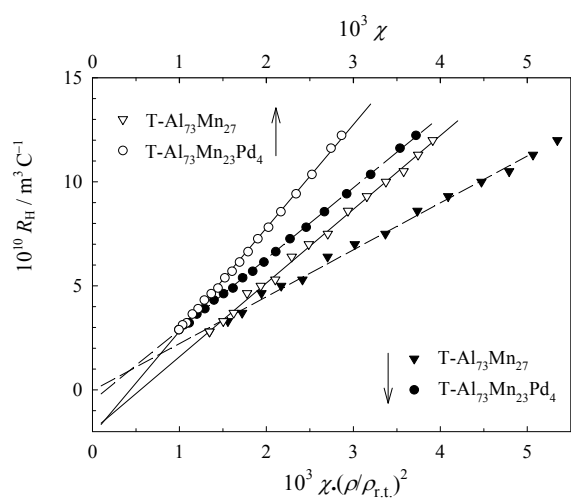


Figure 5. The Hall coefficient of $T-Al_{73}Mn_{27-x}Pd_x$ as a function of paramagnetic susceptibility χ , and as a function of $\chi \cdot (\rho/\rho_{r.t.})^2$.

Table 1. The normal Hall coefficient R_0 and the anomalous Hall coefficient R_S (295 K) deduced from R_H vs. χ , and R_H vs. $\chi \cdot (\rho/\rho_{r.t.})^2$ plots

Alloy composition	$10^{10} R_0 / \text{m}^3 \text{ C}^{-1}$		$10^7 R_S / \text{m}^3 \text{ C}^{-1}$	
	from χ	from $\chi \cdot \rho^2$	from χ	from $\chi \cdot \rho^2$
$Al_{73}Mn_{27}$	–	–14	–	160
$T-Al_{73}Mn_{27}$	–1.6	0.3	4.8	2.7
$T-Al_{73}Mn_{25}Pd_2$	–2.1	–0.6	5.4	3.9
$T-Al_{73}Mn_{23}Pd_4$	–2.1	–0.5	5.0	3.4
$T-Al_{73}Mn_{21}Pd_6$	–1.7	–0.3	5.6	3.7

The absolute values of R_0 lower than $1 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ do not imply a higher carrier concentration but the mutual cancellation of the contributions from the electron-like and hole-like regions of the Fermi surface.

The anomalous Hall coefficient of all the samples is rather large which is due to their high resistivity. The values of R_S of the $T-Al_{73}Mn_{27-x}Pd_x$ samples are close to those determined for the $T-Al_{80}Mn_{20}$ and $T-Al_{78}Mn_{22}$ phases (3.2 and $4.8 \times 10^{-7} \text{ m}^3 \text{ C}^{-1}$ respectively).⁹ Recently, for the quasicrystalline icosahedral $Al_{70.4}Pd_{20.8}Mn_{8.8}$ a high value of $R_S = 1.8 \times 10^{-5} \text{ m}^3 \text{ C}^{-1}$, close to those obtained for the triclinic $Al_{73}Mn_{27}$ alloy, has been reported.¹⁰

CONCLUSION

Hall coefficient $R_H(T)$ of the triclinic $Al_{73}Mn_{27}$ and Taylor (T) $Al_{73}Mn_{27-x}Pd_x$ ($0 \leq x \leq 6$) complex metallic alloys is a linear function of $\chi(T) \cdot \rho^2(T)$. This enables the separation of the normal and anomalous Hall coefficients R_0 and R_S respectively, and at the same time confirms the proposition that in these materials the Hall effect is dominated by the spin-orbit interaction and by the side-jump effect. The anomalous Hall coefficient of all the investigated alloys is very large, due to the high resistivity of these alloys. In the $T-Al_{73}Mn_{27-x}Pd_x$ samples, R_S is of the order $10^{-7} \text{ m}^3 \text{ C}^{-1}$ and in the triclinic $Al_{73}Mn_{27}$ sample it is of the order $10^{-5} \text{ m}^3 \text{ C}^{-1}$. The conduction electron densities estimated from R_0 are characteristic for metals and are of the order 10^{23} cm^{-3} in the $T-Al_{73}Mn_{27-x}Pd_x$ samples, and of the order 10^{22} in the triclinic $Al_{73}Mn_{27}$ sample.

Acknowledgements. 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 has been supported in part by the Ministry of Science, Education and Sports of the Republic of Croatia through the Research Project No. 035-0352826-2848. We thank A. Smontara for the fruitful discussion.

REFERENCES

1. H. Klein, M. Boudard, M. Audier, M. de Boissieu, H. Vincent, L. Beraha, and M. Duneau, *Philos. Mag. Lett.* **75** (1997) 197–208.
2. S. Balanetsky, G. Meisterernst, M. Heggen, and M. Feuerbacher, *Intermetallics* **16** (2008) 71–87.
3. C. M. Hurd, *The Hall Effect in Metals and Alloys*, Plenum, New York, 1972.
4. N. A. Sinitsyn, *J. Phys.: Condens. Matter* **20** (2008) 023201 1–17.
5. J. Dolinšek, J. Slanovec, Z. Jagličić, M. Heggen, S. Balanetsky, M. Feuerbacher, and K. Urban, *Phys. Rev. B* **77** (2008) 064430 1–18.
6. T. R. McGuire, R. J. Gambino, and R. C. O’Handley, in: C. L. Chien and C. R. Westgate (Eds.), *The Hall Effect and Its Applications*, Plenum, New York, 1980, pp. 137–200.
7. L. Berger, *Phys. Rev. B* **2** (1970) 11, 4559–4566.
8. D. Stanić, J. Ivkov, A. Smontara, Z. Jagličić, J. Dolinšek, M. Heggen, and M. Feuerbacher, *Z. Kristallogr.* **224** (2009) 49–52.
9. A. Gozlan, C. Berger, G. Fourcaudot, R. Omari, J. C. Lasjaunias, and J. J. Préjean, *Phys. Rev. B* **44** (1991) 2, 575–583.
10. A. Poddar, S. Das, D. Plachke, and H. D. Carstanjen, *J. Magn. Mater.* **300** (2006) 263–272.

SAŽETAK

Hallov efekt u triklinskim $Al_{73}Mn_{27}$ i $T-Al_{73}Mn_{27-x}Pd_x$ ($0 \leq x \leq 6$) kompleksnim metalnim slitinama

Jovica Ivkov,^a Denis Stanić,^a Zvonko Jagličić,^b Janez Dolinšek,^c
Marc Heggen^d i Michael Feuerbacher^d

^aLaboratorij za fiziku transportnih svojstava, Institut za fiziku, Bijenička c. 46,
P. P 304, HR-10001 Zagreb, Hrvatska

^bInstitute of Mathematics, Physics and Mechanics & Faculty of Civil and Geodetic Engineering,
University of Ljubljana, Jadranska 19, SI-1000 Ljubljana, Slovenia

^cJožef Stefan Institute, University of Ljubljana, Jamova 39, SI-1000 Ljubljana, Slovenia

^dInstitut für Festkörperforschung, Forschungszentrum Jülich, Jülich D-52425, Germany

Prikazani su rezultati mjerenja Hallovog koeficijenta, R_H , triklinske $Al_{73}Mn_{27}$ slitine i Taylorovih (T) $Al_{73}Mn_{27-x}Pd_x$ ($x = 0, 2, 4$ i 6) faza kompleksnih metalnih slitina u intervalu temperatura od 90 do 400 K. Hallov koeficijent svih slitina je pozitivan i brzo opada s porastom temperature T . Za odijeljivanje normalnog, R_0 , i anomalnog, R_S , Hallovog koeficijenta korišteni su rezultati za paramagnetsku susceptibilnost, $\chi(T)$, i električnu otpornost, $\rho(T)$. Dobro definirana linearna ovisnost R_H o $\chi(T) \cdot \rho^2(T)$ potvrđuje pretpostavku da je u tim materijalima R_H pretežito određen spin-orbit interakcijom. Vrijednosti određene iz R_H vs. χ i R_H vs. $\chi \cdot \rho^2$ prikaza za $T-AlMnPd$ slitine, padaju između $-2 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ i 0 za R_0 , a iznose oko $5 \times 10^{-7} \text{ m}^3 \text{ C}^{-1}$ za R_S . Vrijednosti određene iz R_H vs. $\chi \cdot \rho^2$ prikaza za triklinsku $Al_{73}Mn_{27}$ slitinu iznose oko $-15 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ za R_0 , i oko $1.5 \times 10^{-5} \text{ m}^3 \text{ C}^{-1}$ za R_S .