CROATICA CHEMICA ACTA CCACAA, ISSN 0011-1643, e-ISSN 1334-417X Croat. Chem. Acta 83 (1) (2010) 7–10. CCA-3381

Original Scientific Paper

Hall Coefficient of the Y-Al-Ni-Co Decagonal Approximant*

Denis Stanić,^{a,**} Jovica Ivkov,^a Janez Dolinšek,^b and Peter Gille^c

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

^bJožef Stefan Institute, University of Ljubljana, Jamova 39, SI-1000 Ljubljana, Slovenia ^cLudwig-Maximilians-Universität München, Department of Earth and Environmental Sciences, Crystallography Section, Theresienstrasse 41, D-80333 München, Germany

RECEIVED AUGUST 6, 2008; ACCEPTED FEBRUARY 16, 2009

Abstract. We have investigated anisotropic Hall coefficient of Y-Al-Ni-Co decagonal approximant of the $Al_{76}Co_{22}Ni_2$ composition. The crystalline-direction-dependent studies were performed along three orthogonal directions a^* , b and c of the Y-Al-Ni-Co unit cell, where (a,c) monoclinic atomic planes are stacked along the perpendicular b direction. Hall coefficient R_H exhibits pronounced anisotropy, while at the same time the magnetic field in a given crystalline direction yields the same R_H for the current along the other two crystalline directions in the perpendicular plane. The observed anisotropy is studied in terms of the anisotropic structure of the Y-Al-Ni-Co phase and Fermi surface. The results are compared to the anisotropy of the Hall coefficient of the d-Al-Ni-Co decagonal quasicrystal reported in literature.

Keywords: complex metallic alloys, decagonal approximants, Y-Al-Ni-Co, Hall effect

INTRODUCTION

Decagonal quasicrystals (d-QCs) can be structurally displayed as a periodic stack of quasiperiodic atomic planes, so that d-QCs are two-dimensional quasicrystals, whereas they are periodic crystals in a direction perpendicular to the quasiperiodic planes. This makes d-QCs especially suitable for studying the influence of quasiperiodicity on the transport properties of the material, as a given property may be investigated on the same sample along the quasiperiodic (Q) and periodic (P) directions. A striking example of the anisotropic nature of d-QCs is their electrical resistivity, which shows positive temperature coefficient with metallic values along the P direction $\rho_{\rm P}^{\rm r.t.} \approx 40 \ \mu\Omega$ cm in d-Al-Cu-Co and d-Al-Ni-Co,¹ whereas the resistivity in the quasiperiodic plane is considerably larger $\rho_Q^{r.t.}\approx 330~\mu\Omega$ cm and exhibits a negative temperature coefficient. The degree of anisotropy is related to the structural details of a particular decagonal phase, depending on the number of quasiperiodic layers in one periodic unit.²⁻⁴ The most anisotropic case belongs to the phases with just two layers, realized in d-Al-Ni-Co and d-Al-Cu-Co, where the periodicity length along the periodic axis is about 0.4 nm and the resistivity ratio at room temperature amounts typically to $\rho_0/\rho_P \approx 6-10^{.1,5-8}$ Other d-phases contain more quasiperiodic layers in a periodic unit and show smaller anisotropies. The basic question is whether the observed anisotropy is a consequence of complex local atomic order on the scale of near-neighbor atoms with no direct relationship to the quasiperiodicity. Approximant phases are characterized by large unit cells, which periodically repeat themselves in space, but the structure of the unit cell resembles closely to d-QCs. Atomic layers are stacked periodically and the periodicity lengths along the stacking direction are almost identical to those along the periodic direction of d-QCs. Moreover, atomic planes of approximants and d-QCs show locally similar quasiperiodic patterns, so that their structures on the scale of near-neighbor atoms closely resemble each other. Recently, the anisotropic transport properties (resistivity, Hall coefficient, ...) measured along three orthogonal crystalline directions, have been reported for the Al₄(Cr,Fe) complex metallic alloy with composition $Al_{80}Cr_{15}Fe_5$.^{9,10} The in-plane electrical resistivity of this compound exhibits nonmetallic behavior with negative temperature coefficient, while the resistivity along the stacking direction shows positive temperature coefficient. The anisotropy of the resistivity along the three orthorhombic directions is a consequence



^{*} 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.

^{**} Authors to whom correspondence should be addressed. (E-mail: dstanic@ifs.hr)

of local atomic order on the scale of nearest neighbor atoms, pertinent to the specific structure of the Al₄(Cr,Fe) phase, with no direct relationship to the intermediate-scale quasiperiodic patterns in the structure. Hall coefficient $R_{\rm H}$, shows a metallic behavior in all cases and exhibits pronounced anisotropy. As a part of the systematic investigation of the transport properties, we here report a study of the anisotropic Hall coefficient of a complex metallic alloy $Al_{13-x}(Co_{1-v}Ni_v)_4$, known also as the Y-Al-Ni-Co phase, which is a monoclinic approximant of the decagonal phase with two atomic layers within one periodic unit along the stacking direction and a relatively small unit cell, comprising 32 atoms. Due to the considerably smaller unit cell, the structure of Y-Al-Ni-Co is relatively simple as compared to the much higher complexity of the aforementioned Al₄(Cr,Fe) phase with 306 atoms/unit cell. The measurements of the anisotropy in Y-Al-Ni-Co thus allow a comparison of this simple decagonal approximant to the Al₄(Cr,Fe) phase,^{9,10} a decagonal approximant with a considerably higher structural complexity as well as a comparison to the closely related d-Al-Ni-Co quasicrystal.¹

EXPERIMENTAL

The single crystal, used in our study, was grown from an incongruent Al-rich melt of initial composition Al_{81.9}Co_{14.5}Ni_{3.6} by the Czochralski method. The composition of the crystal was Al₇₆Co₂₂Ni₂ and its structure matched well to the monoclinic unit cell of the Zhang et *al.* model¹² (who studied the composition $Al_{75}Co_{22}Ni_3$). In order to perform crystalline-direction-dependent studies of Hall coefficient, we have cut from the ingot three bar-shaped samples of dimensions $2 \times 2 \times 6 \text{ mm}^3$, with their long axes along three orthogonal directions. The long axis of the first sample was along [0 1 0] direction (designated in what follows as b), which corresponds to the periodic direction in the related d-Al-Ni-Co quasicrystal. The (a,c) monoclinic plane corresponds to the quasiperiodic plane in d-QCs and the second sample was cut with its long axis along [0 0 1] (c) direction, whereas the third one was cut along the direction perpendicular to the (b,c) plane. This direction is designated as a^* (it lies in the monoclinic plane at an angle of 26° with respect to *a* and perpendicular to *c*). For each sample, the orientation of the other two crystalline directions was also known. The so-prepared samples enabled us to determine the anisotropic Hall coefficient along the three orthogonal directions of the investigated monoclinic Al₇₆Co₂₂Ni₂, abbreviated henceforth as Y-Al-Ni-Co. The Hall effect measurements were performed by the five-point method using standard ac technique in magnetic fields up to 10 kOe. The current through the samples was in the range of 10-50 mA. The



Figure 1. Temperature-dependent electrical resistivity of Y-Al-Ni-Co along three orthogonal crystalline directions a^* , b and c.

measurements were performed in the temperature interval from 90 to 370 K.

RESULTS AND DISCUSSION

Electrical resistivity $\rho(T)$, measured in all three crystalline directions, is displayed in Figure 1. The resistivity along the direction perpendicular to the atomic monoclinic plane ρ_b is considerably smaller than in-plane resistivities ρ_{a^*} and ρ_c by a factor of around 3, while the two in-plane resistivities are much closer, $\rho_{a^*}/\rho_c \approx 1.3$ (Table 1). The obtained resistivity values reveal that Y-Al-Ni-Co is good electrical conductor along all three crystalline directions while in-plane resistivity of d-Al-Ni-Co quasicrystal is qualitatively different, showing nonmetallic temperature behaviour, and about 10 times larger value at room temperature than the resistivity along the periodic direction.^{5,13}

The temperature-dependent Hall coefficient $R_{\rm H} = E_y/j_x B_z$ is displayed in Figure 2. In order to determine the anisotropy of $R_{\rm H}$, three sets of experiments were performed with the current along the long axis of each sample (thus along a^* , b and c, respectively), whereas the magnetic field was directed along each of the other two orthogonal crystalline directions, making six experiments altogether. For all combinations of directions,

Table 1. Anisotropic resistivity data $\rho(T)$ of Y-Al-Ni-Co for various crystalline directions

Crystalline direction	$ ho^{ m r.t.}$ / $\mu\Omega$ cm	$ ho^{2\mathrm{K}}$ / $\mu\Omega\mathrm{cm}$
b	25	10
С	60	29
a^*	81	34



Figure 2. Anisotropic temperature-dependent Hall coefficient $R_{\rm H} = E_y/j_x B_z$ of Y-Al-Ni-Co for different combinations of directions of the current j_x and magnetic field B_z : (open triangles (j_b, B_a^*) and full triangles (j_c, B_a^*) ; open circles (j_{a^*}, B_c) and full circles (j_b, B_c) ; open squares (j_c, B_b) and full squares (j_{a^*}, B_b)). The superscript a^* , b or c on $R_{\rm H}$ denotes the direction of the magnetic field.

the $R_{\rm H}$ values are typical metallic values of the order of 10^{-10} m³ C⁻¹, showing weak temperature dependence. This temperature dependence shows tendency to disappear at higher temperatures. R_H's exhibit pronounced anisotropy with the following regularity: the six $R_{\rm H}$ sets of data form three groups of two practically identical $R_{\rm H}$ curves, where the magnetic field in a given crystalline direction yields the same $R_{\rm H}$ for the current along the other two crystalline directions in the perpendicular plane. Thus, identical Hall coefficients are obtained for combinations $R_{\rm H}^{a^*} = E_b/j_c B_{a^*}$ and $R_{\rm H}^{a^*} = E_c/j_b B_{a^*}$ (where additional superscript on the Hall coefficient denotes the direction of the magnetic field), being $R_{\rm H}^{a^*}$ (r.t.) = 8.5 × 10⁻¹⁰ m³ C⁻¹, $R_{\rm H}^{b} = E_{a^*}/j_c B_b$ and $R_{\rm H}^{b} = E_c/j_{a^*}B_b$ with $R_{\rm H}^{\ b}({\rm r.t.}) \approx 0$ and $R_{\rm H}^{\ c} = E_b/j_a * B_c$ and $R_{\rm H}^{\ c} = E_{a^*}/j_b B_c$ with $R_{\rm H}^{\ c}({\rm r.t.}) \approx 4.5 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$. The anisotropic $R_{\rm H}$ values are also given in Table 2. The two rather high positive values $R_{\rm H}^{a^*}$ and $R_{\rm H}^{c}$ for the field lying in the (*a*,*c*) atomic plane and the almost zero value of $R_{\rm H}^{\ b}$ for the field along the perpendicular b direction (periodic direction in d-QCs) reflect strong anisotropy of the Fermi surface¹⁴ that consists mostly of hole-like parts, whereas electron-like and hole-like parts are of comparable importance for the field perpendicular to the (a,c) plane. Comparing the $R_{\rm H}$ anisotropy of Y-Al-Ni-Co to the d-QCs, we find qualitative similarity. For d-QCs, universality of the Hall-effect anisotropy was reported for the d-Al-Ni-Co, d-Al-Cu-Co and d-Al-Si-Cu-Co,6,8 where $R_{\rm H} > 0$ for the field lying in the quasiperiodic plane (corresponding to the in-plane coefficients $R_{\rm H}^{a^*}$, $R_{\rm H}^{c} > 0$ of Y-Al-Ni-Co), whereas $R_{\rm H}$ changes sign to negative for

Table 2. Anisotropic Hall coefficient $R_{\rm H} = E_y / j_x B_z$ of Y-Al-Ni-Co for various directions of the current

Current direction (j_x)	Magnetic field direction (B_z)	$\frac{10^{10} R_{\rm H}^{\rm r.t.}}{\rm m^3 \ C^{-1}}$
<i>a*</i>	b	0.0
	С	4.6
b	<i>a</i> *	8.5
	С	4.3
с	<i>a</i> *	8.5
	b	-0.2

the field along the periodic direction (corresponding to $R_{\rm H}^{\ b} \approx 0$ of Y-Al-Ni-Co). The above d-QCs also exhibit similar weak temperature dependence of $R_{\rm H}$ as Y-Al-Ni-Co, so that there is a good analogy between d-QCs and their approximant Y-Al-Ni-Co.

CONCLUSION

We have studied Hall coefficient of the Y-Al-Ni-Co decagonal approximant. Hall coefficient $R_{\rm H}$ of Y-Al-Ni-Co exhibits pronounced anisotropy. The magnetic field in a given crystalline direction yields the same $R_{\rm H}$ for the current along the other two crystalline directions in the perpendicular plane. Two rather high positive values $R_{\rm H}^{a^*}$ and $R_{\rm H}^{c}$ for the field lying in the (*a*,*c*) atomic plane and the almost zero value of $R_{\rm H}^{\ b}$ for the field along the perpendicular b direction reflect strong anisotropy of the Fermi surface that consists mostly of hole-like parts, whereas electron-like and hole-like parts are of comparable importance for the field perpendicular to the (a,c)plane. There is a complete analogy between the $R_{\rm H}$ anisotropy of Y-Al-Ni-Co and d-QCs, where, in the latter case $R_{\rm H} > 0$ for the field lying in the quasiperiodic plane (corresponding to the in-plane coefficients $R_{\rm H}^{a^*}$, $R_{\rm H}^{\ c} > 0$ of Y-Al-Ni-Co), whereas $R_{\rm H}$ changes sign to negative for the field along the periodic direction (corresponding to $R_{\rm H}^{\ b} \approx 0$ of Y-Al-Ni-Co). Therefore, the results confirm the analogy between the anisotropic Hall coefficient of the quasiperiodic decagonal quasicrystal on one hand and the periodic decagonal approximants on the other hand. This suggests that the long-range quasiperiodicity of the structure is of marginal importance for the anisotropy, which originates from the complex local atomic order on the scale of nearestneighbor atoms.

Acknowledgements. This work has been 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 useful discussion.

REFERENCES

- S. Martin, A. F. Hebard, A. R. Kortan, and F. A. Thiel, *Phys. Rev. Lett.* 67 (1991) 719–722.
- 2. M. Krajčí and J. Hafner, Phys. Rev. B 58 (1998) 5378-5383.
- 3. G. T. de Laissardière and T. Fujiwara, *Phys. Rev. B* **50** (1994) 9843–9850.
- J. Dolinšek and A. Smontara, Anisotropic Physical Properties of Complex Metallic Alloys, in: Esther Belin-Ferre (Ed.), Complex Metallic Alloys: A Primer, London WC2H 9HE, England World Scientific Publishing (UK) Ltd., 2010, in press.
- T. Shibuya, T. Hashimoto, and S. Takeuchi, J. Phys. Soc. Jpn. 59 (1990) 1917–1920.
- Y. P. Wang and D. L. Zhang, *Phys. Rev. B* 49 (1994) 13204–13207.
- S. Y. Lin, X. M. Wang, L. Lu, D. L. Zhan, L. X. He, and K. X. Kuo, *Phys. Rev. B* 41 (1990) 9625–9627.

- D. L. Zhang, L. Lu, X. M. Wang, and S. Y. Lin, *Phys. Rev. B* 41 (1990) 8557–8559.
- J. Dolinšek, P. Jeglič, M. Komelj, S. Vrtnik, A. Smontara, I. Smiljanić, A. Bilušić, J. Ivkov, D. Stanić, E. S. Zijlstra, B. Bauer, and P. Gille, *Phys. Rev. B* 76 (2007) 174207-1-174207-13.
- J. Dolinšek, S. Vrtnik, A. Smontara, M. Jagodič, Z. Jagličić, B. Bauer, and P. Gille, *Philos. Mag.* 88 (2008) 2145–2153.
- 11. B. Zhang, V. Gramlich, and W. Steurer, Z. Kristallogr. 210 (1995) 498–503.
- 12. Y. P. Wang, D. L. Zhang, and L. F. Chen, *Phys. Rev. B* 48 (1993) 10542–10545.
- J. Dolinšek, O. S. Barišić, and A. Smontara, Z. Kristallogr. (2008) 224 (2009) 64–66.
- M. Komelj, J. Ivkov, A. Smontara, P. Gille, and J. Dolinšek, *Solid State Commun.* 149 (2009) 515–518.

SAŽETAK

Hallov koeficijent dekagonalnog aproksimanta Y-Al-Ni-Co

Denis Stanić,^a Jovica Ivkov,^a Janez Dolinšek^b i Peter Gille^c

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

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

`Ludwig-Maximilians-Universit"at M"unchen, Department of Earth and Environmental Sciences,

Crystallography Section, Theresienstrasse 41, D-80333 München, Germany

Prikazani su rezultati ispitavanja anizotropije Hallovog koeficijenta $R_{\rm H}$ monokristala Al₇₆Co₂₂Ni₂ slitine. Ta je slitina dekagonalni aproksimant Y-Al-Ni-Co sustava. Hallov koeficijent je mjeren za tri kristalografska smjera a^* , bi c jedinične ćelije Y-Al-Ni-Co, kod koje su monoklinske atomske ravnine (a,c) naslagane duž b smjera okomitog na njih. $R_{\rm H}$ pokazuje zamjetnu anizotropiju koja je posljedica anizotropne strukture kao i Fermijeve površine. Rezultati su uspoređeni s anizotropijom Hallovog koeficijenta dekagonalnog kvazikristala d-Al-Ni-Co iz literature.