

Anisotropic Transport Properties of the Orthorhombic $\text{Al}_{13}\text{Co}_4$ Approximant to the Decagonal Quasicrystal*

Jagoda Lukatela,^{a,***} Denis Stanić,^a Petar Popčević,^a Jovica Ivković,^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

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Abstract. Anisotropic transport properties (electrical resistivity, thermoelectric power, Hall coefficient and thermal conductivity) of the *o*- $\text{Al}_{13}\text{Co}_4$ orthorhombic approximant to the decagonal phase were investigated. The crystalline-direction-dependent measurements were performed along the *a*, *b* and *c* directions of the orthorhombic unit cell, where (*b*, *c*) atomic planes are stacked along the perpendicular *a* direction. Anisotropic electrical and thermal conductivities are the highest along the stacking *a* direction. The anisotropic thermoelectric power changes sign with the crystalline direction and so does the anisotropic Hall coefficient, which changes from the negative electron-like to the positive hole-like for different combinations of the electric current and magnetic field direction. The transport properties of the *o*- $\text{Al}_{13}\text{Co}_4$ phase were compared to the literature data on approximant to the decagonal quasicrystals, the $\text{Al}_{76}\text{Co}_{22}\text{Ni}_2$ and the $\text{Al}_{80}\text{Cr}_{15}\text{Fe}_5$, allowing for the study of evolution of transport properties with increasing structural complexity and unit cell size.

Keywords: transport properties, approximant to the decagonal quasicrystal, orthorhombic $\text{Al}_{13}\text{Co}_4$

INTRODUCTION

Recently, anisotropic transport properties (electrical resistivity, thermoelectric power, Hall coefficient and thermal conductivity), measured along three orthogonal crystalline directions, have been reported for the $\text{Al}_{76}\text{Co}_{22}\text{Ni}_2$ compound.^{1–4} This material belongs to the derivative of the $\text{Al}_{13}\text{TM}_4$ (TM is a transition metal) compounds and is a monoclinic approximant to the decagonal quasicrystals with two atomic layers within one periodic unit of ≈ 0.4 nm along the stacking direction and a relatively small unit cell, comprising 32 atoms. The investigated transport properties of the $\text{Al}_{76}\text{Co}_{22}\text{Ni}_2$ were found to be metallic in all crystalline directions, showing a pronounced anisotropy, with both the electrical and heat conductivity being the highest along the stacking direction of the crystal (corresponding to the periodic direction in decagonal quasicrystals (d-QCs)). Anisotropic transport properties have also been reported for the $\text{Al}_{80}\text{Cr}_{15}\text{Fe}_5$ ^{5,6} complex metallic

alloy. It is a derivative of orthorhombic Al_4TM compounds⁷ which are approximants to the decagonal quasicrystals with six atomic layers in a periodic unit of 1.25 nm and 306 atoms in the giant unit cell. The measurements showed that the in-plane electrical resistivity of this compound exhibits nonmetallic behavior with a maximum in the resistivity at lower temperatures, whereas the resistivity along the stacking direction shows a metallic behavior.

Here, we report the measurements of anisotropic transport properties (electrical resistivity, thermoelectric power, Hall coefficient and thermal conductivity) of the orthorhombic *o*- $\text{Al}_{13}\text{Co}_4$ complex metallic alloy, which is a derivative of the $\text{Al}_{13}\text{TM}_4$ compound, with four atomic layers within one periodic unit of ≈ 0.8 nm along the stacking direction and a unit cell comprising 102 atoms. These measurements complement our previous work on the anisotropic transport properties of the $\text{Al}_{76}\text{Co}_{22}\text{Ni}_2$ approximant to the decagonal quasicrystals with two atomic layers within one periodic unit of ≈ 0.4 nm

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

and 32 atoms in the relatively small unit cell and the $Al_{80}Cr_{15}Fe_5$ with six atomic layers within one periodic unit of ≈ 1.25 nm and 306 atoms in the giant unit cell. The $o\text{-}Al_{13}Co_4$ phase with four atomic layers and 102 atoms in the unit cell is thus intermediate to the other two approximant phases regarding the number of layers in one periodic unit and the size of the unit cell. A comparison of the three phases can give us an insight into the way the anisotropic transport properties of the approximant to the decagonal quasicrystals evolve with increasing structural complexity and the unit cell size.

EXPERIMENTAL

The $o\text{-}Al_{13}Co_4$ single crystal used in our study was grown by the Czochralski technique and its structure matched well the orthorhombic unit cell.⁸ In order to perform crystalline-direction-dependent studies we have cut, from the ingot, three bar-shaped samples of dimensions $1 \times 1 \times 7$ mm³, with their long axes along three orthogonal directions. The long axis of the first sample was along the [1 0 0] stacking direction (designated as *a*), which corresponds to the pseudo-tenfold axis of the $o\text{-}Al_{13}Co_4$ structure and is equivalent to the periodic (tenfold) direction in the related d-QCs. The (*b*, *c*) orthorhombic plane corresponds to the quasiperiodic plane in the d-QCs and the second sample was cut with its long axis along the [0 1 0] (*b*) direction and the third one along the [0 0 1] (*c*) direction. For each sample, the orientation of the other two crystalline directions was also known. The so-prepared samples enabled us to determine the anisotropic transport properties of the $o\text{-}Al_{13}Co_4$ approximant to the decagonal quasicrystals along the three principal orthorhombic directions of the unit cell.

RESULTS AND DISCUSSION

Electrical Resistivity

The electrical resistivity, $\rho(T)$, was measured between 2 K and 300 K using the standard four-terminal technique. The $\rho(T)$ data along the three crystalline directions are shown in Figure 1. The resistivity is the lowest along the stacking *a* direction perpendicular to the atomic planes, where its room temperature (r.t.) value amounts $\rho_a^{\text{r.t.}} = 69 \mu\Omega \text{ cm}$ and the residual resistivity is $\rho_a^{2 \text{ K}} = 47 \mu\Omega \text{ cm}$. The two in-plane resistivities are higher, amounting $\rho_b^{\text{r.t.}} = 169 \mu\Omega \text{ cm}$ and $\rho_b^{2 \text{ K}} = 113 \mu\Omega \text{ cm}$ for the *b* direction, and $\rho_c^{\text{r.t.}} = 180 \mu\Omega \text{ cm}$ and $\rho_c^{2 \text{ K}} = 129 \mu\Omega \text{ cm}$ for the *c* direction. The anisotropy of the two in-plane resistivities is small, amounting at r.t. to $\rho_c^{\text{r.t.}} / \rho_b^{\text{r.t.}} = 1.1$, whereas the anisotropy to the stacking direction is considerably larger, $\rho_c^{\text{r.t.}} / \rho_a^{\text{r.t.}} = 2.6$ and $\rho_b^{\text{r.t.}} / \rho_a^{\text{r.t.}} = 2.5$. The observed anisotropic resistivities thus appear in the order $\rho_a < \rho_b < \rho_c$ (even the inequality

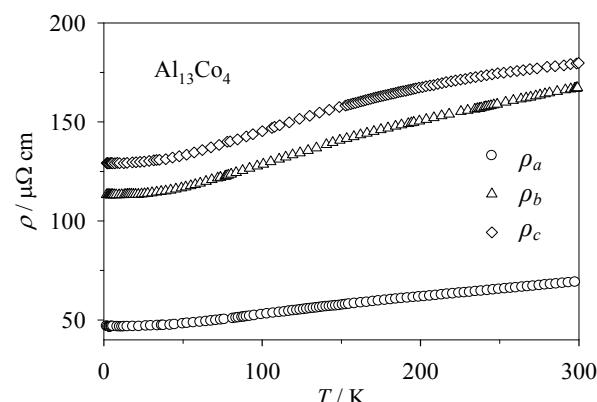


Figure 1. Temperature-dependent electrical resistivity $\rho(T)$ of $o\text{-}Al_{13}Co_4$ along three orthogonal crystalline directions *a*, *b* and *c*.

$\rho_a \ll \rho_b < \rho_c$ may be considered to hold), so that the stacking *a* direction is the most electrically conducting one. The metallic behavior of the resistivity along all three crystalline directions demonstrates predominant role of the electron-phonon scattering mechanism.

Thermoelectric Power

The thermoelectric power, $S(T)$, was measured between 2 K and 300 K using a standard temperature-gradient technique.⁹ The thermopower data, measured along the three crystalline directions *a*, *b* and *c*, are shown in Figure 2. The thermopower appears in the order $S_a > S_b > S_c$ and shows unusual anisotropy: it is positive along the stacking *a* direction with the r.t. value $S_a^{\text{r.t.}} = 18.6 \mu\text{V K}^{-1}$, it becomes almost symmetrically negative for the in-plane *b* direction with $S_b^{\text{r.t.}} = -17.1 \mu\text{V K}^{-1}$ and is close to zero for the second in-plane *c* direction, amounting $S_c^{\text{r.t.}} = -2.9 \mu\text{V K}^{-1}$. While S_a and S_b exhibit relatively strong linear-like temperature dependence (a small change of slope at about 50 K may be noticed, a feature that is often associated with electron-phonon

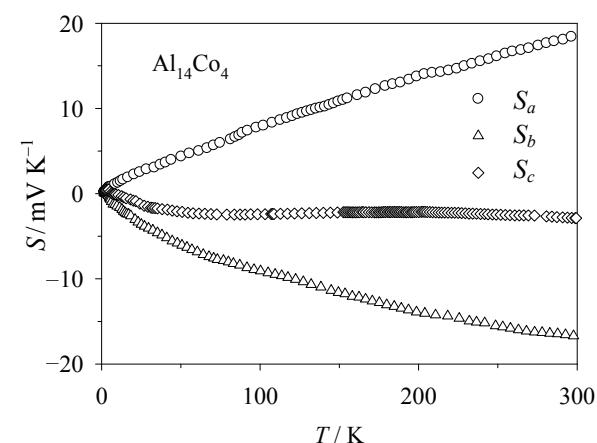


Figure 2. Temperature-dependent thermoelectric power, $S(T)$, of $o\text{-}Al_{13}Co_4$ along three orthogonal crystalline directions *a*, *b* and *c*.

effects), S_c shows almost no temperature dependence in the temperature range investigated. The observed anisotropy of the thermopower, ranging between positive and negative values, suggests that the Fermi surface is highly anisotropic, consisting of electron-like and hole-like parts, which may compensate each other for a certain crystalline direction to yield a thermopower close to zero. Moreover, the anisotropy of the phonon spectrum may contribute to the anisotropic thermopower via the anisotropic electron–phonon interaction as well (the importance of phonons in the electron transport of $o\text{-}Al_{13}Co_4$ is manifested in the metallic electrical resistivity of Figure 1).

Hall Coefficient

The Hall-effect measurements, $R_H(T)$, were performed by a five-point method using a standard ac technique in magnetic fields up to 10 kOe. The current through the samples was in the range 10–50 mA. The measurements were performed in the temperature interval from 90 K to 390 K. The temperature-dependent Hall coefficient $R_H = E_y/j_x B_z$ is shown in Figure 3. In order to determine the anisotropy of R_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, the R_H values are typical metallic of the order of $10^{-10} \text{ m}^3 \text{ C}^{-1}$. R_H exhibits pronounced anisotropy with the following regularity: the six R_H sets of data form three groups of two practically identical R_H curves, where 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. Thus, identical Hall coefficients are obtained for combinations

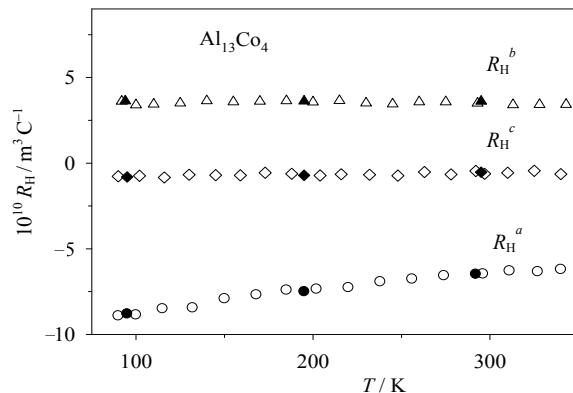


Figure 3. Anisotropic temperature-dependent Hall coefficient $R_H = E_y/j_x B_z$ of $o\text{-}Al_{13}Co_4$ for different combinations of direction a , b , c , current j_x and magnetic field B_z (given in the legend). The superscript a , b or c on R_H denotes the direction of the magnetic field.

$E_b/j_c B_a = E_c/j_b B_a = R_H^a$ (where the additional superscript on the Hall coefficient denotes the direction of the magnetic field), amounting $R_H^a(\text{r.t.}) = -6.5 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$, $E_a/j_c B_b = E_c/j_a B_b = R_H^b$ with $R_H^b(\text{r.t.}) = 3.5 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ and $E_b/j_a B_c = E_a/j_b B_c = R_H^c$ with $R_H^c(\text{r.t.}) = -0.6 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$. R_H^b and R_H^c are practically temperature-independent within the investigated temperature range, whereas R_H^a shows moderate temperature dependence that tends to disappear at higher temperatures. The observed R_H anisotropy reflects the complicated structure of the Fermi surface. The negative $R_H^a < 0$ is electron-like for the magnetic field along the stacking a direction, whereas the positive $R_H^b > 0$ is hole-like for the field along the in-plane b direction. For the field along the second in-plane direction c , $R_H^c \approx 0$ suggests that the electron-like and hole-like contributions are of comparable importance. This orientation-dependent mixed electron-like and hole-like behavior of the anisotropic Hall coefficient is analogous to the anisotropy of the thermopower, presented in Figure 2, which also changes sign with crystalline orientation. In both cases there is no simple explanation of this dual behavior, which requires knowledge of the details of the Fermi surface pertinent to the $o\text{-}Al_{13}Co_4$ phase. Here, it is worth mentioning that the anisotropic Hall coefficient of the d-Al-Ni-Co-type QC shows a similar duality, being also hole-like for the field lying in the quasiperiodic plane and electron-like for the field along the periodic direction.^{10,11} This duality is proposed to be a universal feature of d-QCs.

Thermal Conductivity

The thermal conductivity, $\kappa(T)$, of $o\text{-}Al_{13}Co_4$ was measured along the a , b and c directions using an absolute steady-state heat-flow method.¹² The thermal flux through the samples was generated by a 1 k Ω RuO₂ chip-resistor, glued to one end of the sample, while the other end was attached to a copper heat sink. The temperature gradient across the sample was monitored by a chromel-constantan differential thermocouple. The phonon contribution, $\kappa_{\text{ph}} = \kappa - \kappa_{\text{el}}$, was estimated by subtracting the electronic contribution, κ_{el} , from the total conductivity, κ , using the Wiedemann-Franz law, $\kappa_{\text{el}} = \pi^2 k_B^2 T \sigma(T) / 3e^2$ and the measured electrical conductivity data $\sigma(T) = \rho^{-1}(T)$ from Figure 1. The total thermal conductivity, κ , along the three crystalline directions is displayed in Figure 4 and the corresponding electronic contribution, κ_{el} , is shown in small symbol. At 300 K, we get the following anisotropy: $\kappa^a = 12.5 \text{ W m}^{-1} \text{ K}^{-1}$, $\kappa_{\text{el}}^a = 10.2 \text{ W m}^{-1} \text{ K}^{-1}$ with their ratio $(\kappa_{\text{el}}^a/\kappa^a)_{\text{r.t.}} = 0.8$, $\kappa^b = 6.1 \text{ W m}^{-1} \text{ K}^{-1}$, $\kappa_{\text{el}}^b = 4.4 \text{ W m}^{-1} \text{ K}^{-1}$ with $(\kappa_{\text{el}}^b/\kappa^b)_{\text{r.t.}} = 0.7$ and $\kappa^c = 6.2 \text{ W m}^{-1} \text{ K}^{-1}$, $\kappa_{\text{el}}^c = 4.1 \text{ W m}^{-1} \text{ K}^{-1}$ with $(\kappa_{\text{el}}^c/\kappa^c)_{\text{r.t.}} = 0.7$. Electrons (and holes) are thus the majority heat carriers at room temperature for all three directions. The anisotropic thermal conductivities

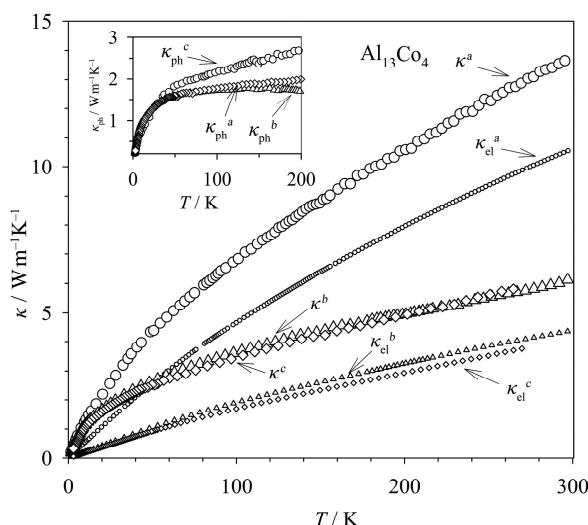


Figure 4. Total thermal conductivity, $\kappa(T)$, of $o\text{-}Al_{13}Co_4$ along three crystalline directions a , b and c . Electronic contribution κ_{el} , estimated from the Wiedemann-Franz law, are shown with small symbol. Inset: Phonon thermal conductivity $\kappa_{ph} = \kappa - \kappa_{el}$ along the three crystalline directions.

appear in the order $\kappa^a > \kappa^b \approx \kappa^c$, and the same order applies to the electronic parts $\kappa_{el}^a > \kappa_{el}^b \approx \kappa_{el}^c$. The thermal conductivity is thus the highest along the stacking a direction, whereas the in-plane conductivity is smaller with no noticeable anisotropy between the two in-plane directions b and c . Figure 2 shows that the electrical conductivity of $o\text{-}Al_{13}Co_4$ is also the highest along a (appearing in the order $\sigma_a > \sigma_b > \sigma_c$). Thus this material is the best conductor, along the stacking a direction perpendicular to the (b, c) atomic planes, for both the electricity and heat. The phonon thermal conductivity, κ_{ph} , is shown in the inset of Figure 4. We observe that the anisotropy of κ_{ph} is small and no systematic differences between the three directions can be claimed unambiguously.

CONCLUSION

We have measured electrical resistivity, thermoelectric power, Hall coefficient and thermal conductivity of the orthorhombic $o\text{-}Al_{13}Co_4$ complex metallic alloy, belonging to the derivative of the $Al_{13}TM_4$ compound, with four atomic layers within one periodic unit of ≈ 0.8 nm along the stacking a direction and comprising 102 atoms in the giant unit cell. Our main objective was to determine the crystalline-direction-dependent anisotropy of the investigated transport coefficients when measured within the (b, c) atomic planes, corresponding to the quasiperiodic planes in the related d-QCs, and along the stacking a direction perpendicular to the planes, corresponding to the periodic direction in d-QCs. The electrical resistivity of $o\text{-}Al_{13}Co_4$ is relatively low in all

crystalline directions, with the room temperature values in the range $70\text{--}80 \mu\Omega \text{ cm}$. There exists a significant anisotropy between the in-plane resistivity and the resistivity along the stacking a direction by a factor about 2.5, whereas the anisotropy between the two in-plane directions b and c is much smaller. Thermal conductivity is the highest along the stacking a direction, whereas the in-plane conductivity is smaller with no noticeable anisotropy between the b and c directions. Since the electrical conductivity of $o\text{-}Al_{13}Co_4$ is also the highest along a , this material is the best conductor for both the electricity and heat along the stacking a direction perpendicular to the (b, c) atomic planes. Electrons (and holes) are the majority heat carriers at room temperature along all three crystalline directions.

Both the anisotropic thermopower and the Hall coefficient results point to the complicated Fermi surface that consists of the electron-like and hole-like parts.

The observed anisotropic behavior of transport properties is quite universal for decagonal quasicrystals. Thus, typical values found for the resistivity^{13–15} were $\rho_{qc} \approx 250 \mu\Omega \text{ cm}$ and $\rho_c \approx 50 \mu\Omega \text{ cm}$, thermal conductivity^{16,17} $\kappa_{qc} \approx 1 \text{ W m}^{-1} \text{ K}^{-1}$ and $\kappa_c \approx 10 \text{ W m}^{-1} \text{ K}^{-1}$ and thermopower¹⁸ $S_{qc} \approx -5 \mu\text{V K}^{-1}$ and $S_c \approx +7 \mu\text{V K}^{-1}$, along a direction in the quasiperiodic plane and periodic direction respectively. Values for the Hall coefficient^{10,11} were found to be $R_{H\perp qc} \approx -|1\text{--}10| 10^{-4} \text{ cm}^3 \text{ C}^{-1}$ with the magnetic field perpendicular to the quasicrystal plane and $R_{H\perp c} \approx (1\text{--}10) 10^{-4} \text{ cm}^3 \text{ C}^{-1}$ with the magnetic field perpendicular to the crystalline direction.

Comparing the transport properties of the three stacked-layer phases of increasing structural complexity, the $Al_{76}Co_{22}Ni_2$ two-layer phase, the $o\text{-}Al_{13}Co_4$ four-layer phase and the $Al_{80}Cr_{15}Fe_5$ six-layer phase, some general conclusions can be drawn on the evolution of the anisotropic transport properties of the approximant to the decagonal quasicrystals with increasing structural complexity and the unit cell size. The electrical resistivity shows similar anisotropy, being weak between the two in-plane directions and stronger in the stacking direction. The resistivity values increase with increasing complexity of the compounds, being the lowest for the $Al_{76}Co_{22}Ni_2$, significantly higher for the $o\text{-}Al_{13}Co_4$ and even higher for the $Al_{80}Cr_{15}Fe_5$. The anisotropic thermal conductivity behaves in complete analogy to the electrical resistivity, by showing weak in-plane anisotropy and considerable anisotropy to the stacking direction. For all three compounds, the thermal conductivity is the highest along the stacking direction, so that the investigated decagonal approximant phases are the best conductors for both the electricity and heat along the stacking direction perpendicular to the atomic planes. At room temperature, $Al_{76}Co_{22}Ni_2$ shows the highest thermal conductivity and $Al_{80}Cr_{15}Fe_5$ the lowest one, indicating that increasing complexity of the structure

results in less efficient electronic and phonon transport of the heat.

The anisotropic Hall coefficient shows the following regularity: the application of the field along the stacking direction always yields the lowest value of the Hall coefficient (for $o\text{-}Al_{13}Co_4$ and $Al_{80}Cr_{15}Fe_5$, the corresponding Hall coefficient is negative, whereas for the $Al_{76}Co_{22}Ni_2$, it is practically zero), whereas the application of the field in-plane results in higher R_H values and a change of sign to positive for at least one of the in-plane directions. No systematic change of R_H with increasing structural complexity can be claimed. Regarding the anisotropic thermopower, no systematic differences between the three compounds can be inferred from the available experimental data.

Comparing the transport properties of the above approximants to the decagonal quasicrystals to the true d-QCs we find that the two kinds of compounds are in complete analogy. A comparison to the currently best studied d-Al-Ni-Co-type d-QCs with two atomic layers within one periodic unit shows the following similarities: (i) electrical^{14,15,19,20} and thermal^{13,16-18} conductivity is the highest along the periodic direction and (ii) the Hall coefficient is the lowest for the magnetic field along the periodic direction, whereas R_H changes sign to positive for the field along the in-plane directions.^{11,12,20} The investigated approximants to the decagonal quasicrystals of increasing structural complexity thus exhibit anisotropic transport properties qualitatively similar to that of the decagonal quasicrystals. Both types of compounds have in common the atomic planes that are stacked periodically. The stacked-layer structure appears to be at the origin of the anisotropy of the investigated transport coefficients, whereas the in-plane structural details seem to be of marginal importance for the anisotropic electronic transport properties.

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SAŽETAK

Anizotropna transportna svojstva ortorompskog $Al_{13}Co_4$ aproksimanta dekagonalnih kvazikristala

**Jagoda Lukatela,^a Denis Stanić,^a Petar Popčević,^a Jovica Ivkov,^a
Janez Dolinšek^b i Peter Gille^c**

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

^b*Jožef Stefan Institute, University of Ljubljana, Jamova 39, SI-1000 Ljubljana, Slovenia*

^c*Ludwig-Maximilians-Universität München, Department of Earth and Environmental Sciences,
Crystallography Section, Theresienstrasse 41, D-80333 München, Germany*

Eksperimentalno su istražena anizotropna svojstva (električni otpor, termostruju, Hallov efekt i termičku vodljivost) ortorombskog aproksimanta, $o\text{-}Al_{13}Co_4$, dekagonalne faze. Mjerena je ovisnost navedenih svojstava o a , b , c kristalnom smjeru ortorombske jedinične celije pri čemu se atomske ravnine (b , c) slažu u a smjeru okomitom na njih. Anizotropna električna i toplinska vodljivost imaju najveće vrijednosti u smjeru slaganja a . Anizotropna termostruja mijenja predznak s promjenom kristalnog smjera. Anizotropni Hallov koeficijent također mijenja predznak iz negativnog (elektronski doprinos) u pozitivan (Šupljinski doprinos) za različite smjerove električne struje i magnetskog polja. Transportna svojstva $o\text{-}Al_{13}Co_4$ faze uspoređena su s podacima iz literature za aproksimante dekagonalne faze $Al_{76}Co_{22}Ni_2$ i $Al_{80}Cr_{15}Fe_5$. Što je omogućilo promatranje transportnih svojstava u ovisnosti o složenosti strukture kao i o veličini jedinične celije.