

## Hall Effect of the $\text{Al}_{13}\text{Fe}_4$ Decagonal Approximant and Its Ternary Extension $\text{Al}_{13}(\text{Fe},\text{Ni})_4^*$

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**Abstract.** We have measured Hall coefficient and electrical resistivity of the  $\text{Al}_{13}\text{Fe}_4$  and  $\text{Al}_{13}(\text{Fe},\text{Ni})_4$  monoclinic approximants to the decagonal quasicrystal. While the  $\text{Al}_{13}\text{Fe}_4$  crystals are structurally well ordered, the ternary extension  $\text{Al}_{13}(\text{Fe},\text{Ni})_4$  contains quenched disorder and can be viewed as a disordered version of  $\text{Al}_{13}\text{Fe}_4$ . The crystallographic-direction-dependent Hall effect measurements were performed along the  $a^*$ ,  $b$  and  $c$  directions of the monoclinic unit cell, where ( $a^*,c$ ) atomic planes are stacked along the perpendicular  $b$  direction. The stacking  $b$  direction is the most conducting direction for the electricity. The effect of quenched disorder in  $\text{Al}_{13}(\text{Fe},\text{Ni})_4$  is manifested in the large residual resistivity  $\rho(T \rightarrow 0)$  as compared to the ordered  $\text{Al}_{13}\text{Fe}_4$ . The Hall coefficient,  $R_H$ , values for all combinations of directions, are typical metallic. The anisotropic Hall coefficient reflects complicated structure of the anisotropic Fermi surface that contains electron-like and hole-like parts. Depending on the combination of directions of the current and magnetic field electron-like ( $R_H < 0$ ) or hole-like ( $R_H > 0$ ) contributions may dominate, or the two contributions compensate each other ( $R_H \approx 0$ ).

**Keywords:** complex metallic alloys, decagonal approximants, Hall effect, electrical resistivity

### INTRODUCTION

Transport properties of decagonal quasicrystals (d-QCs) can be consequently studied along the quasiperiodic (Q) and periodic (P) crystallographic directions on the same sample.<sup>1–6</sup> A consequence of the structural anisotropy of d-QCs is anisotropic transport (electrical resistivity, thermopower, Hall coefficient) properties. The electrical resistivity generally shows positive temperature coefficient (PTC) at metallic values along the periodic direction, whereas the resistivity in the quasiperiodic plane is considerably larger and exhibits negative temperature coefficient (NTC) and sometimes also a maximum somewhere below room temperature or approaching 0 K. The anisotropy of the Hall coefficient  $R_H$  is one of intriguing features of d-QCs, being positive hole-like ( $R_H > 0$ ) for the magnetic field lying in the quasiperiodic plane, whereas it changes sign to negative ( $R_H < 0$ ) for the field along the periodic direction, thus becoming electron-like. This  $R_H$  anisotropy was reported for the d-Al-Ni-Co, d-Al-Cu-Co and d-Al-Si-Cu-Co and is considered to be a universal feature of d-QCs.<sup>5,6</sup>

The degree of anisotropy of the transport coefficients is related to the structural details of a particular decagonal phase, depending on the number of quasiperiodic layers in one periodic unit.<sup>7,8</sup> The most anisotropic case are the phases with just two layers, realized in d-Al-Ni-Co and d-Al-Cu-Co, with anisotropy of electrical resistivity  $\rho_Q/\rho_P$  at room temperature in the range 6–10,<sup>1–3</sup> while the other  $d$  phases contain more quasiperiodic layers in a periodic unit and show smaller anisotropies. So, in d-Al-Co, d-Al-Ni and d-Al-Si-Cu-Co there are four quasiperiodic layers with anisotropy of electrical resistivity at room temperature in the range  $\rho_Q/\rho_P = 2–4$ .<sup>4</sup> d-Al-Mn, d-Al-Cr and d-Al-Pd-Mn phases contain six layers and the anisotropy amounts  $\rho_Q/\rho_P = 1.2–1.4$ , whereas d-Al-Pd and d-Al-Cu-Fe phases with eight layers are close to isotropic ( $\rho_Q/\rho_P \approx 1$ ). While the origin of the anisotropic electron transport coefficients is the anisotropic Fermi surface, the anisotropy of which originates from the specific stacked-layer crystal structure of the d-QC phases and the chemical decoration of the lattice, the lack of translational periodicity within the quasiperiodic planes prevents any

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Dedicated to Professor Boran Leontić on the occasion of his 80<sup>th</sup> birthday.

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quantitative analysis of this appearance. The problem can be prevailed by considering approximant phases to the decagonal phase. Approximant phases are characterized by large unit cells, which periodically repeat in space with the atomic decoration closely resembling that of d-QCs. Atomic layers are again stacked periodically and the atomic planes of approximants and d-QCs show locally similar patterns, so that their structures on the scale of near-neighbor atoms closely resemble each other. Decagonal approximants thus offer valid comparison to the decagonal quasicrystals.

Recently, we have reported the anisotropic transport properties for three decagonal approximant phases of increasing structural complexity comprising two (Y-Al-Ni-Co),<sup>9,10</sup> four (*o*- $Al_{13}Co_4$  decagonal approximant)<sup>11</sup> and six atomic layers ( $Al_4(Cr,Fe)$ )<sup>12,13</sup> in the unit cell. Common to all these phases is strong anisotropy of transport properties between the stacking and in-plane directions, where the crystals show the highest conductivity for the electricity along the stacking direction (corresponding to the periodic direction in d-QCs), whereas the in-plane anisotropy is considerably smaller.

In this paper we present anisotropic Hall coefficients together with the electrical resistivity of two other decagonal approximant phases from the  $Al_{13}TM_4$  (TM = transition metal) class of complex intermetallics, which are the monoclinic  $Al_{13}Fe_4$  and its ternary extension  $Al_{13}(Fe,Ni)_4$ . Both alloys comprise four atomic layers within one periodic unit which comprises 102 atoms. The  $Al_{13}Fe_4$  crystals can be currently grown to the highest structural perfection within the  $Al_{13}TM_4$  class, containing small amount of quenched structural disorder, and the  $Al_{13}Fe_4$  phase is considered to be the "parent phase" of this class. The introduction of a small amount of Ni atoms into the structure of the ternary extension  $Al_{13}(Fe,Ni)_4$  (the fraction of Ni atoms about 2 % in our case) creates disorder within the more or less perfect  $Al_{13}Fe_4$  structure, so that the comparison of  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  demonstrates the effect of quenched disorder on the transport properties of decagonal approximants.

## EXPERIMENTAL

The  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  single crystals used in our study were grown by the Czochralski technique and the details of preparation are described elsewhere.<sup>14</sup> The chemical composition of the  $Al_{13}(Fe,Ni)_4$  crystal was  $Al_{76.5}Fe_{21.3}Ni_{2.2}$  and it was grown from a melt of composition  $Al_{85}Fe_{10}Ni_5$ . The  $Al_{13}(Fe,Ni)_4$  phase is a ternary extension of the  $Al_{13}Fe_4$  with the maximum solubility of Ni in  $Al_{13}Fe_4$  of 8.9 %.<sup>15</sup> The structure of both investigated compounds matched well to the monoclinic unit cell of the Grin *et al.* model,<sup>16</sup> and there was practically no difference between the cell parameters of the  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  compounds. They are recognized as an approximants phase to the  $Al_{71}Ni_{24}Fe_5$  decagonal quasicrystal.<sup>17</sup>

The main objective was to determine the crystallographic-direction-dependent anisotropy of the investigated Hall coefficient when measured within the ( $a^*,c$ ) atomic planes, corresponding to the quasiperiodic planes in the related d-QCs, and along the stacking  $b$  direction perpendicular to the planes, corresponding to the periodic direction in d-QCs. In order to perform such studies, we have cut from the ingot of each compound three bar-shaped samples of dimensions  $1 \times 1 \times 8$  mm<sup>3</sup>, with their long axes along three orthogonal directions. The long axis of the first sample was along the [010] stacking direction (designated in the following as  $b$ ), which corresponds to the periodic direction in the related d-QCs. The ( $a,c$ ) monoclinic plane corresponds to the quasiperiodic plane in d-QCs and the second sample was cut with its long axis along the [001] ( $c$ ) 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 17° with respect to  $a$  and perpendicular to  $c$ ). For each sample, the orientation of the other two crystallographic directions was also known. The so-prepared samples enabled us to determine the anisotropic resistivity and Hall coefficient along the three orthogonal directions of the investigated monoclinic  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$ . The Hall-coefficient measurements were performed by the five-point method using standard AC technique in magnetic fields up to 1 T. The current through the samples was in the range 10–50 mA. The measurements were performed in the temperature interval from 90 to 370 K, while electrical resistivity was measured between 2 and 300 K using the standard four-terminal technique.

## RESULTS AND DISCUSSION

### Electrical Resistivity

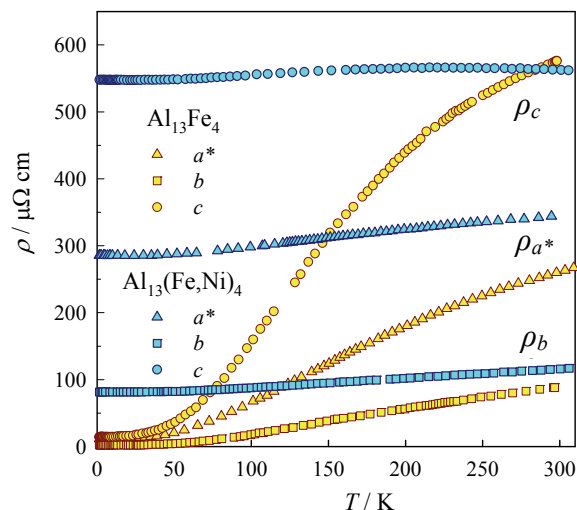
The electrical resistivity,  $\rho(T)$ , data for  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  along the three crystallographic directions are displayed in Figure 1. For both compounds, the anisotropic resistivities appear in the order  $\rho_b < \rho_{a^*} < \rho_c$ , so that the resistivity is the lowest along the stacking  $b$  direction perpendicular to the atomic planes. The anisotropy between the two in-plane directions  $a^*$  and  $c$  is also substantial (Table 1).

The resistivities of the two compounds exhibit different temperature dependence.  $Al_{13}Fe_4$  exhibits small residual resistivities  $\rho(T \rightarrow 0)$ , amounting at 2 K  $\rho_{a^*}^{2K} = 11 \mu\Omega$  cm,  $\rho_b^{2K} = 2 \mu\Omega$  cm and  $\rho_c^{2K} = 14 \mu\Omega$  cm, and large PTC of the resistivity along all three crystallographic directions, demonstrating an important role of the electron-phonon scattering mechanism. At the room temperature, r.t., the resistivities reach the values  $\rho_{a^*}^{r.t.} = 268 \mu\Omega$  cm,  $\rho_b^{r.t.} = 88 \mu\Omega$  cm and  $\rho_c^{r.t.} = 576 \mu\Omega$  cm. In contrast to that, the residual resistivities of

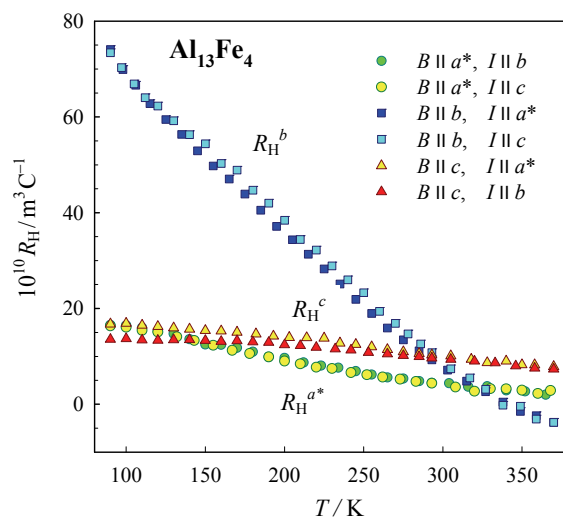
$\text{Al}_{13}(\text{Fe,Ni})_4$  are much larger, amounting  $\rho_{a^*}^{2\text{K}} = 286 \mu\Omega \text{ cm}$ ,  $\rho_b^{2\text{K}} = 81 \mu\Omega \text{ cm}$  and  $\rho_c^{2\text{K}} = 548 \mu\Omega \text{ cm}$ , and the PTC of the resistivity is very small for all three crystallographic directions. The 300 K resistivity values of  $\text{Al}_{13}(\text{Fe,Ni})_4$  are almost the same as those of  $\text{Al}_{13}\text{Fe}_4$  (Table 1). The marked difference between the residual resistivities of the two compounds can be explained by the presence of quenched structural disorder in  $\text{Al}_{13}(\text{Fe,Ni})_4$  and its absence in  $\text{Al}_{13}\text{Fe}_4$ . In the structurally well-ordered  $\text{Al}_{13}\text{Fe}_4$ , the amount of quenched disorder is small, yielding small residual resistivities and large PTC due to the high density of phonons in the lattice that are at the origin of the positive temperature coefficient. The large non-zero residual resistivity of  $\text{Al}_{13}(\text{Fe,Ni})_4$  is, on the other hand, a consequence of quenched disorder in this compound, together with the large effective masses of electrons and small electron velocities which are characteristic for complex metallic alloys.<sup>12</sup> The strong anisotropy of the residual resistivity suggests that the amount of quenched disorder depends on the crystallographic direction.

### Hall Effect

The anisotropic temperature-dependent Hall coefficient  $R_H = E_y/j_x B_z$  (where  $B_z$  is magnetic field perpendicular to the current of the density  $j_x$ , and  $E_y$  is transverse Hall field) of  $\text{Al}_{13}\text{Fe}_4$  is shown in Figure 2, whereas  $R_H$  of  $\text{Al}_{13}(\text{Fe,Ni})_4$  is shown in Figure 3. In order to determine the anisotropy of  $R_H$ , three sets of experiments were performed with the current  $j_x$  along the long axis of each sample (thus along  $a^*$ ,  $b$  and  $c$ , respectively), whereas the magnetic field  $B_z$  was directed along each of the other two orthogonal crystallographic directions, making six experiments altogether for each compound. For all combinations of directions, the  $R_H$  absolute values are typical metallic in the range  $10^{-9} - 10^{-10} \text{ m}^3 \text{ C}^{-1}$  (with the experimental uncertainty of  $\pm 0.1 \times 10^{-10} \text{ m}^3 \text{ C}^{-1}$ ).  $R_H$ 's exhibit pronounced anisotropy with the following regularity. For each compound six  $R_H$  sets of data form three groups of two practically identical  $R_H$  curves, where the magnetic field in a given crystallographic direction yields the same  $R_H$  for the current along the other two crystallographic directions in the perpendicular plane. Thus, identical Hall coefficients are obtained for the pair combinations  $E_b/j_c B_{a^*} = E_c/j_b B_{a^*} = R_H^{a^*}$  (where the additional superscript on the Hall coefficient denotes the direction of the magnetic field),  $E_{a^*}/j_c B_b = E_c/j_{a^*} B_b = R_H^b$  and  $E_{a^*}/j_b B_c = E_b/j_{a^*} B_c = R_H^c$ . At 100 K, the anisotropic  $R_H$  values of  $\text{Al}_{13}\text{Fe}_4$  amount  $R_H^{a^*} = 1.7 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ ,  $R_H^b = 6.8 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$  and  $R_H^c = 1.6 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ , whereas the corresponding values of  $\text{Al}_{13}(\text{Fe,Ni})_4$  are  $R_H^{a^*} = -1.1 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ ,  $R_H^b = -4.6 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$  and  $R_H^c = 4.9 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ . For both compounds,  $R_H^{a^*}$  and



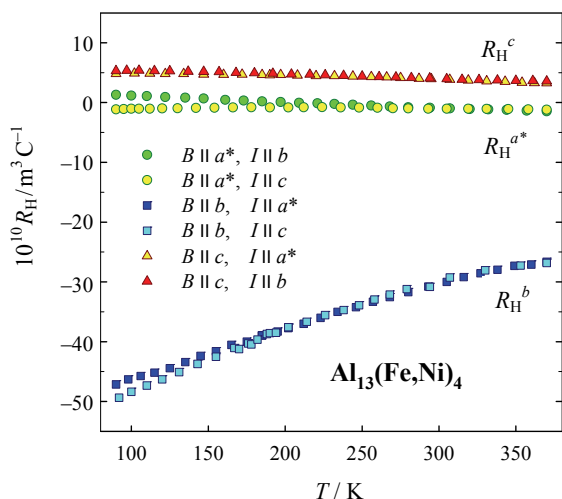
**Figure 1.** Temperature-dependent electrical resistivity of  $\text{Al}_{13}\text{Fe}_4$  and  $\text{Al}_{13}(\text{Fe,Ni})_4$  along three orthogonal crystallographic directions  $a^*$ ,  $b$  and  $c$ .



**Figure 2.** Anisotropic temperature-dependent Hall coefficient  $R_H = E_y/j_x B_z$  of  $\text{Al}_{13}\text{Fe}_4$  for different combinations of directions of the current  $j_x$  and magnetic field  $B_z$ . The superscript  $a^*$ ,  $b$  or  $c$  on  $R_H$  denotes the direction of the magnetic field.

**Table 1.** The values of the electrical resistivity of the  $\text{Al}_{13}\text{Fe}_4$  and  $\text{Al}_{13}(\text{Fe,Ni})_4$  alloys for three orthogonal crystallographic directions  $a^*$ ,  $b$  and  $c$  at 2 K and the room temperature

Alloy	$\rho$ (2 K) / $\mu\Omega \text{ cm}$			$\rho$ (r.t.) / $\mu\Omega \text{ cm}$		
	$a^*$	$b$	$c$	$a^*$	$b$	$c$
$\text{Al}_{13}\text{Fe}_4$	11	2	14	268	81	576
$\text{Al}_{13}(\text{Fe,Ni})_4$	286	88	548	344	116	562



**Figure 3.** Anisotropic temperature-dependent Hall coefficient  $R_H = E_y / j_x B_z$  of  $Al_{13}(Fe,Ni)_4$  for different combinations of directions of the current  $j_x$  and magnetic field  $B_z$ . The superscript  $a^*$ ,  $b$  or  $c$  on  $R_H$  denotes the direction of the magnetic field.

$R_H^c$  are almost temperature-independent within the investigated temperature range, whereas  $R_H^b$  shows quite strong temperature dependence, being negative in  $Al_{13}(Fe,Ni)_4$ , while in  $Al_{13}Fe_4$  it change the sign from large positive to negative values.

The Hall coefficients of  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  show complicated temperature-dependent anisotropy for different combinations of the current and field directions. For some combinations, Hall coefficient is electron-like ( $R_H < 0$ ), for others it is hole-like ( $R_H > 0$ ) or practically zero (e.g.  $R_H^{a^*}$  of  $Al_{13}(Fe,Ni)_4$ ). All this reflects complicated structure of the anisotropic Fermi surface that contains electron-like and hole-like contributions. Depending on the combination of crystallographic directions, either electron-like ( $R_H < 0$ ) or hole-like ( $R_H > 0$ ) contributions dominate, or the two contributions compensate each other ( $R_H \approx 0$ ). Similar complicated anisotropic behavior was observed also in the thermopower.<sup>18</sup>

Due to the large positive  $R_H^b$  of  $Al_{13}Fe_4$  at low temperatures, it is not possible to establish an analogy between the  $R_H$  anisotropy in this alloy and the anisotropy in d-QCs, which was mentioned in the introduction. However, in  $Al_{13}(Fe,Ni)_4$   $R_H \geq 0$  for magnetic field in plane which corresponds to the quasiperiodic plane in d-QC, and  $R_H < 0$  for  $B$  perpendicular to this plane. Therefore, there is partial analogy between the  $R_H$  of this alloy and the anisotropy of d-QC. Here we note that similar Hall-coefficient anisotropy was reported also for the related decagonal approximants Y-Al-Ni-Co<sup>9,10</sup> and  $o$ - $Al_{13}Co_4$ .<sup>11</sup>

Quantitative analysis of the Hall effect may be very difficult. It requires knowledge of the anisotropic Fermi surface and, sometimes, even of the electrons

scattering times. The  $R_H$  anisotropy was reproduced theoretically, quantitatively for the Y-Al-Ni-Co and qualitatively for  $o$ - $Al_{13}Co_4$ , using the *ab-initio* calculated anisotropic Fermi surfaces of these compound.<sup>10,11</sup> However, in those calculations it was possible to use (thanks to the small temperature dependence of  $R_H$ ) the approximation of isotropic scattering times. This approximation is evidently not appropriate at all for  $Al_{13}Fe_4$ , for which it is possible to calculate the Fermi surface.<sup>17</sup> For  $Al_{13}(Fe,Ni)_4$  compound this approximation may give some qualitative results, but the calculation of the Fermi surface is uncertain due to the chemical disorder of the compound. Therefore, the detailed theoretical analysis is left for the future. At present, an approximate analysis of the temperature dependence of  $R_H$  in terms of electron-like and hole-like charge carriers with anisotropic scattering times is under the way.

As to the rather large temperature dependence of  $R_H$  (for magnetic field in  $b$  direction at least) it ought to emphasize that it is not due neither to the anomalous Hall effect nor to the increasing number of the current carriers. Measurements of the magnetic properties<sup>17</sup> have shown that both compounds have too low magnetic susceptibility ( $< 10^{-5}$ ) to yield a contribution of the order of  $10^{-10} \text{ m}^3 \text{ C}^{-1}$ . Next, from the temperature dependence of the resistivity it is evident that no increase of the number of the charge carriers is taking place.

## CONCLUSION

We have investigated Hall coefficient and the electrical resistivity of the  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  monoclinic approximants to the decagonal quasicrystals. While the  $Al_{13}Fe_4$  crystals are structurally well ordered, as can be deduced from the resistivity results, the introduction of Ni atoms creates chemically disordered ternary extension  $Al_{13}(Fe,Ni)_4$ . The large sensitivity of the electrical properties of these alloys on the Ni content may enable the detailed study of the effects of disorder on the properties of complex intermetallic compounds in general.

Both the Hall coefficient and electrical resistivity of the  $Al_{13}Fe_4$  and  $Al_{13}(Fe,Ni)_4$  alloys exhibit a pronounced anisotropy. In the Hall effect, depending on the combination of directions of the current and magnetic field, electron-like ( $R_H < 0$ ) or hole-like ( $R_H > 0$ ) contributions may dominate, or the two contributions compensate each other ( $R_H \approx 0$ ). The anisotropy of  $R_H$  in  $Al_{13}(Fe,Ni)_4$  can be correlated to the anisotropy of  $R_H$  in d-QCs, and Y-Al-Ni-Co and  $o$ - $Al_{13}Co_4$  compounds as well.  $R_H$  of  $Al_{13}Fe_4$  alloy exhibits unusually large temperature dependence for magnetic field perpendicular to the stacking planes.

The theoretical analysis of the Hall effect is postponed because it would require the knowledge of, especially for  $Al_{13}Fe_4$  alloy, the anisotropic scattering times as well as the knowledge of the influence of chemical disorder on the Fermi surface of  $Al_{13}(Fe,Ni)_4$  alloy.

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## SAŽETAK

### Hallov efekt u $Al_{13}Fe_4$ dekahagonalnom aproksimantu i njegovoj ternarnoj ekstenziji $Al_{13}(Fe,Ni)_4$

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Izmjerali smo Hallov koeficijent i električnu otpornost u  $Al_{13}Fe_4$  i  $Al_{13}(Fe,Ni)_4$  monoklinskim aproksimantima dekahogonalnih kvazikristala. Dok su  $Al_{13}Fe_4$  kristali dobro uređeni, ternarna ekstenzija  $Al_{13}(Fe,Ni)_4$  sadrži zamrznuti nered i može se smatrati neuređenom verzijom  $Al_{13}Fe_4$ . Mjerenja Hallovog efekta ovisnog o kristalografskim smjerovima izvedena su duž  $a^*$ ,  $b$  i  $c$  smjerova monoklinske jedinične ćelije, gdje su ( $a^*$ ,  $c$ ) ravnine pakirane duž okomite  $b$  osi. Smjer pakiranja  $b$  je najvodljiviji smjer za električnu struju. Utjecaj nereda zamrznutog u  $Al_{13}(Fe,Ni)_4$  ogleda se, u usporedbi s uređenim  $Al_{13}Fe_4$ , u velikoj rezidualnoj otpornosti  $\rho(T \rightarrow 0)$ . Vrijednosti Hallovog koeficijenta su za sve kombinacije smjerova svojstvene metalima. Anizotropni Hallov koeficijent odražava složenu strukturu anizotropne Fermijeve površine koja je sastavljena od dijelova svojstvenih elektronima i onih svojstvenih šupljina. Ovisno o kombinaciji smjerova struje i magnetskog polja dominira jedan od doprinosa, elektronski ( $R_H < 0$ ) ili šupljinski ( $R_H > 0$ ), ili se ta dva doprinosa međusobno poništavaju ( $R_H \approx 0$ ).