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HALL EFFECT IN THE CRYSTALLINE ORTHOROMBIC *o*-$Al_{13}Co_4$ APPROXIMANT TO THE DECAGONAL QUASICRYSTALS

HALLOV EFEKT V KRISTALINIČNEM ORTOROMBIČNEM PRIBLIŽKU *o*-$Al_{13}Co_4$ DEKAGONALNIM KVAZIKRISTALOM

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We have investigated the anisotropic Hall effect of the *o*-$Al_{13}Co_4$ orthorhombic approximant to the decagonal phase. The crystalline-direction-dependent measurements were performed along the *a*, *b* and *c* directions of the orthorhombic unit cell. The Hall effect has been measured for all the combinations of the electrical current and magnetic field directions. The Hall coefficients R_H change with the crystallographic direction from negative electron-like or zero to positive hole-like for different combinations of the current and field directions. The results for the anisotropy of R_H is well correlated with the anisotropy of R_H in the *d*-Al-Ni-Co and *d*-Al-Cu-Co quasicrystals. The Hall coefficients of the o-$Al_{13}Co_4$ phase were compared to the literature data on single crystals of the $Al_{76}Co_{22}Ni_2$ and the $Al_{80}Cr_{15}Fe_5$ approximants to the decagonal quasicrystals, allowing a study of the evolution of the Hall coefficient with an increasing structural complexity and unit-cell size.

Keywords: complex intermetallics, quasicrystalline approximants, Hall effect

Raziskali smo anizotropni Hallov efekt v ortorombičnemu približku *o*-$Al_{13}CO_4$ dekagonalni fazi. Meritve, odvisne od kristalne orientacije, so bile izvršene v smereh *a, b* in *c* ortorombične osnovne celice. Hallov efekt je bil izmerjen za vse kombinacije električnega toka in smeri magnetnega polja. Hallov koeficient R_H se spremeni s kristalografsko orientacijo od negativnega podobnega elektronom, do pozitivnega, podobnega vrzelim, po različnih kombinacijah toka in smeri polja. Rezultati o anizotropiji za R_H se dobro korelirajo z anizotropijo R_H v kvazikristalih *d*-Al-Ni-Co in v *d*-Al-Cu-Co. Hallov koeficient faze *o*-$Al_{13}CO_4$ je bil primerjan s podatki iz literature o monokristalnih približkih $Al_{76}Co_{22}Ni_2$ in $Al_{80}Cr_{15}Fe_5$ dekagonalnim kvazikristalom, kar omogoča študij evolucije Hallovega koeficienta z naraščajočo strukturno kompleksnostjo in velikostjo osnovne celice.

Ključne besede: kompleksni intermetaliki, kvazikristalni približki, Hallov efekt

1 INTRODUCTION

Decagonal quasicrystals (*d*-QCs) can be structurally viewed 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. A consequence of this anisotropic structure are the anisotropic physical properties,[1–9] when measured along the different crystalline directions. The anisotropy of the Hall coefficient R_H of *d*-QCs is especially intriguing, 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*-AlNiCo, *d*-AlCuCo and *d*-AlSiCuCo, and is considered to be a universal feature of *d*-QCs.[5,6] The lack of translational periodicity within the quasiperiodic layers prevents any quantitative theoretical analysis of this phenomenon. The problem can, however, be overcome by considering periodic approximant phases to the *d*-QCs that are characterized by a large unit cell, which periodically repeats in space, while the structure of the unit cell closely resembles *d*-QCs. Atomic layers that correspond to the quasiperiodic layers are again stacked periodically and the periodicity lengths along the stacking direction are almost identical to those along the periodic direction of the *d*-QCs. Approximant phases thus offer a valid comparison with the *d*-QCs. Here, we report measurements of the anisotropic Hall coefficient of the orthorhombic o-$Al_{13}Co_4$ complex metallic alloy, which belongs 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 direction and a unit cell comprising 102 atoms. These measurements complement our previous work on the anisotropic Hall coefficient of the $Al_{76}Co_{22}Ni_2$ and $Al_{80}Cr_{15}Fe_5$ approximants to the decagonal quasicrystals. $Al_{76}Co_{22}Ni_2$ has two atomic layers within one periodic unit of ≈0.4 nm and 32 atoms in a relatively small unit cell,[10–12] and the $Al_{80}Cr_{15}Fe_5$ has six atomic layers within one periodic unit of ≈1.25 nm and 306 atoms in a giant unit cell.[13–14] The *o*-$Al_{13}Co_4$

phase with four atomic layers and 102 atoms in the unit cell is thus intermediate to the other two approximant phases in terms of 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 Hall coefficient of the approximant to the decagonal quasicrystals evolve with the increasing structural complexity and the unit cell size.

2 EXPERIMENTAL

The o-$Al_{13}Co_4$ single crystal used in our study was grown using the Czochralski technique and its structure matched well with the orthorhombic unit cell.[15] In order to perform crystalline-direction-dependent studies we cut three bar-shaped samples of dimensions 1×1×7 mm^3 from the ingot, with their long axes along three orthogonal directions. The long axis of the first sample was along the [100] stacking direction (designated as *a*), which corresponds to the pseudo-tenfold axis of the o-$Al_{13}Co_4$ 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 [010] (*b*) direction and the third one along the [001] (*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 Hall coefficients of the o-$Al_{13}Co_4$ approximant to the decagonal quasicrystals along the three principal orthorhombic directions of the unit cell. The Hall-effect measurements were performed using a five-point method with a standard ac technique in magnetic fields up to 1 T.[16] 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.

3 RESULTS

The temperature-dependent Hall coefficient $R_H = E_y/j_xB_z$ of the o-$Al_{13}Co_4$ is displayed in **Figure 1**. In order to determine the anisotropy of the 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 of a metal, in agreement with the electrical resistivity (**Figure 2a**), of the order 10^{-10} m^3 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. The room-temperature values of the Hall coefficient of these

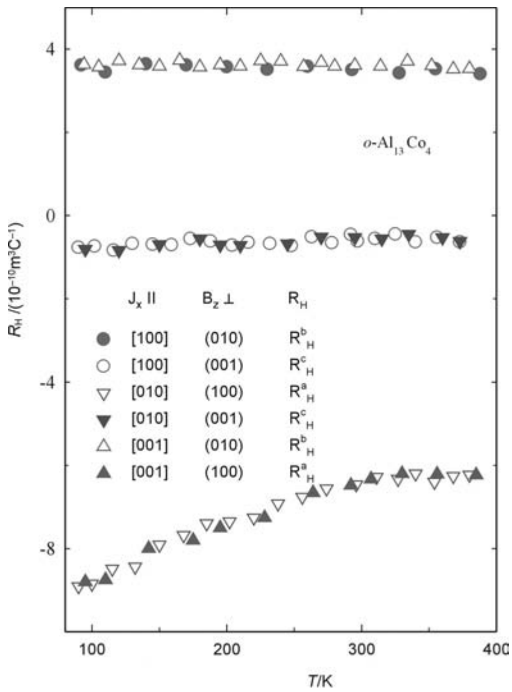

Figure 1: Anisotropic temperature-dependent Hall coefficient $R_H = E_y/j_xB_z$ of o-$Al_{13}Co_4$ for different combinations of direction *a, b, c,* of the current j_x and magnetic field B_z (given in the legend). The superscript *a*, *b* or *c* on R_H denotes the direction of the magnetic field.

Slika 1: Anizotropen tempereturno odvisen Hallov koeficient $R_H = E_y/j_xB_z$ za o-$Al_{13}CO_4$ za različne kombinacije smeri *a, b, c* toka j_x in magnetnega polja B_z (v legendi). Označba *a, b, c* na R_H je smer magnetnega polja.

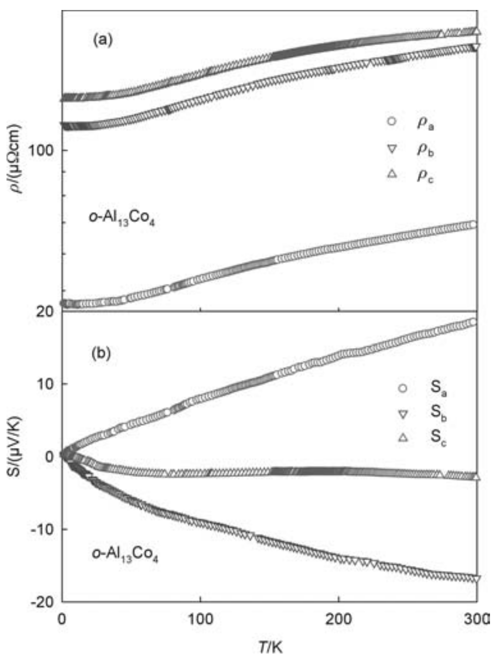

Figure 2: **(a)** Temperature-dependent electrical resistivity (*T*) of o-$Al_{13}Co_4$ along three orthogonal crystalline directions *a*, *b* and *c*. **(b)** Temperature-dependent thermoelectric power *S* of o-$Al_{13}Co_4$ along three orthogonal crystalline directions *a*, *b* and *c*

Slika 2: **(a)** Tempereturno odvisna električna upornost *(T)* za o-$Al_{13}CO_4$ v treh ortogonalnih kristalnih smereh *a, b, c*. **(b)** Temperaturno odvisna termoelekrična napetost *S* za o-$Al_{13}CO_4$ v treh ortogonalnih kristalnih smereh *a, b, c* pairs are $R^a_H = E_b/j_cB_a = E_c/j_bB_a = -6.5 \cdot 10^{-10}$ m^3 C^{-1}, $R^b_H = E_a/j_cB_b = E_c/j_aB_b = 3.5 \cdot 10^{-10}$ m^3 C^{-1} and $R^c_H = E_a/j_bB_c = E_b/j_aB_c = -0.6 \cdot 10^{-10}$ m^3 C^{-1}, where the additional superscript on the Hall coefficient denotes the direction of the magnetic field. R^b_H and R^c_H are practically temperature-independent within the investigated temperature range, whereas R^a_H shows a 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 $R^a_H < 0$ is electron-like for the magnetic field along the stacking *a* direction, whereas the positive $R^b_H > 0$ is hole-like for the field along the in-plane *b* direction. For the field along the second in-plane direction *c*, $R^c_H \approx 0$ 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 measured on the same specimens, presented in **Figure 2b**, which also changes sign with crystalline orientation. In both cases there is no simple explanation of this dual behavior, which would require knowledge of the details of the Fermi surface pertinent to the *o*-$Al_{13}Co_4$ phase.

4 DISSCUSION

We have measured the Hall coefficient of the orthorhombic *o*-$Al_{13}Co_4$ complex metallic alloy, with four atomic layers within one periodic unit. Our main objective was to determine the crystalline-direction-dependent anisotropy of the investigated Hall 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 Hall coefficient results point to a complicated Fermi surface that consists of electron-like and hole-like parts. Comparing the Hall coefficient of the three stacked-layer phases – the $Al_{76}Co_{22}Ni_2$ two-layer phase, the *o*-$Al_{13}Co_4$ four-layer phase and the $Al_{80}Cr_{15}Fe_5$ six-layer phase – some general conclusions can be drawn on the anisotropic Hall coefficient of the approximant to the decagonal quasicrystal with increasing structural complexity and unit-cell size. 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*-$Al_{13}Co_4$ 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 Hall coefficient 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 with the currently best-studied *d*-Al-Ni-Co-type *d*-QCs with two atomic layers within one periodic unit shows the following similarities: the Hall coefficient is the lowest, negative and electronic-like for the magnetic field along the periodic direction, whereas R_H changes sign to positive hole-like for the field along the in-plane directions.[5,6] This duality is suggested to be a universal feature of *d*-QCs.

5 CONCLUSION

The investigated approximants to the decagonal quasicrystals of increasing structural complexity exhibit an anisotropic Hall coefficient qualitatively similar to that of the decagonal quasicrystals. Both types of compounds have in common atomic planes that are stacked periodically. The stacked-layer structure appears to be at the origin of the anisotropy of the investigated Hall coefficient.

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