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THERMAL CONDUCTIVITY OF Al-Cr-Fe APPROXIMANT COMPOUNDS

ŽELJKO BIHAR[a], ANTE BILUŠIĆ[a], ANA SMONTARA[a,1] and JANEZ DOLINŠEK[b]

[a]*Institute of Physics, Bijenička 46, POB 304, HR–10 001 Zagreb, Croatia*

[b]*J. Stefan Institute, University of Ljubljana, Jamova 39, SI–1000 Ljubljana, Slovenia*
E-mail address: [1]*ana@ifs.hr*

Dedicated to the memory of Professor Zvonko Ogorelec

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We have investigated electrical resitivity and thermal conductivity of quasicrystalline compounds of the Al-Cr-Fe system: a gamma-brass phase (γ-AlCrFe) and a mixture of two orthorhombic approximants of the decagonal phase (O_1/O_2-AlCrFe) with properties which are in many respects intermediate to regular metals and quasicrystals. The electrical resistivities show very weak temperature dependence and the resistivity values are in between regular metals and aluminum-based quasicrystals. Thermal conductivity data show that the electronic and lattice contributions are of comparable size. While the electronic contribution can be described by the Wiedemann–Franz law, the lattice contribution can be reproduced by a sum of a long-wavelength phonons and hopping terms. At the lowest measured temperature, scattering of phonons on stacking-fault-like defects limits the heat transport. This type of defects has also been observed in the structural investigations. The results indicate that transport properties of quasicrystalline approximants are not determined by the short-range atomic order only, but are affected by both, the short-range quasiperiodic and long-range periodic atomic orders.

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1. Introduction

Quasicrystalline compounds exhibit a type of long-range order that is not compatible with translation periodicity [1]. Different kinds of quasicrystals were uncovered during the two last decades. Specifically, icosahedral quasicrystals and decagonal quasicrystals are rather frequent in aluminum-based alloys. Some are stable compounds, which may be prepared by slow cooling of liquid alloys followed by adequate heat treatment. Most frequently, quasicrystals are surrounded in the phase diagram by crystalline compounds of similar composition, called approximants, which present a closely related short-range order. Several such approximant crystals were discovered in the Al-Fe-Cr system [2]. Recently, two stable approximant phases of the Al-Cr-Fe system were investigated. The first (labeled O_1/O_2) is a large-unit-cell phase consisting of a mixture of two orthorhombic approximants of the decagonal phase, whereas the second is a γ-brass phase (a superstructure of the β-cubic phases), isostructural with rhombohedral Al_8Cr_5, which was considered as an approximant of both, icosahedral and decagonal quasicrystals. In the present article, we focus on the electrical and thermal conductivities of these compounds and compare them to the conductivities measured for regular metals and quasicrystals.

2. *Electrical resistivity*

The samples of compositions (at. %) $Al_{77.5}Cr_{16.5}Fe_6$ (noted O_1/O_2-AlCrFe hereafter), and $Al_{65}Cr_{28}Fe_7$ (noted γ-AlCrFe) were prepared by melting the pure constituents under helium gas atmosphere in an induction furnace equipped with a cold copper crucible. The as-cast ingots were then crushed into powder (granulometry range selected between 20 and 50 μm) and pressed in a graphite cell. They were sintered under purified helium using a uniaxial pressure of 15 MPa in order to produce ingots of 30 mm diameter. The sintering temperature was monitored at constant heating rate up to a plateau of 30 min duration at 980° C and 1060° C for the O_1/O_2-AlCrFe and γ-AlCrFe samples, respectively. Upon cooling, this step was followed by an annealing treatment of 2 h at 100° C below the plateau in order to eliminate all traces of undesirable phases [3 – 4]. After this preparation process, the AlCrFe ingots consisted of stable polycrystalline phases. Both samples under present investigation were shaped in the form of prisms, with dimensions 1.6 mm × 1.8 mm × 1.4 mm (γ-AlCrFe) and 2.0 mm × 1.9 mm × 5.9 mm (O_1/O_2-AlCrFe), respectively.

The electrical resistivity, $\rho(T)$, was determined in the temperature interval between 300 and 4 K using the standard four-terminal DC technique. The resistivities are displayed in Fig. 1, whereas in Fig. 2, the resistivities normalized to their room-temperature values are shown. The resistivity of the γ-AlCrFe sample at 300 K amounts 2.8 $\mu\Omega$m and exhibits a small positive temperature coefficient, resembling regular, periodic metals. A change of slope at about 70 K is evident. In Fig. 2 it is observed that the total variation of the resistivity is rather small, reaching 93 % of the room temperature value at 4 K. The resistivity of the O_1/O_2-AlCrFe is larger, amounting to $\rho_{300\,\mathrm{K}} = 4.2$ $\mu\Omega$m. Its resistivity exhibits a small, negative temperature coefficient from 300 to 160 K (Fig. 2), stays approximately constant between 160 and 30 K and shows an insignificant drop below 30 K. The total variation in the resistivity across the whole investigated range is very small, amounting only to 4 % of the room temperature value. The negative temperature coefficient of resistivity suggests that the O_1/O_2-AlCrFe sample is similar to dis-

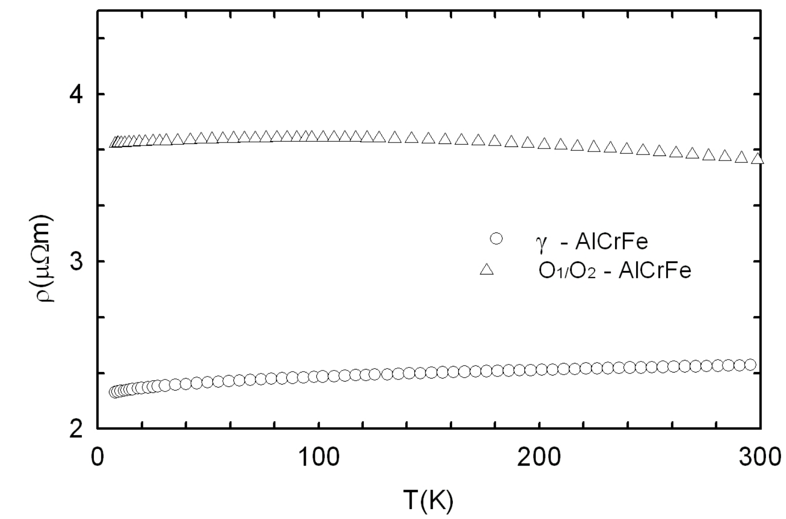

Fig. 1. Temperature dependent electrical resistivities, $\rho(T)$.

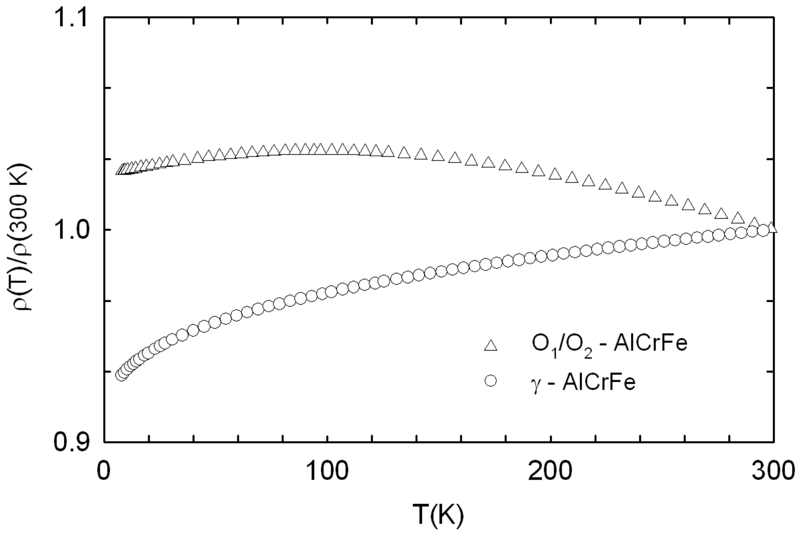

Fig. 2. Temperature dependent electrical resistivities, $\rho(T)/\rho(300\,\mathrm{K})$, normalized to the 300 K *value.*

ordered metals and/or quasicrystals, where high-quality icosahedral quasicrystals show particularly large negative temperature coefficient. In view of the structural disorder [5], recently found in this sample as well as the existing quasiperiodic short-range order on the scale of interatomic distances, which both promote a negative temperature coefficient of the resistivity, we are not able to tell which of these plays the dominant role. Most likely, however, it is a combination of both. The fact that the overall temperature dependence of the resistivities of both, the γ-AlCrFe and the O_1/O_2-AlCrFe approximants, are so weak, offers the following qualitative explanation. This kind of resistivity can be considered as an almost perfect balance of a regular metallic positive temperature coefficient of the resistivity due to the scattering of conduction electrons by phonons and/or localized lattice vibrations, and of the negative temperature coefficient of the resistivity due to gradual temperature-dependent electron localization by structural disorder and/or quasiperiodicity, typical of disordered metals and quasicrystals. This kind of duality can be easily anticipated for quasicrystalline approximants which are periodic on a large scale, promoting a metallic positive temperature coefficient of the resistivity and quasiperiodic on a small scale, promoting a negative temperature coefficient of the resistivity typical of quasicrystals. The considerably smaller unit–cell size of the γ-AlCrFe sample (which also displays quasiperiodic short-range order) makes this sample more similar to regular periodic solids, so that the metallic contribution in this sample is larger than that in the large-unit-cell system O_1/O_2-AlCrFe. Consequently, the positive temperature coefficient appears to dominates the overall resistivity behaviour of the γ-AlCrFe sample. The stronger metallic character of the γ-AlCrFe is also consistent with its smaller resistivity value. The relatively strong metallic contribution to the resistivities of the investigated γ-AlCrFe and O_1/O_2-AlCrFe samples is also evident from the absolute values of their resistivities, which are one to two orders of magnitude smaller than those of icosahedral aluminum-based quasicrystals [6].

3. *Thermal conductivity*

The thermal conductivities were measured between 300 and 8 K using an absolute steady-state heat-flow method. The thermal flux through the samples were generated by a 1 kΩ RuO_2 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 samples was monitored by a chromel-constantan differential thermocouple (more detailed description can be found elsewhere [7]). The temperature dependence of the measured thermal conductivity of both samples is displayed in Fig. 3. The thermal conductivity values at room temperatures are 6.0 W/mK and 4.2 W/mK, for the γ-AlCrFe and O_1/O_2-AlCrFe samples, respectively. These low thermal conductivity values are similar to those of quasicrystals, where they are considered to be a consequence of both the low electronic density of states at Fermi energy and nonperiodicity of the lattice, making the electronic and lattice contributions to the heat transport small [8]. The electronic contribution to the thermal conductivity, $K_{\mathrm{el}}(T)$, can be calculated using the Wiedemann–Franz law and the measured electrical resistivity, presented in Fig. 1, $K_{\mathrm{el}} = L_0 T/\rho$, where $L_0 = 2.45\times10^{-8}$ WΩ/K^2. The lattice contribution to the thermal conductivity, $K_{\mathrm{meas}} - K_{\mathrm{el}}$, is analyzed, in analogy to quasicrystals and amorphous solids, by considering propagation of long-wavelength phonons within the Al-Cr-Fe structure and, at elevated temperatures, hopping of localized vibrations. While the long-wavelength phonons may be ana-

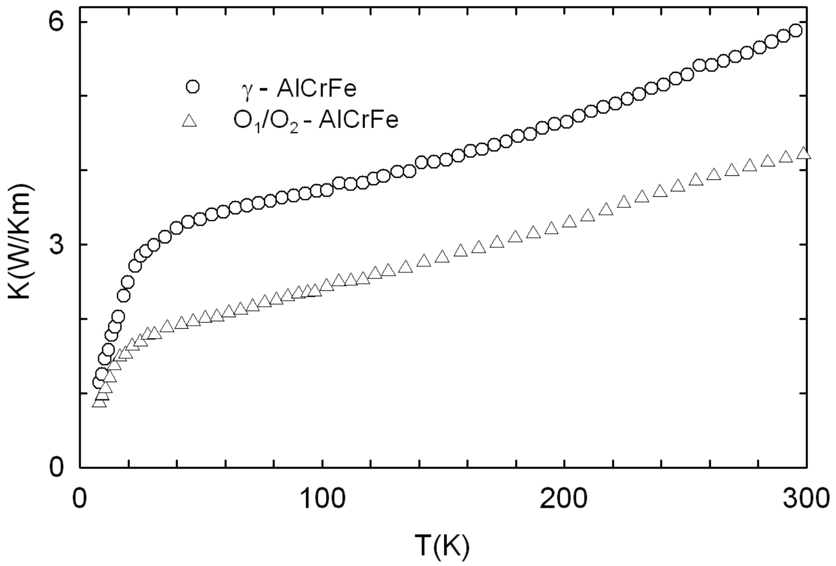

Fig. 3. Temperature dependent measured thermal conductivities, $K_{\mathrm{meas}}(T)$*, of the* γ-AlCrFe *and* $\mathrm{O_1/O_2}$-AlCrFe *samples between 8 and 300* K.

lyzed within the Debye model, the thermal activation of local vibrational modes opens a channel for the heat transfer by hopping excitations, which generally can be described by an activation law with activation energy E_{a}. Within this model, hopping yields a contribution to the thermal conductivity $K_{\mathrm{hopp}} = K^0_{\mathrm{hopp}} \exp\{-E_{\mathrm{a}}/(k_{\mathrm{B}}T)\}$, where K^0_{hopp} is a constant. The Debye thermal conductivity is written as [9]

$$K_{\mathrm{D}} = C_{\mathrm{D}} T^3 \int\limits_0^{\Theta/T} \tau(x) \frac{x^4 \mathrm{e}^x}{(\mathrm{e}^x - 1)^2}\, \mathrm{d}x, \tag{1}$$

where $C_{\mathrm{D}} = k_{\mathrm{B}}^4/(2\pi^2 \bar{v} \hbar^3)$, $\bar{v}$ is the average sound velocity, Θ the Debye temperature, τ the phonon relaxation time and $x = \hbar\omega/(k_{\mathrm{B}}T)$, where $\hbar\omega$ is the phonon energy. The different phonon scattering processes are incorporated in the relaxation time $\tau(x)$ and we assume that the Matthiessen's rule is valid, $\tau^{-1} = \sum \tau_j^{-1}$, where τ_j^{-1} is a scattering rate related to the j-th scattering channel. In amorphous solids and quasicrystals, the two dominant phonon scattering processes at low temperatures are Casimir scattering at the sample boundaries and scattering on tunnelling states. As in our experiment, performed between 300 and 8 K, we do not really enter the low temperature regime, these two processes may be neglected. However, two other processes are known that dominate thermal conduction in the investigated temperature regime. The back-scattering images of the γ-AlCrFe and

O_1/O_2-AlCrFe sintered-powder samples show porosity on the micrometer scale at the grain boundaries. In the O_1/O_2-AlCrFe sample there exist also phase boundaries between the O_1 and O_2 phases. In addition, metallographic observation of the γ-AlCrFe sample clearly shows a twinning- or stacking-faults structure of the material [5]. We consider all these structural defects to be stacking-faults-like, for which the scattering rate is given by [10], $\tau_{\mathrm{sf}}^{-1} = (7/10)(a^2/\bar{v})\gamma^2\omega^2 N_{\mathrm{s}}$, where a is a lattice parameter, γ the Grüneisen parameter and N_{s} the linear density of stacking faults. The second scattering mechanism are the umklapp processes. In periodic crystals, the acoustic vibrational spectrum exhibits a gap only at the Brillouin zone boundary and the umklapp scattering rate is described by an exponential factor, $\tau_{\mathrm{um}}^{-1} \propto \omega^2 T \exp(-\Theta_D/\beta T)$, where β is a dimensionless parameter of the order one. In quasicrystals, on the other hand, the vibrational spectrum contains a dense distribution of energy gaps, which implies that umklapp-type phonon scattering processes are much more frequent. The model of quasiumklapp phonon scattering in a quasiperiodic lattice yields power-law-type frequency and temperature dependence of the scattering rate $\tau_{\mathrm{um}}^{-1} \propto \omega^2 T^4$. Since the local atomic order in systems is quasicrystals-like, one can expect a power-law-type umklapp scattering rate also in this compound. As many different power-law expressions, such as $\tau_{\mathrm{um}}^{-1} \propto \omega^3 T$, $\omega^2 T^2$, $\omega^2 T^4$, can be found in the literature, we assume phenomenologically $\tau_{\mathrm{um}}^{-1} \propto B x^\alpha T^4$, where the exponent α (yielding the power-law dependence $\tau_{\mathrm{um}}^{-1} \propto \omega^\alpha T^{4-\alpha}$) should be determined from the fit.

The thermal conductivity data of Fig. 3 were fitted using the expression

$$K_{\mathrm{meas}}(T) = K_{\mathrm{el}}(T) + K_{\mathrm{D}}(T) + K_{\mathrm{h}}(T), \tag{2}$$

where the scattering rate in $K_{\mathrm{D}}(T)$ is the sum $\tau^{-1} = \tau_{\mathrm{sf}}^{-1} + \tau_{\mathrm{um}}^{-1}$. The Debye temperatures of the related Al-Pd-Mn and Al-Cu-Fe quasicrystals were commonly found close to 500 K. We assumed that the situation is similar also in our Al-Cr-Fe samples and made the fit with a fixed $\Theta = 500$ K value. Since our thermal conductivity data are available only up to 300 K, it turned out that the fit was very insensitive to the change of this Θ value. The Debye constant C was also not taken as a free parameter, but instead it was calculated by using $\bar{v} = 4000$ ms^{-1}, a very common value for aluminum-based quasicrystals obtained from ultrasonic data [11]. There still remains a number of parameters involved in the fitting procedure – K_{hopp}^0 and E_{a} for the hopping contribution and A, B and α for the Debye contribution. In spite of the excellent fits to the thermal conductivity data obtained with Eq. (2) for $K(T)$ to be presented in the following, and of the reasonable values of the fit parameters, the fits should not be considered more than just qualitative.

The theoretical fits of thermal conductivity for the γ-AlCrFe (Fig. 4) and O_1/O_2-AlCrFe (Fig. 5) samples show good agreement with the experimental data in the whole investigated temperature range, and the fit parameters are given in

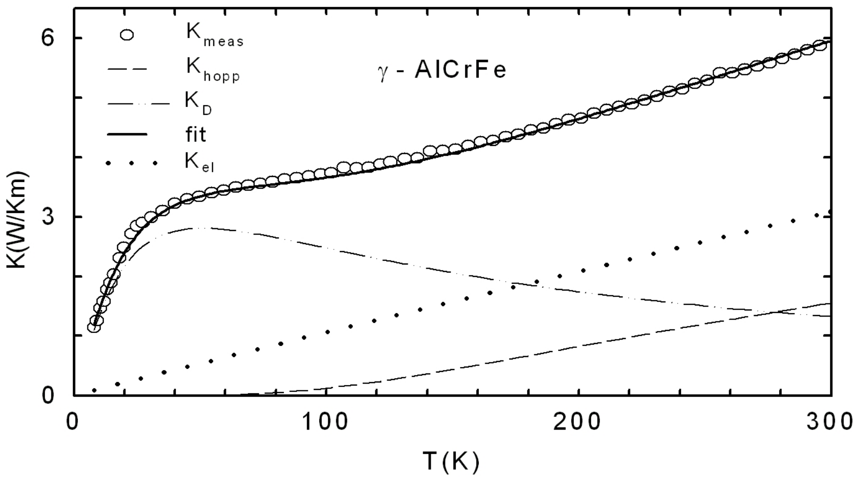

*Fig. 4. Temperature-dependent measured thermal conductivity data of the γ-*AlCrFe *sample reproduced theoretically (solid line) by Eq. (2). The three contributions to the measured thermal conductivity $K_{\rm meas}$ (circles) are shown separately: the electronic contribution $K_{\rm el}$ – dotted line, the Debye contribution $K_{\rm D}$ – dashed-dot dot line and the hopping contribution $K_{\rm hopp}$ – dashed line.*

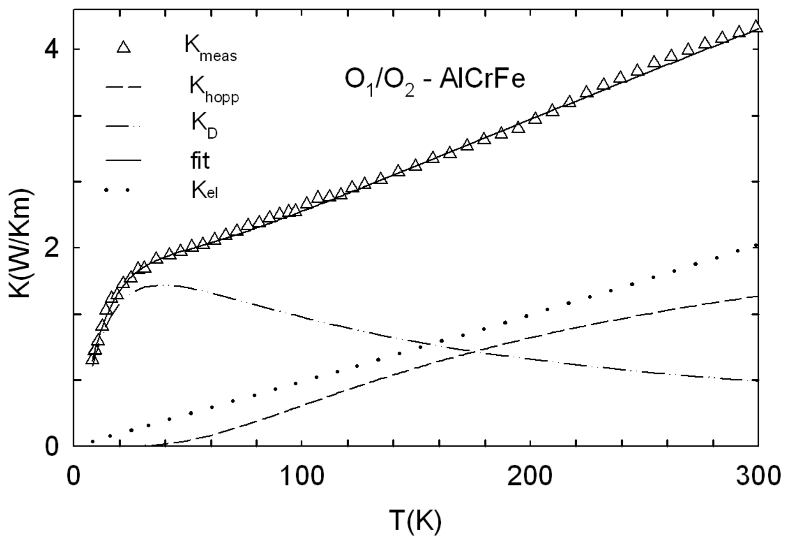

Fig. 5. Temperature-dependent thermal conductivity data of the O_1/O_2-AlCrFe *sample reproduced theoretically (solid line) by Eq. (2). The three contributions to the measured thermal conductivity $K_{\rm meas}$ (circles) are shown separately: the electronic contribution $K_{\rm el}$ – dotted line, the Debye contribution $K_{\rm D}$ – dashed-dot dot line and the hopping contribution $K_{\rm hopp}$ – dashed line.*

TABLE 1. Fit parameters of the thermal conductivity from Figs. 4 and 5.

Parameter	γ-AlCrFe	O_1/O_2-AlCrFe
E_a	15.3 meV	15.6 meV
K^0_{hopp}	5.3 W/mK	3.1 W/mK
A	0.9×10^7 s^{-1}K^{-2}	1.1×10^7 s^{-1}K^{-2}
B	5.4×10^3 s^{-1}K^{-4}	9.5×10^3 s^{-}K^{-4}
α	2.0	2.85

Table 1. In both graphs, the separate contributions of $K_{\mathrm{el}}(T)$, $K_{\mathrm{D}}(T)$ and $K_{\mathrm{hopp}}(T)$ to the total $K(T)$ are also displayed. The Debye contribution $K_{\mathrm{D}}(T)$ shows a maximum and a decrease at higher temperatures. Such behaviour is commonly found in periodic solids, where it is attributed to *umklapp* processes and phonon-phonon scattering [12]. For both samples, the fracton hopping contribution K_{hopp} becomes significant above about 100 K, so that it does not interfere much with the lattice contribution, which exhibits maximum at temperatures close to 40 K. The activation energies for the hopping were found similar for both samples, amounting $E_{\mathrm{a}}^{\gamma} = 15.3$ meV and $E_{\mathrm{a}}^{O_1/O_2} = 15.6$ meV. These values correlate with the inelastic neutron [13 – 14] and X-ray [16] scattering data on icosahedral Al-Pd-Mn quasicrystals, where dispersionless vibrational states were identified for energies higher than 12 meV. Such dispersionless states are considered to be a consequence of a dense distribution of energy gaps in the phonon excitation spectrum, which prevents extended phonons to propagate through the lattice, whereas localized vibrations may still be excited. The parameter A, which is for both samples close to 10^7 s^{-1}K^{-2}, allows estimating the linear density of stacking faults N_{s}. We take typical values for the lattice parameter $a \approx 1$ nm and the Grüneisen parameter $\gamma \approx 2$ and get $N_{\mathrm{s}} = 0.8\ \mu\mathrm{m}^{-1}$. This micrometer-scale value compares well with the μm-scales on which porosity, phase boundaries and twinning or stacking faults structure of the investigated materials are observed [5]. These structural defects (of extrinsic origin) may thus be considered as the source of the phonon scattering at low temperatures in the γ-AlCrFe and O_1/O_2-AlCrFe samples. The parameters B and α define phonon scattering by *umklapp* processes in a phenomenological way. For the γ-AlCrFe, $\alpha \approx 2$ so that $\tau_{\mathrm{um}}^{-1} \propto T^2$, whereas for the O_1/O_2-AlCrFe, $\alpha = 2.85$ and $\tau_{\mathrm{um}}^{-1} \propto T^{1.15}$. Consequently, K_{D} (Figs. 4, 5) decays at temperatures above the maximum roughly as a power-law $1/T^x$ with x between 1 and 2. This is qualitatively similar to regular periodic solids, where the rate of decline is generally given by [12] $K_{\mathrm{lat}} \approx 1/T^x$ with the x in the same interval.

In addition, we compare the ratio of the electronic and lattice contributions to the thermal conductivity of the γ-AlCrFe and O_1/O_2-AlCrFe approximants. At 300 K we find, for both samples, $K_{\mathrm{el}}/(K_{\mathrm{meas}} - K_{\mathrm{el}}) \approx 0.7$, thus they are of comparable value. This is different from both, simple metals, where the electronic contribution is usually $1-2$ orders of magnitude larger than the lattice contribution, and aluminium-based quasicrystals, where electrons carry less than 1 % of the heat. From this point of view, the investigated Al-Cr-Fe quasicrystalline approximants lie somewhere in between regular metals and quasicrystals.

4. Conclusion

The investigated Al-Cr-Fe quasicrystalline materials are sintered powders, showing inevitable micrometer-scale porosity at the grain boundaries [5]. The γ-AlCrFe is well-ordered, single-phase material, but exhibits twinning or stacking-fault structure, while the O_1/O_2-AlCrFe is a two-phase material, exhibiting phase boundaries, significant disorder and inclusion of small aluminum precipitates. These structural defects can significantly affect the intrinsic transport properties of the investigated samples. Indeed, they exhibit transport properties that are in many respects intermediate to regular metals on one hand and to qauasicrystals on the other hand. The electrical resistivity values of the γ-AlCrFe and O_1/O_2-AlCrFe samples were found to be $1-2$ orders of magnitude larger than those of simple metals and $1-2$ orders smaller than those of aluminium-based qauasicrystals. The temperature variation of the resistivity is very weak, amounting to few % in the investigated range from 300 to 4 K, which is intermediate to simple metals (showing generally large positive temperature coefficients) and quasicrystals (characterized by negative temperature coefficients). Regarding the thermal conductivity, the electronic and the lattice contributions were found to be of comparable size, classifying the Al-Cr-Fe approximants in between regular metals (where the electronic contribution is dominant) and quasicrystals (dominant lattice contribution). The electronic contribution can be well described by the Wiedemann–Franz law, the lattice contribution can be reproduced by a sum of a long-wavelength phonons and localized-vibrations hopping terms. The hopping is the dominant lattice heat-carrying channel at elevated temperatures (above 100 K), and the long-wavelength phonons dominate at low temperatures exhibiting a typical maximum in $K_\mathrm{D}(T)$ at temperature close to 40 K. At the lowest measured temperature, scattering of phonons on stacking-fault-like defects limits the heat transport, whereas at higher temperatures, *umklapp* processes become excited.

In conclusion, our results give a direct indication that the transport properties of the quasicrystalline approximants are not determined by the short-range atomic order only, as sometimes considered, but are affected by both the short-range quasiperiodic and long-range periodic atomic orders.

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TOPLINSKA VODLJIVOST FAZNO-DIJAGRAMSKIH BLIZNACA Al-Cr-Fe

Istraživali smo električnu otpornost i toplinsku vodljivost kvazikristalnih spojeva sustava Al-Cr-Fe: γ-fazu (γ-AlCrFe) i smjesu dvaju ortorombskih faznodijagramskih bliznaca dekagonalne faze (O_1/O_2-AlCrFe) čija su svojstva u mnogim pogledima između običnih metala i kvazikristala. Električne otpornosti pokazuju vrlo slabu temperaturnu ovisnost i njihove su vrijednosti između onih običnih metala i kvazikristala na osnovi aluminija. Podaci o toplinskoj vodljivosti pokazuju da su doprinosi elektrona i rešetke podjednaki. Elektronski doprinos može se opisati Wiedemann–Franzovim zakonom, a doprinos rešetke zbrojem doprinosa dugovalnih fonona i članova od preskakanja. Na najnižoj temperaturi mjerenja, raspršenje fonona na nepravilnostima rešetke sličnima defektima u slaganju atoma, ograničuje vođenje topline. Ta je vrsta nepravilnosti opažena i u istraživanjima strukture. Rezultati pokazuju da transportna svojstva kvazikristalnih bliznaca nisu određena samo kratko-dosežnom atomskom strukturom, već ovise i o kratko-dosežnim kvaziperiodičnim i o dugodosežnim periodičnim atomskim strukturama.