We have investigated anisotropic physical properties (the magnetic susceptibility, the electrical resistivity, the thermoelectric power, the Hall coefficient, and the thermal conductivity) of the single-crystalline Taylor-phase $T$-$Al_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ complex intermetallic that is an orthorhombic approximant to the $d$-$Al$-$Mn$-$Pd$ decagonal quasicrystal. The measurements were performed along the $a$, $b$, and $c$ directions of the orthorhombic unit cell, where ($a$, $c$) atomic planes are stacked along the perpendicular $b$ direction. The $T$-$Al_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ shows spin-glass behavior below the spin-freezing temperature $T_f=29$ K with a small anisotropy in the magnetic susceptibility. The anisotropic electrical resistivities are rather large and show negative temperature coefficient. The resistivity is lowest along the stacking direction, which appears to be a common property of the decagonal-approximant phases with a stacked-layer structure. The temperature-dependent resistivity was theoretically reproduced by the quantum transport theory of slow charge carriers. The thermopower is positive for all three crystallographic directions, indicating that holes are the majority charge carriers, and no anisotropy can be claimed within the experimental precision. The same conclusion on the holes being the dominant charge carriers follows from the Hall-coefficient measurements, which is a sum of the (positive) normal Hall coefficient and the anomalous term, originating from the magnetization. The anisotropy of the thermal conductivity is practically negligible. The $T$-$Al_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ Taylor phase can be considered as a “close-to-isotropic” complex intermetallic. The relation of the anisotropic physical properties of the Taylor phase to other families of decagonal-approximant phases with the stacked-layer structure is discussed.

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I. INTRODUCTION

Decagonal quasicrystals (d-QCs) can be structurally viewed as a periodic stack of quasiperiodic atomic planes, so that $d$-QCs are two-dimensional (2D) quasicrystals, whereas they are periodic crystals in a direction perpendicular to the quasiperiodic planes. A consequence of the structural anisotropy are anisotropic magnetic, electrical, and thermal transport properties, when measured along the periodic and quasiperiodic crystallographic directions. The lack of translational periodicity within the quasiperiodic planes prevents any quantitative theoretical analysis of the anisotropic physical properties of $d$-QCs. The problem can be overcome by considering approximant phases to the decagonal phase, for which—being periodic solids in three dimensions (3D)—theoretical simulations are straightforward to perform. Approximant phases are characterized by large unit cells, which periodically repeat in space with the atomic decoration closely resembling that of the $d$-QCs. Atomic layers are again 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 patterns, so that their structures on the scale of near-neighbor atoms closely resemble each other. Decagonal approximants thus furnish valid comparison to the $d$-QCs.

The degree of anisotropy of the physical properties of $d$-QCs is related to the structural details of a particular decagonal phase, depending on the number of quasiperiodic layers in one periodic unit. The most anisotropic case are 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. Other $d$ phases contain more quasiperiodic layers in a periodic unit and show smaller anisotropies. In $d$-$Al$-$Co$, $d$-$Al$-$Ni$, and $d$-$Al$-$Si$-$Cu$-$Co there are four quasiperiodic layers with the periodicity about 0.8 nm; $d$-$Al$-$Mn$, $d$-$Al$-$Cr$, and $d$-$Al$-$Mn$-$Pd$ phases contain six layers with the periodicity of about 1.2 nm, whereas $d$-$Al$-$Pd$ and $d$-$Al$-$Cu$-$Fe$ phases contain eight layers in a periodicity length of 1.6 nm. Recently, large single-crystalline samples of several decagonal-approximant phases were successfully grown and their anisotropic physical properties (the magnetic susceptibility, the electrical resistivity, the thermoelectric power, the Hall coefficient and the thermal conductivity) were measured along three orthogonal crystallographic directions. The first was the $Al_{76}$Co$_{22}$Ni$_{2}$ compound, known as the Y-phase of Al-Ni-Co (denoted as Y-Al-Ni-Co), which belongs to the $Al_{13}$TM$_{4}$ (TM=transition metal) class of complex intermetallics and is a monoclinic approximant to the decagonal phase 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 second group were the orthorhombic $o$-$Al_{13}$Co$_{4}$ (Ref. 5), the monoclinic $Al_{13}$Fe$_{4}$ and its ternary extension $Al_{13}$Fe$_{4}$Ni$_{4}$ (Ref. 6) decagonal approximants, also belonging to the $Al_{13}$TM$_{4}$ class of intermetallics, but comprising four atomic layers.
The T-phase in the Al-Mn-Fe system is stable at high temperatures only, similar to the T-phase in the binary Al-Mn system. It is observed at room temperature (RT) only due to rapid quenching of the material after the growth. On the other hand, the T-phase in the Al-Mn-Pd system remains stable also at lower temperatures. From this point of view, the T-phase in the Al-Mn-Fe system is more similar to the binary T-phase in the Al-Mn system than to the ternary one in the Al-Mn-Pd system. The reason is probably the fact that the similarity between the manganese and iron atoms is higher than the similarity between the manganese and palladium atoms; iron is just beside manganese in the periodic table. It was also reported that the Al72.5Mn21.5Fe6 composition behaves similar to additional annealing of the quenched T-Al73Mn21Fe6 Taylor phase resulted in a transformation to the decagonal phase.

Our study of the anisotropic physical properties of the Taylor phase was performed on an oriented single crystal T-Al73Mn21.5Fe6.0 grown by the Czochralski technique. The crystal was pulled from a melt of composition Al73Mn21Fe6 at a rate of 1 mm/h under Argon atmosphere. The growth was performed in a temperature range between 1100 and 1040 °C according to the phase-diagram information and growth conditions reported by Balanetskyy and co-workers. The material was not annealed after growth. In order to perform crystallographic-direction-dependent studies, three rectangular bar-shaped samples of dimensions 2 × 1 × 1 mm³ were cut from the oriented single crystal with their long axes along the a, b, and c crystallographic directions, where for each sample, the orientation of the other two axes was known as well. The so-prepared samples enabled us to determine the anisotropic physical properties of the T-Al73Mn21Fe6 Taylor phase along the three orthogonal directions.

Recently we have reported the magnetic properties of a polygran binary T-Al73Mn and a ternary T-Al73(Mn,Pd) and T-Al73(Mn,Fe) series of samples of compositions T-Al73Mn27−xPdx (x = 2, 4, 6) and T-Al73Mn27−xFe6 (x = 2, 4) and the decagonal quasicrystal d-Al73Mn27Fe6. All samples exhibited a pronounced magnetic memory effect that has lead to the application of this material for the thermal storage of digital information, serving as a new kind of a digital memory element—a thermal memory cell.

III. ANISOTROPIC PHYSICAL PROPERTIES OF THE SINGLE-CRYSTALLINE T-Al72.5Mn21.5Fe6.0

A. Magnetization and magnetic susceptibility

Magnetic properties of the polygran binary T-Al73Mn and the ternary T-Al73Mn27−xPdx (x = 2, 4, 6), T-Al73Mn27−xFex
of the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6}$. The results can be summarized as follows. (1) All samples show a transition to a spin-glass-type nonergodic phase at the spin-freezing temperature $T_f \approx 20–23$ K, where below $T_f$, the zero-field-cooled (zfc) and field-cooled (fc) magnetic susceptibilities become unequal. This shows frustration of interactions between the magnetic moments of the Mn and Fe ions. (2) Within the nonergodic phase, typical broken-ergodicity phenomena were observed, including the frequency-dependent freezing temperature, $T_f(\omega)$, hysteresis and remanence, ultraslow decay of the thermoremanent magnetization, the memory effect (a state of the spin system reached upon isothermal aging can be retrieved after a negative temperature cycle) and rejuvenation (small positive temperature cycle within the nonergodic phase erases the effect of previous aging). (3) The memory effect was employed to construct a new kind of a memory element using the T-Al$_3$(Mn,Fe) material, where an eight-bit byte of digital information was stored by a specific temperature-time profile in the absence of any external electric, magnetic, or electromagnetic field. In this way, arbitrary ASCII characters, representing text in computers, were successfully written by pure thermal manipulation of the Taylor-phase polygran material. Our single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ sample was subjected to the same set of magnetic experiments as the polygran material studied in Ref. 15, and the results are in complete accordance with those of the polygran samples. Since the polygran and single-crystalline morphologies mainly affect the long-range electrical and thermal transport properties of the material, where the electrons and phonons travel over macroscopic distances, no pronounced differences between the magnetic properties of the polygran and the single-crystalline samples are expected. We, therefore, do not repeat here the general magnetic results of the Taylor phase, but rather concentrate on the anisotropy of magnetic properties of the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$.

Magnetic properties were investigated in the temperature interval between 300 and 2 K and magnetic fields up to 50 kOe using a Quantum Design MPMS XL-5 SQUID magnetometer. In the orientation-dependent measurements, the magnetic field was directed along the long axis of each sample, thus along the $a$, $b$, and $c$ crystallographic directions.

1. zfc and fc magnetic susceptibilities

The zfc and fc susceptibilities $\chi=M/H$ in the temperature range 2–50 K in magnetic fields $H=100$ Oe and 50 kOe are shown in Fig. 1. In the following, the anisotropic susceptibilities are labeled by the index $k$, denoting the direction of the magnetic field with respect to the crystallographic axes ($k=a, b, c$). In the low field of 100 Oe [Fig. 1(a)], $\chi_{a,b,c}^{\text{zfc}}$ exhibit a pronounced cusp at the spin-freezing temperature $T_f=29$ K, whereas $\chi_{a,b,c}^{\text{fc}}$ remain more or less constant below $T_f$. At $T_f$, the system becomes nonergodic and $\chi_{a,b,c}^{\text{zfc}}$ and $\chi_{a,b,c}^{\text{fc}}$ become unequal. The orientation-dependent susceptibilities show the following anisotropy: there is practically no anisotropy between the two in-plane directions $a$ and $c$, whereas there exists a moderate anisotropy between the in-plane and the stacking $b$ directions, where the susceptibility along $b$ is somewhat higher. The temperature-dependent susceptibility difference $\chi_{b}^{\text{zfc}} - \chi_{b}^{\text{fc}}$ below $T_f$ does not show anisotropy and there is also no anisotropy in the freezing temperature for the three crystallographic directions. The lack of anisotropy in $T_f$ was verified also from the orientation-dependent measurements of the ac susceptibility $\chi'$ (not shown), where the temperature of the cusp in $\chi'$ did not depend on the direction of the magnetic field. It is worth mentioning that $T_f=29$ K of the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is by $\Delta T=7$ K higher from that of the polygran decagonal quasicrystal d-Al$_{73}$Mn$_{21}$Fe$_{6}$ ($T_f=22$ K) of practically the same composition, studied in the preceding paper. Different structures of the two compounds, the small difference in the transition-metal (Mn+Fe) concentration and the extrinsic disorder effects in the polygran sample may be at the origin of this difference.

The $\chi_{a}^{\text{zfc}}$ and $\chi_{a}^{\text{fc}}$ in the high field of 50 kOe are displayed in Fig. 1(b). We observe that the cusp in $\chi_{a}^{\text{zfc}}$ has rounded and the zfc-fc splitting temperature has shifted to lower temperature of about 8 K. This strong influence of the external magnetic field demonstrates comparable strengths of the spin-spin exchange interactions (responsible for the spin-glass magnetic ordering in the absence of an external magnetic field) and the Zeeman interaction of spins with the magnetic field. The internal spin-glass-type frustrated magnetic structure is thus “soft” and fragile with respect to the external magnetic field. The anisotropy of the susceptibility is preserved in the high field [Fig. 1(b)]. There is again no anisotropy between the two in-plane directions $a$ and $c$ and a
moderate anisotropy to the stacking b direction. The susceptibility difference $\chi_{fc}^k - \chi_{zfc}^k$ does not show anisotropy either.

2. Hysteresis

The $M(H)$ curves for the field along the three crystallographic directions $a$, $b$, and $c$ in the sweep range $\pm 50$ kOe are indistinguishable within the experimental error of measurement. Using the Taylor-phase material for the thermal memory cell application is advantageous, as identical thermal-storage performance is obtained for the single crystalline and polygrain forms of the material, by showing the difference $M_{fc}(t_a=0) - M_{zfc}(t_a)$. Between the reference (no-stop) $zfc$ magnetization curve $M_{fc}(t_a=0)$ and the $zfc$ magnetization curves $M_{zfc}(t_a)$ with a stop at the aging temperature $T_f=14$ K for aging times $t_a$ between 1 min and 4 h. For more details on the memory effect see Ref. 15.

Our ME experiments on the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ (T$_f$=29 K) were performed at a set of different aging temperatures $T_f$ between 1 K and aging times between 1 min and 4 h. No anisotropy of the ME between the three crystallographic directions was observed. This is evident from Fig. 3, where we show $\Delta M(t_a)$ curves for the $k=a$, $b$, and $c$ directions of the magnetic field in zero magnetic field and the cooling is stopped at the aging temperature $T_f$. The $\Delta M(t_a)$ curve is the source of the $M(H)$ hysteresis. The $M(H)$ loops remain open up to the highest applied field of 50 kOe, demonstrating predominant antiparallel (antiferromagnetic) coupling between the spins (for the parallel ferromagnetic-type coupling, the loops usually saturate at a much lower field of a few kOe only).

3. Memory effect

We discuss also the paramagnetic susceptibility, which will be employed for the analysis of the temperature-dependent Hall coefficient, to be presented in the following. The $zfc$ and $fc$ susceptibilities, $\chi_{zfc}$ and $\chi_{fc}$, in the temperature range 2–300 K, measured in a magnetic field $H = 1$ kOe along the three crystallographic directions $a$, $b$, and $c$ are displayed in Fig. 4(a). The experimental anisotropy of the magnetic susceptibility is rather small, less than 2%, and cannot be discerned from the graph. The temperature-dependent paramagnetic susceptibility in the high-temperature range $T > 60$ K was analyzed with the Curie-Weiss function.

FIG. 2. (Color online) The $M(H)$ hysteresis loops at $T=2$ K for the field along the three crystallographic directions $a$, $b$, and $c$ in the sweep range $\pm 50$ kOe. The three loops are practically indistinguishable on the graph.

FIG. 3. (Color online) Anisotropy of the memory effect in the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ by showing the difference $\Delta M = M_{fc}(t_a=0) - M_{zfc}(t_a)$ between the reference (no-stop) $zfc$ magnetization curve $M_{fc}(t_a=0)$ and the $zfc$ magnetization curves $M_{zfc}(t_a)$ with a stop at the aging temperature $T_f=14$ K for aging times $t_a$ between 1 min and 4 h. No anisotropy of the ME between the three crystallographic directions was observed. This is evident from Fig. 3, where we show $\Delta M(t_a)$ curves for the $k=a$, $b$, and $c$ directions of the magnetic field in zero magnetic field and the cooling is stopped at the aging temperature $T_f$. The $\Delta M(t_a)$ curve is the source of the $M(H)$ hysteresis. The $M(H)$ loops remain open up to the highest applied field of 50 kOe, demonstrating predominant antiparallel (antiferromagnetic) coupling between the spins (for the parallel ferromagnetic-type coupling, the loops usually saturate at a much lower field of a few kOe only).

4. Paramagnetic susceptibility

We discuss also the paramagnetic susceptibility, which will be employed for the analysis of the temperature-dependent Hall coefficient, to be presented in the following. The $zfc$ and $fc$ susceptibilities, $\chi_{zfc}$ and $\chi_{fc}$, in the temperature range 2–300 K, measured in a magnetic field $H = 1$ kOe along the three crystallographic directions $a$, $b$, and $c$ are displayed in Fig. 4(a). The experimental anisotropy of the magnetic susceptibility is rather small, less than 2%, and cannot be discerned from the graph. The temperature-dependent paramagnetic susceptibility in the high-temperature range $T > 60$ K was analyzed with the Curie-Weiss function.
where $\chi_0$ is the temperature-independent part (including the Larmor diamagnetic core susceptibility $\chi_{dia}$ and the Pauli paramagnetic and Landau diamagnetic susceptibilities of conduction electrons), $C$ is the Curie-Weiss constant and $\theta$ the Curie-Weiss temperature. Due to the very small anisotropy of the paramagnetic susceptibility, only one set of fit parameters ($\chi_0$, $C$, and $\theta$) was determined that should be considered valid for all three directions. The fit (solid curve in Fig. 4(a)) yielded the parameter values $\chi_0 = -4 \times 10^{-6}$ emu/mol, $C = 0.31$ emu K/mol, and $\theta = -27$ K. The negative $\theta$ value supports the consideration of a predominant antiferromagnetic coupling between the magnetic moments. The Larmor contribution was estimated theoretically from literature tables\textsuperscript{18} to be in the range $\chi_{dia} = [-3.8, -5.2] \times 10^{-6}$ emu/mol (depending on the Mn and Fe ionization states), which is about the same as $\chi_0$.

The $M(H)$ curves at $T = 300$ K in the paramagnetic phase for the field along the three crystallographic directions are shown in Fig. 4(b). The curves do not show any anisotropy and hysteresis, but a linear $M(H)$ relation typical of Curie-type paramagnets.

\begin{equation}
\chi = \chi_0 + \frac{C}{T - \theta},
\end{equation}

FIG. 4. (Color online) (a) The anisotropic zfc and fc susceptibilities in the temperature range 2–300 K, measured in a magnetic field $H = 1$ kOe along the three crystallographic directions $a$, $b$, and $c$. Solid curve is the Curie-Weiss fit with Eq. (1) in the high-temperature $T > 60$ K paramagnetic regime and the fit parameters are given in the text. For $T > 60$ K, the three sets of experimental data and the fitting curve are indistinguishable, so that the same fit is valid for all three directions. (b) The $M(H)$ lines at $T = 300$ K in the paramagnetic phase for the field along the three crystallographic directions. The three lines are indistinguishable on the graph.

The electrical resistivity of the single-crystalline $T$-$Al_{72.5}Mn_{21.5}Fe_{6.0}$ was measured between 300 and 2 K using the standard four-terminal technique. The $\rho(T)$ data for the current along the three crystallographic directions are displayed in Fig. 5. All resistivities show a negative temperature coefficient (NTC) by increasing with decreasing temperature. The resistivity is the lowest along the stacking $b$ direction perpendicular to the atomic planes, where its RT value amounts $\rho_{0}^{a} = 441$ $\mu\Omega$ cm and the 2 K resistivity is $\rho_{0}^{b} = 601$ $\mu\Omega$ cm, yielding the increase upon cooling by $(\rho_{k}^{b} - \rho_{0}^{b})/\rho_{0}^{b} = 40\%$. The two in-plane resistivities $\rho_{a}$ and $\rho_{c}$ are higher, amounting $\rho_{0}^{a} = 501$ $\mu\Omega$ cm and $\rho_{0}^{c} = 740$ $\mu\Omega$ cm with the increase $(\rho_{k}^{a} - \rho_{0}^{a})/\rho_{0}^{a} = 48\%$, whereas $\rho_{0}^{c} = 490$ $\mu\Omega$ cm and $\rho_{k}^{c} = 709$ $\mu\Omega$ cm with $(\rho_{k}^{c} - \rho_{0}^{c})/\rho_{0}^{c} = 45\%$. Considering the experimental error to be about 5\% (originating mainly from the uncertainty in the samples’ geometrical parameters), no pronounced anisotropy between the two in-plane $a$ and $c$ directions can be claimed, whereas the anisotropy to the stacking $b$ direction is significant, though still small (at 300 K, the $\rho_{b}$ value is 10\% smaller from $\rho_{a}$ and $\rho_{c}$). It is remarkable that this kind of anisotropy, where the resistivity is lowest along the stacking direction, whereas the in-plane resistivities are higher and show little or no anisotropy, appears to be a general feature of the decagonal-approximant phases with the stacked-layer structure. It was equally observed in the Y-Al-Ni-Co (Ref. 3) and the Al$_{13}$Cr$_{2}$Fe$_{4}$ (Ref. 6) and the Al$_{12}$Fe$_{2}$Co$_{x}$Ni$_{y}$ (Ref. 7) and the Al$_{12}$Cr$_{2}$Fe$_{4}$ (Ref. 8) decagonal approximants. It is also noteworthy that the anisotropic resistivity values of the investigated single-crystalline $T$-$Al_{73}Mn_{27}Fe_{6.0}$ from Fig. 5 are by factors 2–10 smaller from the resistivities of the polycrystal $T$-$Al_{3}Mn$, $T$-$Al_{12}Mn_{27}Fe_{5} (x = 2.4)$ and the decagonal quasicrystal $d$-$Al_{17}Mn_{21}Fe_{6}$ reported in the previous paper.\textsuperscript{17} Though different chemical compositions of the samples (and the structural difference in the case of the decagonal $d$-$Al_{17}Mn_{21}Fe_{6}$) are likely the dominant cause of these differences, the extrinsic effects due to the polycrystal morphology of the samples (grain boundaries) may add significantly to the higher resistivity of the polycrystal samples.
A straightforward way to analyze theoretically the anisotropic resistivity of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ single crystal is to calculate ab initio the anisotropic Fermi surface using published structural models of the T-phase and then perform calculation of the electronic transport coefficients (the electrical resistivity, the thermoelectric power, the Hall coefficient and the electronic contribution to the thermal conductivity) using appropriate transport theory, e.g., the semiclassical Bloch-Boltzmann theory. Such calculations were successfully performed for the Y-Al-Ni-Co (Refs. 3 and 4) and Al$_{4}$Co$_{4}$ (Ref. 5) decagonal approximants. In the case of the T-Al$_3$(Mn,Fe) phase, there are three major problems to perform such calculations. First, to calculate ab initio the Fermi surface, one has to have a reliable structural model of the T-phase for the particular composition Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ of our sample, which is not available at present. Second, in the unit cells of the related T-phase models, e.g., the model with composition Al$_{72.5}$Mn$_{21.5}$Pd$_{3.2}$ by Klein et al., most of the lattice sites show either fractional occupation or mixed TM/Al occupancy, creating structural and chemical disorder on the lattice. For the ab initio calculations, the unit cell should be well defined by having a particular fractionally occupied lattice site either occupied with an atom (occupation 1) or the site is empty (occupation 0) and the sites of mixed occupancy should either contain an Al or a TM atom. Since out of the 25 atomic sites in the unit cell of the Klein et al. model, 11 are fractionally occupied and 6 show mixed occupancy, many different variants of the unit cell can be constructed and most of them will give different results. Third, the NTC resistivity cannot be reproduced within the Bloch-Boltzmann transport theory, which is thus inapplicable to our T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ single crystal. In this situation, we proceed with the resistivity analysis in a semiquantitative way by employing the theory of quantum transport of slow charge carriers by Trambly de Laissardière et al., applicable to metallic alloys and compounds in which the electron mean-free path between the scattering events is small compared to the extension of the conduction-electron wave packet, in which case the electron propagation is non-Boltzmann (nonballistic) and the resistivity exhibits a NTC. The structural and chemical disorders in the Taylor phase due to the fractional and mixed occupancy of the lattice sites imply that the relaxation time τ between the scattering events is short, so that l=υτ (where υ is the electron velocity) is small and the short mean-free-path limit is met, hence justifying the use of the quantum transport theory. This theory has recently been successfully applied to the Al$_4$(Cr,Fe) decagonal approximant, where the temperature-dependent resistivity exhibited a maximum.

In an anisotropic crystal, the electrical conductivity σ (the inverse resistivity $ρ^{-1}$) is generally a symmetric (and diagonalizable) tensor, relating the current density $j$ to the electric field $E$ via the relation $j = ∑_{i=x,y,z}σ_{ij}E_i$, where $i,j = x,y,z$ denote three orthogonal directions in a Cartesian coordinate system. The tensorial ellipsoid exhibits the same symmetry axes as the crystallographic structure. For our orthorhombic T-Al$_3$(Mn,Fe) crystal this implies that the conductivity tensor is diagonal in the basis of the crystallographic directions $a$, $b$, and $c$. The geometry of our samples (their long axes were along the three crystallographic directions) and the direction of the electric field applied along their long axes imply that diagonal elements $σ_{xx} = σ_{yy} = σ_{zz}$ and $σ_{xy} = σ_{yx} = σ_{yz} = σ_{zy} = σ_{zx} = σ_{xz} = 0$, and $σ_{xx} = σ_{yy} = σ_{zz}$ were measured in our experiments, so that the entire conductivity tensor was determined. The quantum theory of slow charge carriers applies to any diagonal element of the conductivity tensor (in Ref. 19, $σ_{xx}$ is considered, but $x$ can be any crystallographic direction).

According to the Einstein relation, the conductivity $σ$ depends on the electronic density of states (DOS) $g(ε)$ and the spectral diffusivity $D(ε)$ within the thermal interval of a few $k_BT$ around the Fermi level $ε_F$. In the case of slowly varying metallic DOS around $ε_F$ it is permissible to replace $g(ε)$ by $g(ε_F)$. For the diffusion constant of slow charge carriers it was shown$^{10}$ that it can be written as $D = v^2τ + L^2(τ)/τ$, where $L^2(τ)$ is the non-Boltzmann (non-Boltzmann) contribution to the square of spreading of the quantum state at energy $ε$ due to diffusion, averaged on a time scale $τ$. $L(τ)$ is bounded by the unit cell length and saturates to a constant value already for short averaging time. The dc conductivity of the system in the crystallographic direction $j$ can be written as$^{19}$

$$σ_j = σ_{Bj} + σ_{NBj} = ε^2g(ε_F)υ_j^2τ_j + ε^2g(ε_F)L_j^2(τ_j)/τ_j,$$

(2)

where $σ_{Bj}$ is the Boltzmann contribution and $σ_{NBj}$ is the non-Boltzmann contribution. The scattering rate $τ_j^{-1}$ will generally be a sum of a temperature-independent rate $τ_j^{-1}$ due to scattering by quenched defects and a temperature-dependent rate $τ_j^{-1}$ due to scattering by phonons and the magnetic scattering by localized paramagnetic spins. The anisotropy of the atomic structure implies that the density of quenched defects and the phonon spectrum will also be anisotropic, so that the scattering rate will generally depend on the crystalline direction, $τ_j^{-1} = τ_{0j}^{-1} + τ_{pj}^{-1}$. In the simplest case, $τ_{pj}$ can be phenomenologically written as a power law of temperature, $τ_{pj} = β_j/T^{ν_j}$, at least within a limited temperature interval. Assuming that $L_j^2(τ_j)$ can be replaced by its limiting value, a constant $L_j^2$, Eq. (2) yields a minimum in the conductivity $σ_j$ as a function of $τ_j$ or temperature (or equivalently, there is a maximum in the resistivity) at the condition $τ_j = L_j/υ_j$. At temperatures below the resistivity maximum, $τ_j$ is long enough that the system is in the Boltzmann regime ($σ_j = σ_{Bj}$) with ballistic propagation of the electrons and a power-law positive-temperature-coefficient (PTC) metallic resistivity. At high temperatures above the resistivity maximum, $τ_j$ is short enough that the system is in the non-Boltzmann (nonballistic) regime and Eq. (2) yields the conductivity of the form

$$σ_j = B_j(1 + C_jT^{ν_j}),$$

(3)

with $B_j = ε^2g(ε_F)L_j^2/τ_{0j}$ and $C_j = τ_{0j}/β_j$ that describes an insulator-like NTC resistivity. The fits of the anisotropic resistivity of T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ using Eq. (3) are shown by solid curves in Fig. 5 and the fit parameters are given in Table I. Successful fits down to the lowest investigated temperature of 2 K demonstrate that the degree of structural and chemical disorder in the T-Al$_3$(Mn,Fe) phase is large enough that the system is in the non-Boltzmann regime.
TABLE I. Fit parameters of the electrical resistivity [solid curves in Fig. 5, as calculated from Eq. (5)]. The units of the coefficients $C_i$ are chosen such that the temperature in the product $C_i T^{\alpha_i}$ is dimensionless.

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within the whole investigated temperature range.

C. Thermoelectric power

The thermoelectric power (the Seebeck coefficient $S$) of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ single crystal was measured between 300 and 2 K by using a standard temperature-gradient technique. The thermopower data, measured along the three crystallographic directions $a$, $b$, and $c$, are displayed in Fig. 6. For all three directions, the thermopower is positive with the electric charge $e^+$ and thus insensitive to the sign of the electric charge, hence distinguished between the electrons and holes. Unlike the electrical resistivity that does not distinguish between the negative (electrons) and positive (holes) charge carriers [the electric charge in Eq. (2) appears as $e^+$ and is thus insensitive to the sign of the charge], the charge in the expression for the thermopower appears as $1/e$, hence distinguishing between the electrons and holes. For that reason, the thermopower reflects better the anisotropy of the Fermi surface of a complex metallic alloy that generally contains electronlike and holelike parts. The almost isotropic thermopower of Fig. 6 indicates that the anisotropy of the Fermi surface of T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is small. Moreover, the positive sign of the thermopower indicates that the holelike parts are dominant.

D. Hall coefficient

Hall coefficient is another physical quantity that distinguishes between the electrons and holes. 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. Using the usual geometry for the Hall effect measurements of a rectangular specimen, where the magnetic field $B_z$ is applied along the $z$ direction, the current density $j_x$ is fed along the $x$ direction and the Hall transverse electric field $E_y$ is generated along the $y$ direction, the Hall coefficient is defined as $R_H = E_y / j_x B_z$. In order to determine the anisotropy of $R_H$, three 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. The experimental uncertainty in $R_H$ was $\pm 0.1 \times 10^{-10}$ m$^2$ C$^{-1}$.

The anisotropic temperature-dependent Hall coefficient of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is shown in Fig. 7. The six $R_H$ sets of data form three groups of two approximately identical $R_H$ curves, where the magnetic field in a given crystallographic direction yields, in accordance with the Onsager relations, the same $R_H$ for the current along the other two crystallographic directions in the perpendicular plane. Thus, approximately identical Hall coefficients are obtained for the pair combinations $E_y / j_x B_z = E_y / j_y B_x = R_H^{ab}$ (where the additional superscript on the Hall coefficient denotes the direction of the magnetic field), $E_y / j_x B_z = E_y / j_y B_x = R_H^{ac}$ and $E_y / j_x B_z = E_y / j_z B_x = R_H^{bc}$. All Hall coefficients strongly increase upon cooling and their temperature-dependence resembles that of the Curie-type magnetic susceptibility $\chi$ within the paramagnetic phase, shown in Fig. 4(a). This indicates that the anomalous (magnetic) contribution to the Hall coefficient is dominant, to be discussed in the following.
In paramagnetic solids with the small-enough susceptibility that the demagnetization fields can be neglected (which is the case for our sample), the anisotropic Hall coefficient for the magnetic field along the \( k \) direction can be written as \(^{21}\)

\[
R_H^k = R_0^k + \chi_k R_S^k,
\]

where \( R_0^k \) is the normal Hall coefficient due to the Lorentz force and \( R_S^k \) is the anomalous Hall coefficient, originating from the magnetization. Due to the second term in Eq. (4), \( R_H^k \) follows the strong Curie-Weiss temperature dependence of \( \chi_k \). In order to separate the normal and anomalous Hall coefficients, \( R_H^k \) is plotted versus the susceptibility \( \chi_k \). Assuming temperature-independent \( R_0^k \) and \( R_S^k \) coefficients, \( R_H^k \) is derived from the intercept and \( R_S^k \) from the slope of the \( R_H^k(\chi_k) \) line. Using the paramagnetic susceptibility from Fig. 4(a), the \( R_H^k(\chi_k) \) plots for all combinations of the current and field directions are shown in Fig. 8(a). Although the \( R_H^k \) curves seem fairly linear in \( \chi_k \), a closer inspection reveals a clear deviation from linearity, demonstrating that the assumption of temperature-independent \( R_0^k \) and \( R_S^k \) is not justified. \( R_H^k \) should show strong temperature dependence only in the case of unequal and strongly temperature-dependent relaxation times of the electrons and holes, but this should also yield a strong temperature dependence of the electrical resistivity. For our T-Al\(_{72.5}\)Mn\(_{21.5}\)Fe\(_{6}\) system, the electrical resistivity increase between RT and 90 K (within the temperature range of the Hall-coefficient experiment) is by about 20\%, whereas the anisotropic Hall coefficients increase in the same temperature range for much larger factors of about 3.5. A significant contribution of \( R_0^k \) to the strong temperature dependence of \( R_H^k \) is thus unlikely and can be neglected with respect to that of \( R_S^k \), so that our further analysis is performed assuming a temperature-independent \( R_0^k \) and a temperature-dependent \( R_S^k \).

It is generally accepted\(^{22}\) that in metallic systems with a high resistivity, the anomalous Hall effect is dominated by a side-jump mechanism, i.e., by a lateral displacement that the electrons undergo during scattering in the presence of spin-orbit interaction.\(^{23}\) In that case, the anomalous Hall coefficient should be proportional to the square of the resistivity, \( R_S \propto \rho^2 \), so that a temperature-dependent \( \rho(T) \) results in a temperature-dependent \( R_S(T) \).\(^{24}\) In an anisotropic crystal, \( \rho \) is replaced by the product \( \rho_{ij} \), where \( i, j \) denote the two orthogonal crystallographic directions perpendicular to the magnetic field direction. By suitably normalizing to the RT \((T=295\,\text{K})\) values, denoted here as \( \rho_{0ij} \), Eq. (4) can be rewritten as

\[
R_H^k = R_0^k + R_S^k \kappa \chi_k \rho_{ij}/\rho_{0ij},
\]

where \( R_S^k \) is the value of the anomalous Hall coefficient \( R_S \) at 295 K. In Fig. 8(b) we plotted \( R_H^k \) as a function of \( \chi_k(\rho_{ij}/\rho_{0ij}) \). In this plot, the anisotropic \( R_H^k \) curves show well-defined linearity, enabling us to extract the \( R_0^k \) and \( R_S^k \) parameters. These parameter values, for all combinations of the current and field directions, are listed in Table II. The differences of the two \( R_0^k \) or \( R_S^k \) values for a particular field direction are small enough to be ascribed to the experimental error. The \( R_0^k \) values are positive for all three crystallographic directions and in the range \([0.5,0.8] \times 10^{-10}\,\text{m}^3\,\text{C}^{-1}\). The positive sign of the normal Hall coefficient indicates that holes are the dominant charge carriers.
in agreement with the positive hole-type thermoelectric power from Fig. 6. The above $R_0^b$ magnitude of the order $10^{-10}$ m$^2$ C$^{-1}$ yields the effective charge-carrier density of the order $10^{13}$ cm$^{-3}$, a typical metallic value. Within the experimental error, no anisotropy of the normal Hall coefficient $R_0$ can be claimed. The $R_0^b$ values are, on the other hand, of the order $10^{-7}$ m$^2$ C$^{-1}$ and show small, but distinct anisotropy. The highest value $R_0^b$ = $(3.1 \pm 0.1) \times 10^{-7}$ m$^2$ C$^{-1}$ is obtained for the field along the $b$ direction, where the in-plane product $\rho_p \rho_c$ is the largest according to the anisotropic resistivities shown in Fig. 5. This supports the consideration that the anisotropy of the resistivity is the dominant cause of the anisotropy of the anomalous Hall coefficient $R_0$. We emphasize that the $R_0^b$ values are rather large, which is certainly due to the high resistivity of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ compound that amounts at RT (depending on the crystallographic direction) between 440 and 500 $\mu$Ω cm. The obtained $R_0^b$ values are close to those determined for the T-Al$_{60}$Mn$_{30}$, T-Al$_{72.5}$Mn$_{21.5}$, and T-Al$_{73}$Mn$_{21}$ phases, amounting (3.2, 4.8, and 3.5) $\times 10^{-7}$ m$^2$ C$^{-1}$, respectively. For comparison, amorphous ferromagnetic alloys with lower resistivities around 150 $\mu$Ω cm show smaller $R_0$ of the order $10^{-8}$ m$^2$ C$^{-1}$ whereas for the high-resistivity icosahedral quasicrystal Al$_{70.4}$Pd$_{20.8}$Mn$_{8.8}$ with $\rho_{RT}$ $= 1000$ $\mu$Ω cm, a higher value $R_0$ = $1.8 \times 10^{-5}$ m$^2$ C$^{-1}$ was reported, showing clearly the influence of the resistivity magnitude on $R_0$.

A basic discrepancy between the Hall-coefficient measurements on the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ and the polygrain Taylor-phase samples studied in Ref. 15 is the sign of the normal Hall coefficient, which is positive holelike for the single crystal and negative electronelectron-like (or the coefficient value is close to zero for some chemical compositions) for the polygrain T-Al$_2$Mn, T-Al$_{17.3}$Mn$_{27−x}$Fe$_x$ ($x \approx 2.4$) and $d$-Al$_{17.3}$Mn$_{27}$Fe$_x$. Since the positive holelike normal Hall coefficient of the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is in agreement with the positive holelike thermopower of this compound, the results of the single crystal should be trusted.

To summarize, the Hall coefficient of the single-crystalline T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is dominated by the magnetic anomalous contribution and shows strong temperature dependence. For all three directions of the magnetic field $B_z$, the Hall coefficient $R(T)$ is a linear function of $\chi_p\rho_c/\rho_p$. This enables the separation of the normal and anomalous Hall coefficients $R_0$ and $R_5$. The values of the normal Hall coefficient $R_0$ are typical metallic of about $(0.7 \pm 0.1) \times 10^{-10}$ m$^2$ C$^{-1}$ and do not exhibit anisotropy within the experimental error. The positive sign of $R_0$ indicates that holes are the majority charge carriers, in agreement with the hole-type positive thermopower of this compound. The anomalous Hall coefficient $R_5$ is very large due to the high resistivity of the compound, with the RT value of the order $10^{-7}$ m$^2$ C$^{-1}$. It exhibits a weak anisotropy that can be attributed to the weak anisotropy of the electrical resistivity.

**E. Thermal conductivity**

Thermal conductivity $\kappa$ of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ single crystal was measured along the $a$, $b$, and $c$ directions using an absolute steady-state heat-flow method. The thermal flux through the samples was 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 sample was monitored by a chromel-constantan differential thermocouple. The anisotropic total thermal conductivities $\kappa_a$, $\kappa_b$, and $\kappa_c$ of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ along the three crystallographic directions, normalized to their 300 K values, are displayed in Fig. 9. The normalization was employed to show that the temperature-dependence of $\kappa_a$, $\kappa_b$, and $\kappa_c$ are practically the same within the experimental error. The absolute values at $T$ = 300 K amount $\kappa_a$ = 3.7 W/mK, $\kappa_b$ = 3.2 W/mK, and $\kappa_c$ = 3.2 W/mK, so that the anisotropy of the thermal conductivity is practically negligible.

The electronic contribution $\kappa_a$ to the total thermal conductivity can be estimated from the Wiedemann-Franz (WF) law, $\kappa_a = \pi^2 k_B^2 T$ $\sigma(T)/e^2$, and the measured electrical conductivity data $\sigma(T)$. Here it is important to recall the validity of the WF law, which is valid under the condition of dominant elastic scattering of the electrons, usually realized at high temperatures $T > \theta_D$, where $\theta_D$ is the Debye temperature (e.g., for typical metals, the WF law is valid already at RT). At low temperatures, the WF law is valid for solids where only the residual electrical resistivity (due to elastic scattering by quenched defects) is observed. The strongly temperature-dependent NTC resistivity of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ single crystal from Fig. 5 suggests that the WF law in not applicable at temperatures below RT, so that we use it only to estimate the RT $\kappa_a$ values. Taking the 300 K $\rho$ values from Fig. 5, we obtain the electronic thermal conductivities at $T$ = 300 K as $\kappa_a^e$ = 1.5 W/mK, $\kappa_b^e$ = 1.6 W/mK, and $\kappa_c^e$ = 1.5 W/mK. This gives the RT ratios of the electronic to the total thermal conductivity as $(\kappa_a^e/\kappa_a)_{300}$ K = 0.41, $(\kappa_b^e/\kappa_b)_{300}$ K = 0.50, and $(\kappa_c^e/\kappa_c)_{300}$ K = 0.47, so that at RT, the electrons and holes carry about half of the heat, the other half being transported by the lattice.

**IV. SUMMARY AND CONCLUSIONS**

We have investigated the magnetic susceptibility, the electrical resistivity, the thermoelectric power, the Hall coefficient and the thermal conductivity of the Taylor-phase...
T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ complex intermetallic compound that is an orthorhombic approximant to the d-Al-Mn-Pd decagonal quasicrystal with six atomic layers within one periodic unit of 1.24 nm and 156 atoms in the giant unit cell. The main objective was to determine the crystallographic-direction-dependent anisotropy of the investigated physical parameters 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.

The T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ shows spin-glass behavior below the spin-freezing temperature $T_f \approx 29$ K. The orientation-dependent magnetic susceptibilities do not show anisotropy for the magnetic field applied along the two in-plane directions $a$ and $c$, whereas there exists a moderate anisotropy between the in-plane and the stacking $b$ directions, where the susceptibility along $b$ is somewhat higher. The temperature-dependent susceptibility difference $\chi_b - \chi_c$ below $T_f$ does not show anisotropy and there is also no anisotropy in the spin-freezing temperature, the $M(H)$ hysteresis curves and the memory effect for the three crystallographic directions.

The anisotropic electrical resistivities are rather large (the RT values in the range 440–500 $\mu\Omega$ cm) and show a NTC along all three crystallographic directions. The resistivity is the lowest along the stacking $b$ direction perpendicular to the atomic planes, whereas the two in-plane resistivities $\rho_a$ and $\rho_c$ are at RT about 10% higher and show almost no anisotropy. It is remarkable that this kind of anisotropy, where the resistivity is the lowest along the stacking direction, appears to be a general feature of the decagonal-approximant phases with the stacked-layer structure. The NTC resistivity was theoretically reproduced by the quantum transport theory of slow charge carriers, applicable to metallic alloys and compounds in which the electron mean-free path $l$ between the scattering events is small compared to the extension of the conduction-electron wave packet, in which the electron propagation is non-Boltzmann (nonballistic) and the resistivity exhibits a NTC. The short mean-free path in the Taylor-phase crystals is a consequence of structural and chemical disorder on the lattice, as most of the lattice sites show either fractional occupation or mixed TM/Al occupancy.

The thermopower is positive for all three crystallographic directions and no anisotropy can be claimed within the experimental precision. The almost isotropic thermopower indicates that the anisotropy of the Fermi surface of the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is small, whereas its positive sign indicates that the holelike parts of the Fermi surface are dominant.

The same conclusion on the holes being the dominant charge carriers in the T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ is inferred from the Hall-coefficient measurements. The anisotropic Hall coefficient is a sum of the normal Hall coefficient due to the Lorentz force and the anomalous term, originating from the magnetization. The anomalous term introduces strong temperature dependence into the Hall coefficient, originating from both the temperature-dependent magnetic susceptibility and the temperature-dependent electrical resistivity. The anomalous Hall coefficient exhibits a weak anisotropy that can be attributed to the weak anisotropy of the electrical resistivity. The normal Hall coefficient is temperature independent and positive, confirming that the holes are the dominant charge carriers. It shows no anisotropy for different combinations of the magnetic field and electric current directions.

The anisotropy of the thermal conductivity of T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ along the three crystallographic directions is practically negligible. An estimation of the electronic contribution to the total thermal conductivity by using the Wiedemann-Franz law suggests that at RT, the electrons and holes carry about half of the heat, the other half being transported by the lattice.

The anisotropy of the investigated magnetic, electrical and thermal transport properties of the Taylor-phase T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ decagonal approximant with six atomic layers within one periodic unit along the stacking direction is small or in some cases negligible within the experimental precision (the thermopower and the thermal conductivity). This result should be contrasted with the anisotropic properties of the decagonal approximants from the Al$_{13}$TM$_4$ family, where the Y-Al-Ni-Co two-layer compound$^{3,4}$ and the Al$_{13}$Co$_4$ (Ref. 5), Al$_{13}$Fe$_4$, and Al$_{13}$(Fe,Ni)$_4$ (Ref. 6) four-layer compounds all show significant anisotropies along the three orthogonal crystallographic directions. However, the Al$_4$(Cr,Fe) (Refs. 7 and 8) six-layer decagonal approximant from the Al$_4$TM family shows anisotropy of a similar magnitude to those observed for the above two- and four-layer Al$_{13}$TM$_4$ phases. The probable reason that the increasing number of atomic layers within one periodic unit decreases the anisotropy of the tensorial physical properties of a decagonal-approximant phase with a stacked-layer structure is thus not generally valid, but the degree of anisotropy rather depends on the structural and chemical details of a particular phase. Our investigation presents experimental evidence that the Taylor-phase six-layer decagonal approximant shows the smallest anisotropy of all the so-far investigated stacked-layer decagonal-approximant phases. The Taylor phase can thus be considered as a “close-to-isotropic” compound. The decisive role of the structural and chemical details of a particular phase for its physical properties is also supported by the fact that the properties of the Taylor phase T-Al$_{72.5}$Mn$_{21.5}$Fe$_{6.0}$ are in many respects very different from those of the Al$_{13}$TM$_4$ and Al$_4$TM families. Although all these phases contain a significant fraction of a magnetic transition metal (Mn, Fe, Co), the Taylor phase is the only one to show strong Curie-type paramagnetism and a spin glass transition at low temperatures. The NTC resistivity of the Taylor phase is also different from the PTC resistivity of the Al$_{13}$TM$_4$ phases, whereas the anisotropic resistivities of the Al$_4$(Cr,Fe) exhibit a maximum with the transition from NTC to PTC upon cooling. The Taylor phase is also the only one to show the dominant anomalous contribution to the Hall coefficient. Therefore, no common footing of the anisotropic
physical properties of the stacked-layer decagonal-approximant phases from different structural families can be inferred.

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