Low-temperature thermal conductivity of icosahedral $Al_{70}Mn_9Pd_{21}$

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We report measurements of the thermal conductivity $\lambda(T)$ of icosahedral $Al_{70}Mn_9Pd_{21}$ in the temperature range 0.06–110 K. The quasilattice thermal conductivity λ_{ph} increases monotonically with T in the temperature range between 0.06 and 25 K and above 55 K. A plateau is observed for $\lambda_{ph}(T)$ between 25 and 55 K, i.e., at substantially higher temperatures than in amorphous solids. Moreover, the thermal conductivity of icosahedral $Al_{70}Mn_9Pd_{21}$ in the λ plateau region is *substantially higher* than that of insulating and metallic glasses. At lower temperatures, between 0.35 and 1.6 K, λ_{ph} varies as $T^{2.06}$. This $\lambda_{ph}(T)$ variation is compatible with a scattering of phonons by tunneling states. Below 0.35 K the thermal conductivity decreases with increasing slope suggestive of a crossover to a regime of excessive phonon scattering characterized by a temperature-independent mean free path.

I. INTRODUCTION

Since the discovery of icosahedral structural symmetry in rapidly quenched Al-Mn alloys[1] the consequences of quasiperiodicity for the physical properties of quasicrystals and possibly related peculiarities of their electronic structure have been studied intensively. It was, however, soon realized that Al-Mn and other metastable quasicrystals possess a high degree of intrinsic disorder that inhibited unambiguous investigations and evaluations of anticipated novel physical properties.

New opportunities for experimental studies of physical properties of condensed matter with quasiperiodic structures are provided by the recent discovery of the thermodynamically stable quasicrystals Al-Cu-(Fe,Ru,Os),[2] which have face-centered icosahedrally ordered structures and a long-range quasiperiodic order with structural coherence lengths up to 10^4 Å compatible with those of well-ordered periodic crystals.[3] Recently, a new stable icosahedral phase was discovered in the Al-Mn-Pd system,[4,5] that does not show phason broadening of Bragg peaks even in the nonannealed state. Structural analysis indicates that this phase forms an ordered icosahedral state[6–8] and therefore it can be considered as an ideal model system for the investigation of its possible nontrivial physical properties.

Most of the studies performed so far on this material were related to its structural characteristics.[6–8] The available information on physical properties include data of electrical transport properties[9,10] supplemented by the complete electrodynamic response,[11] low-temperature thermodynamic and magnetic properties,[12] results from photoemission experiments,[13] and acoustical properties.[14]

Studies of low-temperature heat transport are, to our knowledge, available for metastable phases only. The low-temperature thermal conductivity of both icosahedral and glassy Pd-U-Si alloys was investigated in Ref. 15. The reported results for the two different phases are similar in their temperature dependence, with the icosahedral phase exhibiting an approximately 30% lower thermal conductivity. This is not surprising because icosahedral Pd-U-Si phases are metastable and notorious for a high degree of intrinsic disorder.

We are not aware of investigations of the low-temperature heat transport in thermodynamically stable icosahedral quasicrystals. Below we present the results of measurements of the thermal conductivity of a bulk sample of icosahedral $Al_{70}Mn_9Pd_{21}$ with high structural perfection.

II. EXPERIMENT

The sample of icosahedral $Al_{70}Mn_9Pd_{21}$ was synthesized from 99.997% pure aluminum, 99.9% pure palladium, and 99.94% pure manganese by arc-melting suitable amounts of the constituents to a single piece and remelting it several times to provide homogeneity. The resulting button was annealed for 2 days at 800 °C, and subsequently quenched into water directly from 800 °C. The sample composition is optimal for the formation of the thermodynamically stable icosahedral phase in the Al-Mn-Pd system. The absence of any inclusions of other phases was confirmed by surface analysis from backscattered electron images. Also, selected-area electron-diffraction patterns show a high degree of order and a low density of phason defects. More details concerning the sample preparation, the structural analysis, and its characterization are given elsewhere.[12]

A specimen in the form of a prism with dimensions $1.07\times1.22\times5.80$ mm^3 was cut from the ingot by electroerosion. The thermal conductivity was measured by a standard steady-state heat-flow technique monitoring the temperature gradient along the sample, using a dilution refrigerator from 0.06 to 1.1 K and a pumped ^{4}He cryostat between 1.5 and 110 K.

III. RESULTS, ANALYSIS, AND DISCUSSION

In Fig. 1 the measured total thermal conductivity λ_{tot} of icosahedral $Al_{70}Mn_9Pd_{21}$ is shown on logarithmic scales for the whole temperature range covered in this work. The thermal conductivity λ_{ph} contributed by the

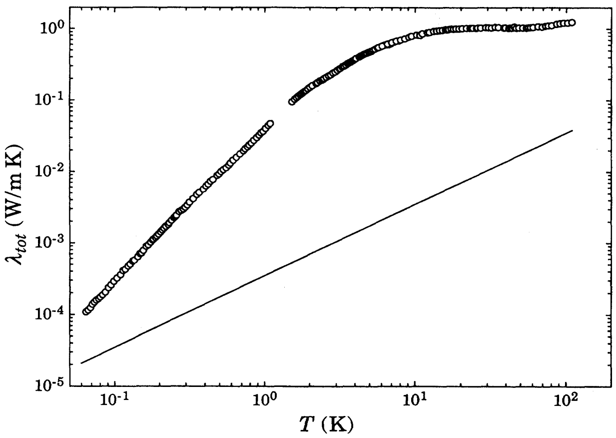

FIG. 1. Measured total thermal conductivity λ_{tot} of icosahedral $Al_{70}Mn_9Pd_{21}$ as a function of temperature between 0.06 and 110 K. The solid line is an estimate of the electronic contribution λ_{el}, as explained in the text.

phonons alone may be obtained by subtracting off an estimate of the electronic contribution λ_{el}. The latter was calculated using the Wiedemann-Franz law and the previously measured electrical conductivity presented in Ref. 10 and obtained from a sample cut from the same ingot. In Fig. 1, the result of this calculation is shown as a solid line. It may be seen that over most of the covered temperature range λ_{el} is at least an order of magnitude less than λ_{tot} and therefore λ_{ph} may be evaluated quite accurately there.

Figure 2 shows the quasilattice thermal conductivity λ_{ph} of icosahedral $Al_{70}Mn_9Pd_{21}$ as established in the way described above. As a first observation we note that λ_{ph} increases monotonically with T in the temperature range between 0.06 and 25 K. A strong tendency to saturation of $\lambda_{ph}(T)$ is evident above approximately 10 K. From 25 to 55 K, λ_{ph} is almost temperature independent and it slowly increases with T above 55 K. This overall

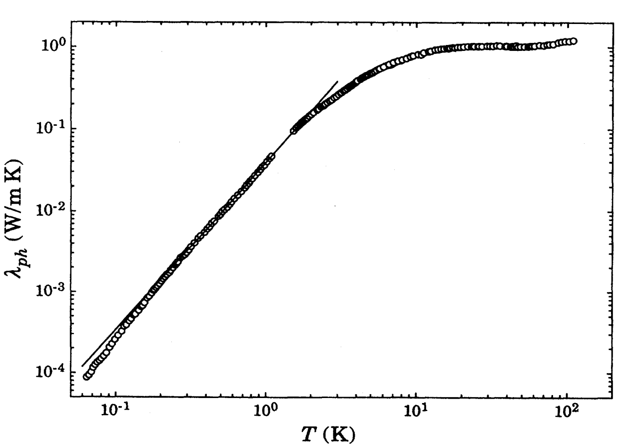

FIG. 2. Quasilattice thermal conductivity λ_{ph} as a function of temperature T. The solid line is a power-law approximation to the data between 0.35 and 1.6 K.

behavior is similar to that of amorphous solids, where generally a λ plateau is observed in a temperature range between approximately 2 and 10 K.

A. Low temperatures $0.06\ K < T < 1.6\ K$

We now analyze the low-temperature behavior of λ_{ph}. At the lowest temperatures between 0.06 and 0.1 K the temperature variation of λ_{ph} is, strictly speaking, characterized by a temperature-dependent slope of the $\lambda_{ph}(T)$ curve but may adequately be approximated by $\lambda_{ph} = AT^n$, where $n = 2.46 \pm 0.08$. Between 0.1 and 0.35 K a distinct change of slope is revealed and between 0.35 and 1.6 K the slope is fairly constant, corresponding to an exponent $n = 2.06 \pm 0.01$ evaluated from the slope of the solid line shown in Fig. 2.

We now focus on the $\lambda_{ph}(T)$ variation in the temperature range from 0.35 to 1.6 K. The power-law behavior of $\lambda_{ph}(T)$ with an exponent close to 2 in this temperature range is similar to what is observed in amorphous solids and compatible with the scattering of phonons by tunneling states.[16] We note that in such cases the actual exponent in the power-law description of $\lambda_{ph}(T)$ is almost never exactly 2. This is thought to arise from a weak energy dependence of the density of tunneling states acting as scattering centers.

The existence of tunneling states in Al-Mn-Pd quasicrystals was recently claimed by Vernier and coworkers[14] by analyzing the results of ultrasound experiments. They revealed a small logarithmic deviation in the sound-velocity variation with temperature and a nonlinear attenuation of acoustic shear waves at low temperatures. These are interpreted as characteristic signatures of the tunneling states.[17]

To test the assumption that phonon scattering by tunneling states dominates the thermal conductivity of icosahedral $Al_{70}Mn_9Pd_{21}$ between 0.35 and 1.6 K, we analyze our data employing the formalism of the tunneling-state model.[16] Assuming an energy-independent density of tunneling states and employing the Debye approximation, the thermal conductivity in this model is given by

$$\lambda_{TS} = \frac{\rho k_B^3 \bar{v}}{6\pi\hbar^2 \bar{P}\gamma^2} T^2 , \tag{1}$$

where ρ is the density, $\bar{v}$ is the average of the sound velocities v_t and v_l for transverse and longitudinal waves, respectively, $\bar{P}$ is the density of tunneling states, and γ describes the average coupling between them and the phonons. A fit of our $\lambda_{ph}(T)$ data to Eq. (1) in the temperature range between 0.35 and 1.6 K yields the value of $6.5\times10^7\ \mathrm{erg\,cm^{-3}}$ for the coupling parameter $\bar{P}\gamma^2$, fairly close to the $\bar{P}\gamma^2$ values previously deduced from the thermal-conductivity data of typical insulating and metallic glasses.[18] Here we used $v_t = 3.8\times10^5$ cm s and $\rho = 5.1\ \mathrm{g\,cm^{-3}}$ from Ref. 14 and we assumed that v_t is close to the sound velocity of longitudinal waves v_l.

Within the framework of the tunneling-state model, independent values of $\bar{P}\gamma^2$ may be obtained from acoustic experiments. Values of $1.6\times10^7\ \mathrm{erg\,cm^{-3}}$ and $1.3\times10^7\ \mathrm{erg\,cm^{-3}}$ for the coupling parameter $\bar{P}\gamma^2$ of an Al-Mn-Pd quasicrystal with slightly different composition than that of our sample were deduced from the sound-velocity variation with temperature and the nonlinear acoustic attenuation of shear waves, respectively.[14] These values are obviously very close to $\bar{P}\gamma^2 = 1.9\times 10^7$ erg cm^{-3} obtained from sound-velocity measurements on the metallic glass Pd-Si.[19] The fact that the coupling constant obtained from sound-velocity experiments is several times lower than the $\bar{P}\gamma^2$ value compatible with the measured thermal conductivity is well known in metallic glasses.[20] It was argued that the presence of conduction electrons in glassy metals may dramatically decrease the relaxation times of tunneling states and, therefore, change the average coupling parameter $\bar{P}\gamma^2$.[21]

Tunneling states in quasicrystals have been associated with phasons.[22] A phason disorder in the atomic occupation of specific sites in a structurally perfect Al-Mn-Pd quasicrystal was recently reported by de Boissieu and coworkers[8] by analyzing the results of anomalous x-ray-diffraction experiments close to the Pd edge. It is difficult to come to any definite conclusions as to whether or not this kind of disorder is connected with tunneling states, in particular because the location of atoms is not known in detail.[7,8] The microscopic origin of tunneling states in amorphous solids is not known either, although the existence of these states is unambiguously established by various independent experiments. They are assumed to be related to a tunneling motion of a group of atoms and it is generally accepted that a continuous density of tunneling states results from some kind of randomness of their structure. It was argued that a density of tunneling states in quasicrystals is discrete and that an energy splitting of these states is constant because the quasicrystalline state is an ordered state.[23] Our $\lambda_{ph}(T)$ data presented here and the results of ultrasound experiments reported in Ref. 14 obviously imply a broad spectrum in the energy splitting of tunneling states and therefore they do not support such an argument, at least for Al-Mn-Pd quasicrystals. We note that a continuous density of tunneling states is characteristic even of periodic crystals. An example is a crystalline $Ti_{0.65}V_{0.33}$ alloy with bcc structure.[24] The low-temperature phonon thermal conductivity of this solid-solution alloy varies as T^2 and the values of λ_{ph} are of the same order of magnitude as those reported for insulating and metallic amorphous solids. This was ascribed to the high degeneracy in this material of energetically similar configurations of positional disorder.[24]

A more comprehensive attempt to interpret our $\lambda_{ph}(T)$ data below 0.35 K is demonstrated in Fig. 3 where we plot λ_{ph}/T^3 vs T. In this figure we show the solid straight line representing a fit of Eq. (1) to our $\lambda_{ph}(T)$ data in the temperature range between 0.35 and 1.6 K, although we realize that an exponent $n>2$ provides a better approximation (see above). The diagram reveals that below 0.35 K, λ_{ph}/T^3 drops below the T^{-1} variation with decreasing T, indicative of a stronger than T^2 variation of $\lambda_{ph}(T)$. We note that the distinct deviation from an approximate T^2 behavior cannot be ascribed to a systematic error since even at 0.06 K the calculated electronic thermal conductivity λ_{el} reaches only 20% of λ_{tot}.

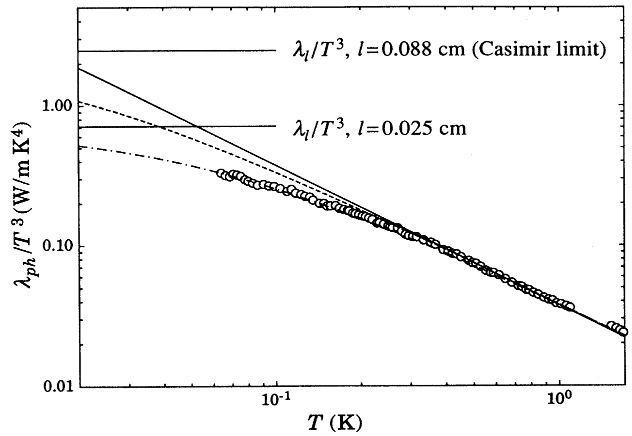

FIG. 3. λ_{ph}/T^3 vs T of icosahedral $Al_{70}Mn_9Pd_{21}$ below 1.6 K. The different lines are different fits, as explained in the text.

This trend suggests that an additional mechanism of phonon scattering becomes effective at low temperatures which limits the phonon mean free path to below that imposed by scattering involving tunneling states. The resulting total mean free path becomes less frequency dependent and it varies approximately at $T^{-0.5}$ at the lowest temperatures of our experiment.

Similar λ_{ph} vs T behavior as shown in Fig. 3 for icosahedral $Al_{70}Mn_9Pd_{21}$, i.e., that the T dependence of λ_{ph} is stronger than T^2 at very low temperatures, is particularly pronounced in composite glassy systems with an additional source of phonon scattering. Examples are fused capillary arrays of borosilicate glass or policarbonate[25] and boroaluminosilicate glass containing mica crystallites.[26] There are some other differences in the behavior of the above-mentioned materials that are not important at this point, however. Introduction of holes or crystallites in a glassy matrix produces a frequency-independent phonon mean free path. This results in a strong suppression of the phonon mean free path below the value determined by the phonon scattering on tunneling states alone. Therefore the thermal conductivity of composite glassy systems also shows a gradual increase of slope in $\lambda_{ph}(T)$ with decreasing temperature. At the lowest temperatures the $\lambda_{ph}(T)$ variation is reported to be close to T^3.[25]

The above-mentioned composite glassy systems represent cases of extremely strong additional phonon scattering corresponding to mean free paths of the order of 10^4–10^{-3} cm. If the mean free path l due to excessive scattering is long enough the T^3 regime will not be reached down to the lowest temperatures usually accessible to thermal-conductivity measurements of the order of 0.05 K.

Below we argue that the behavior of λ_{ph} of icosahedral $Al_{70}Mn_9Pd_{21}$ between 0.06 and 0.35 K most likely manifests a crossover between a regime, where the phonon mean free path is limited by scattering involving tunneling states and varying as T^{-1} and a regime of excessive phonon scattering with a temperature-independent mean free path. The total thermal conductivity that includes scattering of the heat carriers by two-level systems dominating the behavior at higher temperatures, and the excessive scattering characterized by a temperature-independent mean free path at lower temperatures may be obtained by applying a phonon equivalent of Matthiessen's rule:

$$\lambda=(\lambda_l^{-1}+\lambda_{\mathrm{TS}}^{-1})^{-1} , \tag{2}$$

where $\lambda_{\mathrm{TS}}^{-1}$ is the thermal resistance due to phonon scattering involving tunneling states and given by Eq. (1), and λ_l is the thermal conductivity limited by the temperature-independent mean free path l (Ref. 27):

$$\lambda_l=\frac{2\pi^2 k_B^4 l T^3}{15\hbar^3\bar{v}^2} , \tag{3}$$

where $\bar{v}$ is again an appropriate average of the sound velocities for transverse and longitudinal waves.

In a first step we check the possibility that the deviation of $\lambda_{\mathrm{ph}}(T)$ from the approximate T^2 behavior can be explained by the growing importance of a boundary-limited conductivity in the Casimir limit, assuming nonspecular scattering of phonons at the sample surface. Size-limited thermal conductivities are well documented for pure periodic crystals.[28,29] On the contrary, nonspecular boundary scattering is usually absent at surfaces of glassy materials. It was argued that the defects responsible for diffusive scattering of phonons at the surfaces of periodic crystals are not present in amorphous solids.[30] It is not known whether the necessary kind of defects exist at surfaces of quasicrystals.

The calculated $\lambda_{\mathrm{ph}}/T^3$ vs T variation given by Eq. (2) with the sample-boundary-limited mean free path $l=\alpha(4A/\pi)^{1/2}=0.088$ cm, where A is the cross-sectional area of the sample and the factor α accounts for the finite sample length[31] is plotted as a dashed line in Fig. 3. A slightly reduced value of $\bar{P}\gamma^2=6.1\times10^7$ erg cm^{-3} is needed to fit the data above 0.35 K. Below 0.35 K this line strongly deviates from the experimentally measured $\lambda_{\mathrm{ph}}/T^3$ values. This indicates that the effective mean free path for phonons at very low temperatures is substantially shorter than 0.088 cm and, therefore, we cannot attribute the observed deviation of $\lambda_{\mathrm{ph}}/T^3$ from the T^{-1} variation below 0.35 K to the proximity of the Casimir limit, shown as the upper horizontal solid line in Fig. 3. The possibility of a boundary-limited conductivity with specular scattering can also be discarded because it implies an even higher thermal conductivity.

If we now consider the mean free path l as a fitting parameter we may obtain its value from the fit of Eq. (2) to our $\lambda_{\mathrm{ph}}(T)$ data in the temperature range from 0.06 to 1.6 K. The result of this fit, displayed as the dot-dashed line in Fig. 3, yields the length of $l=0.025$ cm. It is not clear which mechanism of phonon scattering is responsible for this length scale. Scattering of phonons on microholes or grain boundaries may limit the phonon mean free path to the above-mentioned length of 0.025 cm. We note that different types of faceted microholes with an average diameter between 20 and 25 μm have been observed in Al-Mn-Pd quasicrystals.[6] The limiting $\lambda_{\mathrm{ph}}/T^3$ value compatible with this mean free path is shown as the lower horizontal solid line in Fig. 3.

B. Temperature range 1.6 K < T < 110 K

Finally we comment on the $\lambda_{\mathrm{ph}}(T)$ behavior between 1.6 and 110 K and compare it to that of amorphous solids. Above we described the main features of $\lambda_{\mathrm{ph}}(T)$ of icosahedral $Al_{70}Mn_9Pd_{21}$ (see Fig. 2). The main feature at higher temperatures is the approximate temperature independence of λ_{ph} in the temperature range between 25 and 55 K. This behavior is reminiscent of the λ plateau characteristic of amorphous solids. We note important differences regarding the thermal-conductivity behavior of icosahedral $Al_{70}Mn_9Pd_{21}$ and that of amorphous solids in the λ-plateau regions, however. The λ plateau of icosahedral $Al_{70}Mn_9Pd_{21}$ is developed at substantially higher temperatures than in amorphous solids, where generally a λ plateau occurs in a temperature range between approximately 2 and 10 K. Moreover, the thermal conductivity of icosahedral $Al_{70}Mn_9Pd_{21}$ in the temperature range where the λ plateau is observed is *substantially higher* than that of insulating and metallic glasses in their respective plateau regions. For example, it exceeds the corresponding value for amorphous SiO_2 by an order of magnitude and that of amorphous Pd-Si by a factor 2 (see Fig. 4).

It was shown by Freeman and Anderson[32] that if the ratio λ_{ph}/K, where

$$K=\frac{k_B^3}{\pi\hbar^2}\frac{\Theta_D^2}{v_s} , \tag{4}$$

with Θ_D as the Debye temperature and $v_s^{-3}=(v_l^{-3}+2v_t^{-3})/3$, is plotted vs T/Θ_D, then the thermal-conductivity data for different amorphous materials can be brought into a common register both at low and high temperatures. We have scaled our thermal-conductivity data using the value of $\Theta_D=362$ K for the Debye temperature obtained from low-temperature specific heat data, measured on a sample cut from the same ingot of icosahedral $Al_{70}Mn_9Pd_{21}$.[12] We found that in the temperature range $T<10^{-2}\Theta_D$ the λ_{ph}/K values are fairly close

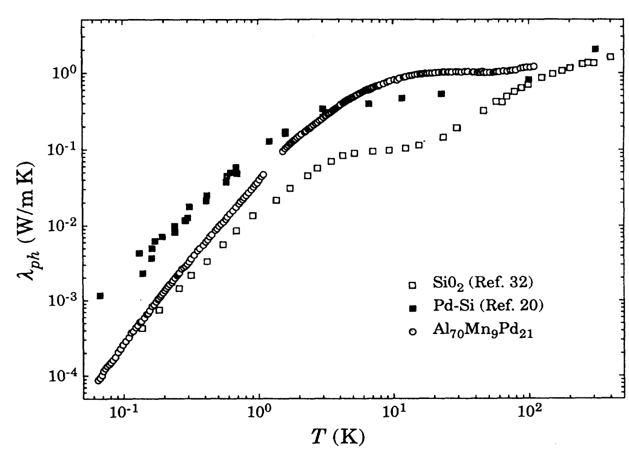

FIG. 4. Temperature dependence of the quasilattice thermal conductivity λ_{ph} of icosahedral $Al_{70}Mn_9Pd_{21}$ in comparison with analogous data for two amorphous materials (insulating and metallic) taken from Refs. 20 and 32.

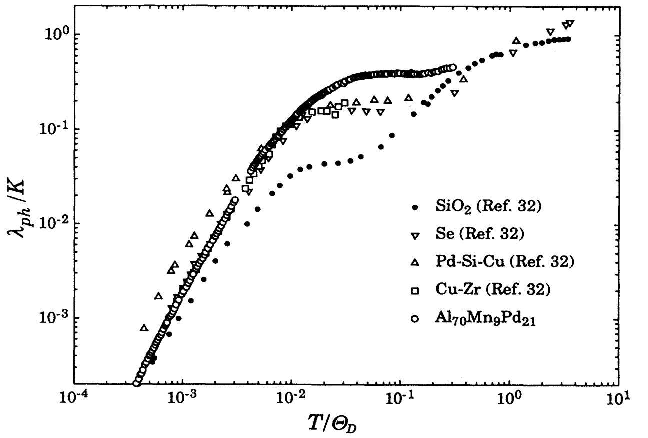

FIG. 5. Scaled quasilattice thermal conductivity λ_{ph}/K of icosahedral $Al_{70}Mn_9Pd_{21}$ in comparison with four insulating and metallic glasses (from Ref. 32), versus T/Θ_D.

to those of most amorphous solids (Fig. 5). In the intermediate temperature range $10^{-2}\Theta_D < T < 10^{-2}\Theta_D$ the thus scaled thermal conductivity λ_{ph}/K exceeds the values typical for amorphous solids. At higher temperatures $T > 10^{-1}\Theta_D$, λ_{ph}/K of quasicrystalline $Al_{70}Mn_9Pd_{21}$ again tends to approach the values typical for amorphous solids.

IV. CONCLUSIONS

The thermal conductivity of icosahedral $Al_{70}Mn_9Pd_{21}$ has been measured in the temperature range between 0.06 and 110 K. Our data imply both common features but also important differences regarding the physical mechanisms dominating the low-temperature thermal conductivity of either quasicrystalline $Al_{70}Mn_9Pd_{21}$ or amorphous solids.

The quasilattice thermal conductivity λ_{ph} of icosahedral $Al_{70}Mn_9Pd_{21}$ increases monotonically with T in the temperature range between 0.06 and 25 K. From 25 to 55 K λ_{ph} is almost temperature independent and it slowly increases with T above 55 K. The λ plateau of icosahedral $Al_{70}Mn_9Pd_{21}$ is developed at substantially higher temperatures than in amorphous solids. This result suggests that a crossover to a strong decrease of the phonon mean free path with increasing frequency takes place at higher phonon frequencies than in amorphous solids.

Below 1.6 K the temperature variation of λ_{ph} is well described by $\lambda_{ph}(T) \sim T^{2.06}$. This $\lambda_{ph}(T)$ variation is compatible with a dominant scattering of phonons by tunneling states and our values of λ_{ph} are of the same order of magnitude as those reported for insulating and metallic amorphous solids. With decreasing temperature, distinct deviations from this behavior become apparent below 0.35 K. They are compatible with a crossover between a regime in which the phonon mean free path is predominantly limited by scattering on the tunneling states and a regime which is characterized by a temperature-independent mean free path. This mean free path is substantially shorter than the Casimir length and is determined by a phonon scattering in the bulk of the sample, presumably by microholes.

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