Transport properties of icosahedral quasicrystal Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$

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Abstract

We present measurements of the thermoelectrical properties of polygrain icosahedral quasicrystal Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$ in the temperature range 10–300 K. Electrical resistivity has a relatively low room temperature value, $\rho_{R.T.} = 1.2$ m$\Omega$ cm, by lowering the temperature it shows an increase with a maximum value at 120 K. Thermoelectric power is positive in the whole temperature ranges with a quite high room temperature value, $S_{R.T.} = 70$ mV/K. Thermal conductivity shows a maximum at 30 K, and a broad minimum around 125 K. The room temperature value is $K_{R.T.} = 3.4$ W/mK. The appearance of the maximum in thermal conductivity at 30 K comes mainly from structural scattering, while at temperatures above 120 K from the contribution of the localized phonon state.

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Since the experimental discovery of quasicrystals [1], they are permanently attracting attention of researchers. Because of the lack of translation symmetry [2], quasicrystals possess unusual electrical [3] and thermal [4–8] properties. It is experimentally established that the main contribution to the thermal conductivity comes from the lattice contribution. The way in which phonons are influenced by the quasiperiodic arrangement of atoms depends on the phonon itself. In spite of lattice non-periodicity, long-wave phonons can be well defined. Moreover, inelastic neutron scattering experiments conducted on single-grain i-Al–Pd–Mn quasicrystals [9] showed the existence of well-defined acoustic phonons with wave vectors smaller then 0.3 Å. Since this wave vector corresponds to a temperature of several tens of Kelvins, the Debye approach can be used below these temperatures. On the high energy side of the phonon spectrum, Janot et al. [10] have observed localized vibration states in i-Al–Cu–Fe quasicrystals. Consequently, just a small number of phonons have heat-carrying capabilities, which along with low electrical conduction, explains why quasicrystals have low values of thermal conductivity. Thermal conductivity of quasicrystals, in general, can be divided into three temperature regions. It is accepted that in the low-temperature region ($T < 1$ K) the main source of the phonon scattering are tunneling states (TLS) and boundary scattering [4,6,11]. Existence of tunneling states in quasicrystals is confirmed by acoustic measurements [12–14], although it is not clear whether or not tunneling states are intrinsic characteristics of quasicrystals [13]. Moreover, recent measurements of thermal conductivity of single-grain i-Y$_{6.6}$Mg$_{34.6}$Zn$_{56.8}$ have shown that in the low-temperature region one does not need tunneling states to interpret the low-temperature thermal conductivity data [8]. At temperatures between 10 and 100 K the existence of a plateau [4,5] and a shallow maximum [6–8] have been observed. Above 100 K the increase in the value of thermal conductivity is common to all quasicrystals.

The Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$ crystal was prepared using the ‘self-flux’ technique, where the ternary melt is first slowly cooled, and then the remaining melt is decanted. The ‘self-flux’ technique does not require large temperature gradients, so samples prepared by this technique have less strain than samples prepared by the Bridgman or Czochralski techniques. In addition, quasicrystals grow via a series of stable thermodynamic steps. Melt decanting ensures that samples are single-phased [15]. The sample, in the shape of a rectangular prism with dimensions $0.5 \times 0.5 \times 5$ mm$^3$, was cut and polished from an as-grown
sample. Electrical conductivity was measured by the standard four contact method. Golden wires of 10 μm diameter were used as contact leads. The comparative steady state method with constantan foil as a reference sample for measuring thermal conductivity. Temperature gradients across both samples are determined by 15-μm chromel–constantan differential thermocouples.

Fig. 1 displays the electrical resistivity of the i-Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$. The room temperature value is 1.2 mΩ cm, and there is in the range of room temperature values observed for other quasicrystals of the i-Al–Pd–Mn family [16]. The temperature behaviour presented in Fig. 1 is not typical for quasicrystals. Anomalous behaviour of electrical resistivity, like that presented in Fig. 1, is observed in periodic structures of noble metals with diluted impurities of transition metals, and has been successfully explained by the theory given by Korringa and Gerritsen [17]. Recently the original KG theory was applied for the explanation of the observed maximum in the electrical resistivity curve of i-Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$ [18]. This curve roughly follows the temperature behaviour of the measured electrical resistivity of i-Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$, but detailed features of our experimental data are not explained by it. The applicability of the KG theory to our case can be justified by the fact that just a small part of the Mn atoms in the i-Al–Pd–Mn quasicrystals are magnetic [12] (between 1 and 1.4%). Thermopower data are presented in Fig. 2. Thermopower is positive in the whole investigated temperature range, and has a relatively large room temperature value S(300 K)≈70 μV/K. Contrary to the case of electrical resistivity, the KG theory could not explain the temperature behaviour of thermopower of i-Al$_{72}$Pd$_{19.5}$Mn$_{8.5}$ as one would expect.

The measured thermal conductivity is presented in Fig. 3. The room temperature value is $K_{\text{R.T.}}=3.4$ W/mK. The
position of the minimum of the thermal conductivity curve is at 120 K, which is higher than in the case of i-Al$_{70}$Pd$_{20}$Mn$_{10}$ [7] and i-Y$_{8.6}$Mg$_{34.6}$Zn$_{56.8}$ [8]. The electron contribution to thermal conduction is estimated by using the Wiedemann–Franz law $K_{\text{el}} = L_0 \alpha T$ (presented by the line in Fig. 3), where $L_0$ is equal to $2.45 \times 10^{-8} \text{ W K}^{-2}$. The electronic contribution to thermal conductivity, $K_{\text{el}}$, can be deduced by subtracting the electron contribution from the total thermal conductivity, $K_{\text{el}}$. The low temperature data ($T < 50$ K) can be well fitted by using the Debye expression [19]. The fitting procedure allowed us to determine $N_e$, a linear density of stacking faults. Using the value of the Grüneisen constant $\gamma$ and the quasilattice constant $a$, the linear density of stacking faults is $1.4 \times 10^6 \text{ 1/m}$. This is an order of magnitude smaller than in other Al–Pd–Mn icosahedral quasicrystals [20], and of the same order as in i-Al–Cu–Fe or i-Y–Mg–Zn.

The increase of thermal conductivity above approximately 100 K is a common property of all quasicrystals. The model proposed by Janot [21] takes into account the high energy and the localized vibration modes, experimentally observed in quasicrystals by neutron diffraction [10]. In that model the interaction between localized quasilattice vibrations and extended ones enables hopping of non-extended phonons. Taking into account structure inflation symmetry of an ideal icosahedral quasicrystal, the temperature behaviour of the thermal conductivity of localised phonon states assumes the power law, i.e., $K_{\text{loc}} \sim T^n$, with $n = 1.5$. If one subtracts the Debye contribution from the quasilattice thermal conductivity for temperatures above 200 K, the thermal conductivity data can really be fitted to the power law $K_{\text{loc}} \sim BT^n$, with $n = 1.7$, which is close to the prediction of Janot’s model. The fact that the position of the minimum of the thermal conductivity curve in our sample is relatively high compared to other icosahedral quasicrystals means a stronger localisation of the high energy quasilattice vibrations.

In conclusion, the measurements of electrical resistivity, thermopower and thermal conductivity of icosahedral quasicrystal i-Al$_{70}$Pd$_{20}$Mn$_{10}$ are presented. Electrical resistivity has an atypical temperature behaviour for quasicrystals which has been observed in periodic crystals with diluted magnetic alloys. The thermal conductivity curve shows temperature behaviour already seen in quasicrystals: it has a shallow maximum at 30 K and a local minimum at 120 K. Below 50 K the thermal conductivity can be fitted by the Debye expression which takes into account the stacking-like faults like structural scattering mechanisms. At higher temperatures the model of variable range hopping of localised phonon states adequately explains the observed rise of thermal conductivity.

References