UDK 669:538.945:536
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
MTAEC9, 44(1)3(2010)

THE THERMAL CONDUCTIVITY OF $Al_{73}Mn_{27-x}Fe_x$ TAYLOR PHASES

TOPLOTNA PREVODNOST TAYLORJEVIH FAZ $Al_{73}Mn_{27-x}Fe_x$

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Prejem rokopisa – received: 2009-07-21; sprejem za objavo – accepted for publication: 2009-08-24

The thermal conductivity (κ) of $Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6) complex metallic alloys has been measured in the temperature interval from 2 K to 300 K. All the alloys are Taylor (T) phases, except $Al_{73}Mn_{21}Fe_6$, which is a decagonal (d) quasicrystal. The behaviours of κ are typical for complex metallic alloys, i.e., a relatively small magnitude, a change of slope at about 50 K and an increase of the conductivity above 100 K. At room temperature the magnitude of κ for all the samples is between 2.7 W/mK and 3.3 W/mK, which is comparable to that of thermally insulating amorphous SiO_2 and Zr/YO_2 ceramics. The reason for such a low thermal conductivity is because both, the electronic and lattice conductivity are low. The electronic contribution to the thermal conductivity is low because of the large electrical resistivity of the samples. The lattice thermal conductivity is greatly reduced because of the enhanced umklapp process of the phonon scattering (caused by the large lattice constant) and by the disorder in the structure.

Keywords: complex metallic alloys, thermal conductivity, spectral conductivity

Toplotna prevodnost (κ) kompleksnih kovinskih zlitin $Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6) je bila izmerjena v razponu temperatur od 2 K do 300 K. Vse zlitine, z izjemo $Al_{73}Mn_{21}Fe_6$, ki je dekagonalni (d) kvazikristal, so Taylorjeve (T) faze. Vedenje toplotne prevodnosti κ je značilno za kompleksne kovinske zlitine: relativno majhna velikost, sprememba naklona pri približno 50 K in povečana prevodnost nad 100 K. Pri sobni temperaturi je velikost κ za vse preizkušance med 2,7 in 3,3 W/mK, kar je primerljivo s toplotno izolirno amorfno SiO_2- in Zr/YO_2-keramiko. Vzrok za tako majhno toplotno prevodnost sta majhni elektronska in mrežna prevodnost. Elektronski prispevek k toplotni prevodnosti je majhen zaradi velike električne upornosti zlitin. Mrežna toplotna prevodnost je zmanjšana zaradi povečanja procesa "umklapp" razpršitve fononov (zaradi velikega parametra mreže) in nereda v strukturi.

Ključne besede: kompleksne kovinske zlitine, toplotna prevodnost, spektralna prevodnost

1 INTRODUCTION

The Al-Mn-Fe system contains several complex metallic alloy phases, which have recently attracted increasing interest. Among them is the orthorhombic Taylor (T) phase, the structure of which is built of atomic layers stacked along the [010]-direction. Along this axis, pentagonal columnar clusters are formed [1]. For this reason, they are considered to be approximants of the decagonal (d) Al-Mn phases. The unit cell of the T-phase contains 156 atoms, with many of the sites having either a fractional occupation or a mixed Al/Mn occupation, so that a great inherent chemical disorder exists on the lattice[2]. As part of a systematic investigation of the transport properties of T-Al-Mn-Fe, here we present the results of the thermal conductivity measurements of T-$Al_{73}Mn_{27-x}Fe_x$ (x = 0,2,4) complex metallic alloys and for a comparison of the d-$Al_{73}Mn_{21}Fe_6$ quasicrystals.

2 EXPERIMENTAL

The polycrystalline samples were produced from the constituent elements by levitation induction melting in a water-cooled copper crucible in argon atmosphere. Parts of the samples were annealed in argon at 900 °C and 930 °C for up to 698 h and subsequently quenched into water[2]. All the samples were T (Taylor) phases of the composition $Al_{73}Mn_{27-x}Fe_x$ (x = 0,2,4,6), except for the d-$Al_{73}Mn_{21}Fe_6$, which was a decagonal quasicrystal.

The thermal conductivity κ of the $Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6) complex metallic alloys was measured in the temperature interval from 2 K to 300 K 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-gold differential thermocouple (gold with 0.07 % of Fe) [3]. The electrical resistivity ρ (conductivity $\sigma = 1/\rho$) was measured between 300 K and 2 K using the standard four-terminal technique.

3 RESULTS AND DISCUSSION

The temperature dependence of the thermal conductivity $\kappa(T)$ for all the investigated samples is shown in **Figure 1.** The thermal conductivity of the T-phases and d-$Al_{73}Mn_{21}Fe_6$ shows characteristic behaviour for the

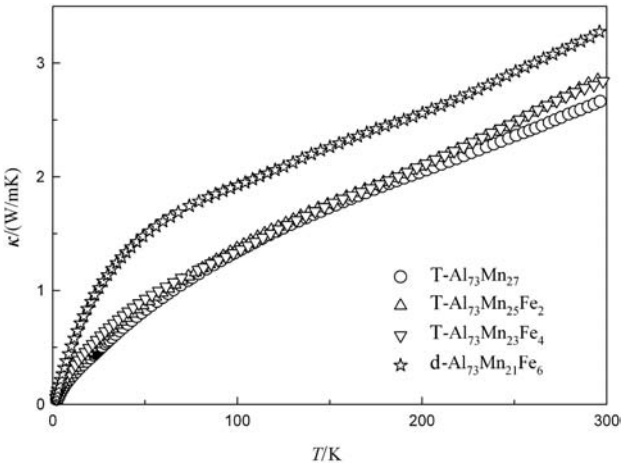

Figure 1: Temperature-dependent thermal conductivity $\kappa(T)$ between 2 K and 300 K for samples (T,d)-$Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6)
Slika 1: Odvisnost med temperaturo in toplotno prevodnostjo $\kappa(T)$ med 2 K in 300 K za zlitine (T,d)-$Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6)

complex metallic alloys ($Mg_{32}(Al,Zn)_{49}$, $Al_{74}Pd_{22}Mn_4$, β-Al_3Mg_2 and ε-phases (Al-Pd-transition metal)[4], which is a relatively small value, a change of slope at about 50 K and a rise of the conductivity above 100 K. At room temperature (300 K) the magnitude of the thermal conductivity is in the interval 2.7–3.3 W/mK for all the samples. Such a small magnitude of the thermal conductivity is a characteristic of thermal insulators like SiO_2 [5] and Zr/YO_2 [6]. The small value of the thermal conductivity has also been found in the icosahedral quasicrystals *i*-Al-Pd-Mn [7,8], which is explained by the small value of electronic density of states at E_F (the small contribution of the electrons to the thermal conductivity) and no periodicity of the sample lattice (the small contribution of the phonons to the thermal conductivity). Our samples (T,d)-$Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6) exhibit a very high electrical resistivity (**Table 1**) compared to simple metals (order of magnitude 1 μΩ cm), so the contribution of the electrons to the thermal conductivity is much smaller than the lattice (phonon) contribution. The low thermal conductivity can be qualitatively explained by the impact of the structure (disorder) of the studied samples on the thermal transport.

Table 1: Values of the electrical resistivity ρ and the thermal conductivity κ at room temperature
Tabela 1: Električna upornost ρ in toplotna prevodnost κ pri sobni temperaturi

Samples	ρ (μΩ cm)	κ (W/mK)
T-$Al_{73}Mn_{27}$	5071	2.69
T-$Al_{73}Mn_{25}Fe_2$	2529	3.01
T-$Al_{73}Mn_{23}Fe_4$	2283	2.83
d-$Al_{73}Mn_{21}Fe_6$	720	3.28

The thermal conductivity model appropriate for complex metallic alloys with a large-scale periodicity of the lattice and a small-scale atomic clustering structure has been described in detail in the previous investigation of the *i*-Al–Pd–Mn system[9].

The electrons (κ_e) and lattice/phonon (κ_l) both contribute to the thermal conductivity $\kappa(T)$:

$$\kappa(T) = \kappa_e(T) + \kappa_l(T) \quad (1)$$

It is common practice to estimate the electronic part using the Wiedemann–Franz law

$$\kappa_e = L_0 T/\rho \quad (2)$$

where $L_0 = 2.44 \times 10^{-8}$ W Ω K^{-2} is the Lorenz number, and ρ is the electrical resistivity.

Here we applied a more elaborate analysis based on the Kubo-Greenwood response theory [10,11,12]. The central quantity of this formalism is the spectral conductivity function that incorporates both the band structure and the transport properties of the system. All the electronic contributions to the transport coefficients, including the resistivity, the thermopower and the thermal conductivity, are related to the spectral conductivity function. We have analyzed both the electrical conductivity and the thermopower to obtain the properties and the shape of the spectral conductivity function in the vicinity of the Fermi level. Our final results on the spectral conductivity function are shown in **Figure 2**. The details of the analysis are given in reference[13]. We should mention that this is a modified version of the procedure originally developed by Landauro and Macia [14,15,16] and is adjusted to suite the experimental data in this class of compounds.

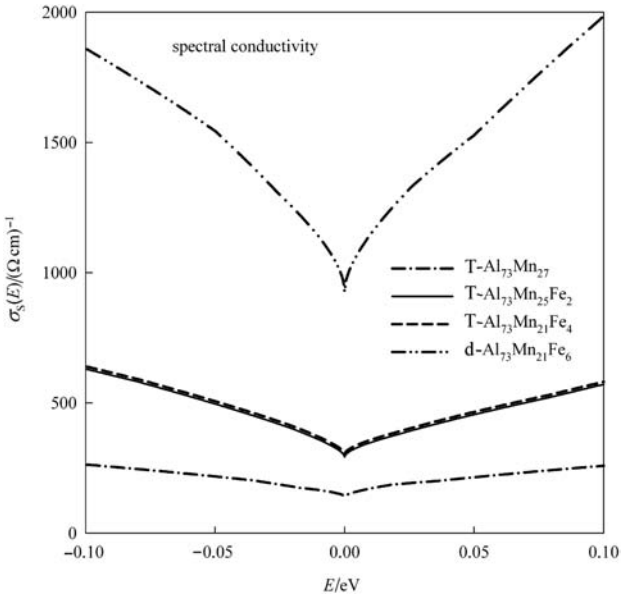

Figure 2: The spectral conductivity function for all the samples. The singularity around the Fermi energy (the energy scale is shifted so that E_F = 0) is clearly pronounced. The sharpness of the pseudogap is directly related to the convex behaviour of the electric conductivity $\sigma(T)$ at low temperatures.
Slika 2: Funkcija spektralne prevodnosti za vse zlitine. Singularnost pri Fermijevi energiji (lestvica za energijo je premaknjena tako, da je E_F = 0) je jasno pokazana. Ostrina psevdoreže je neposredno povezana s konveksnim vedenjem električne prevodnosti $\sigma(T)$ pri nizki temperaturi.

The most important feature of the spectral function in **Figure 2** is the pronounced pseudo gap around the Fermi level. Within the energy range of ±0.1 eV the spectral function loses around 40 % of its spectral weight. Moreover, our analysis, based on transport measurements at low temperatures, reveals the fine structure of the pseudo gap, featuring the $|E|^{1/2}$ singularity at the Fermi level.

Once calculated the spectral conductivity function can be used to determine the electronic contribution, κ_e, to the thermal conductivity. The results of the different contributions at room temperature (RT) are presented in **Table 2**. The lattice part is significantly larger than the electronic part. This is because the electrical resistivity of these samples is very high, so consequently the electrons also have a small contribution to the heat transport.

Table 2: Thermal conductivity κ, electronic contribution κ_e calculated by the model of spectral conductivity, electronic contribution κ_{e0} calculated by the WF law, difference between the two former electronic contributions in (%) and ratio effective and normal Lorentz number L_{eff}/L_o at T = 300 K

Tabela 2: Toplotna prevodnost κ, elektronski delež κ_e, izračunan po modelu spektralne prevodnosti, in elektronski delež κ_{e0} izračunan po WF-zakonu, razlika med obema v odstotkih in razmerje med efektivnim in normalnim Lorentzovim številom L_{eff}/L_o pri T = 300 K.

Sample	κ/ (W/mK)	κ_e/ (W/mK)	$\kappa_{e,o}$/ (W/mK)	$(\kappa_e-\kappa_{e,o})/\kappa$ (%)	L_{eff}/L_o
T-$Al_{73}Mn_{27}$	2.69	0.18	0.15	1	1.25
T-$Al_{73}Mn_{25}Fe_2$	3.01	0.43	0.29	4	1.40
T-$Al_{73}Mn_{23}Fe_4$	2.83	0.41	0.32	3	1.30
d-$Al_{73}Mn_{21}Fe_6$	3.28	1.30	1.02	9	1.30

Although the WF law is not valid, the difference between the electronic contribution obtained by the spectral conductivity model κ_e and the one obtain using the WF law $\kappa_{e,o}$ is only a few percent (except for d-$Al_{73}Mn_{21}Fe_6$, where it reaches up to 10 %). So, we can still use the WF law to predict the electronic contribution to the heat transport as a rough approximation.

The lattice contribution $\kappa_l = \kappa - \kappa_e$ is analyzed by considering the propagation of long-wavelength phonons within the Debye model and the hopping of the localized vibrations. This picture assumes that large atomic clusters of icosahedral symmetry strongly suppress the propagation of phonons in the lattice of complex metallic alloys. The exceptions are the long-wavelength acoustic phonons, for which this material is an elastic continuum, and fracton-like localized vibrations within the cluster substructure that can participate in the heat transfer via thermally activated hopping. In the simplest model, the hopping of localized vibrations is described by the single activation energy E_a, yielding a contribution to the thermal conductivity[9]

$$\kappa_H = \kappa_H^0 \exp\left(-\frac{E_a}{k_B T}\right) \quad (3)$$

where κ_H^0 is a constant. The Debye thermal conductivity is written as:

$$\kappa_D = C_D T^3 \int_0^{\theta_D/T} \tau(x) \frac{x^4 e^x}{(e^x - 1)^2} dx \quad (4)$$

where $C_D = k_B^4/2\pi^2\bar{v}\hbar^3$, $\bar{v}$ is the average velocity of sound, θ_D is the Debye temperature, τ is the phonon relaxation time, $x = \hbar\omega/k_BT$, and $\hbar\omega$ is the phonon energy. The different phonon-scattering processes are incorporated into the relaxation time $\tau(x)$ and we assume that 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 an analogy with the ε-phases in Al-Pd-Mn[9], we consider two dominant scattering processes in the investigated temperature interval (from 2 to 300 K). First, the scattering of phonons on the structural defects of the stacking-fault type with the scattering rate

$$\tau_j^{-1} = \frac{7}{10} \frac{a^2}{\bar{v}} \gamma^2 \omega^2 N_s \quad (5)$$

where a is a lattice parameter, γ is the Grüneisen parameter and N_s is the linear density of the stacking faults.

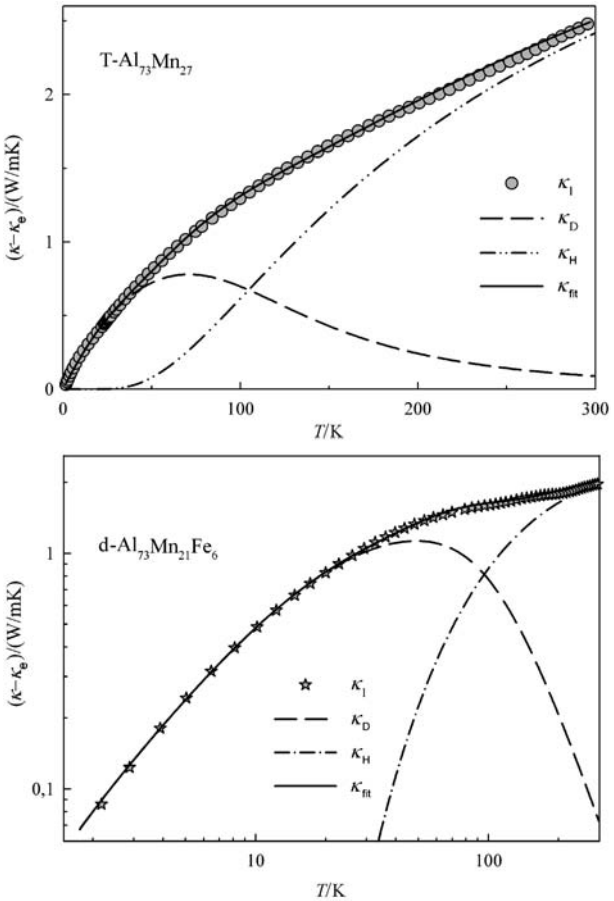

Figure 3: Temperature dependent lattice thermal conductivity κ_l (filled symbols) together with the two contributions: the Debye contribution κ_D (dashed line) and the hopping contribution κ_H (dash-dot line) of T-$Al_{73}Mn_{27}$ and d-$Al_{73}Mn_{21}Fe_6$ shown in different scales.

Slika 3: Temperaturna odvisnosti mrežne toplotne prevodnosti κ_l (polni znaki) z dvema deležema: Debyejev delež κ_D (črtkana črta) in delež "hopping" κ_H (črtopična črta) za T-$Al_{73}Mn_{27}$ in d-$Al_{73}Mn_{21}Fe_6$, prikazani z različnima lestvicama

The second scattering mechanism is the *umklapp* processes with the phenomenological form of the scattering rate pertinent to complex metallic alloys [9], $\tau_{um}^{-1} = Bx^{\alpha} T^{4-\alpha}$ and for the total scattering rate we get $\tau^{-1} = \tau_{sf}^{-1} + \tau_{um}^{-1}$. The Debye temperature of the investigated *T*-phases is not known; therefore, we have used the θ_D value reported for the related icosahedral *i*-Al–Pd–Mn quasicrystals, were θ_D was commonly found to be close to 500 K[9,19]. Since our $\kappa(T)$ data are available only up to 300 K, it turns out that the fit is insensitive to a slight change of this θ_D value, so a fixed $\theta_D = 500$ *K* is used. The Debye constant C_D was also not taken as a free parameter, but was instead calculated using $\bar{v}$ = 4000 m s^{-1}, a value determined for the *i*-Al–Pd–Mn from ultrasonic data. The results of a fitting procedure using the relation $\kappa_l(T) = \kappa_D(T) + \kappa_H(T)$ to the experimental data are shown for T-$Al_{73}Mn_{27}$ and for d-$Al_{73}Mn_{21}Fe_6$ in **Figure 3**. The parameters of the fitting procedure for all the investigated samples are shown in **Table 3**. From **Figure 3** it is easy to see that the Debye contribution $\kappa_D(T)$ has a maximum at about 50 K, although it becomes smaller at higher temperatures. A similar behavior is conventional for periodic structures, where the origin of such behaviour is in the phonon-phonon umklapp scattering processes[21].

Table 3: The fit parameters for the lattice thermal conductivity $\kappa_l = \kappa_D + \kappa_H$

Tabela 3: Parametri ujemanja z mrežno toplotno prevodnost $\kappa_l = \kappa_D + \kappa_H$

Sample	*A* (10^7 s^{-1} K^{-2})	*B* (10^4 s)	κ_{H0} (W/mK)	E_a (meV)
T-$Al_{73}Mn_{27}$	5.6	1.8	4.8	17.7
T-$Al_{73}Mn_{25}Fe_2$	4.8	1.7	5.2	19.6
T-$Al_{73}Mn_{23}Fe_4$	4.4	2.1	4.9	18.5
d-$Al_{73}Mn_{21}Fe_6$	2.3	2.3	3.3	11.3

The parameter *A*, which describes the structural defects of the stacking-fault type, is close to 10^7 s^{-1} K^{-2} for all samples. It is possible to estimate from *A* the linear density of stacking faults N_s. If we take typical values for the lattice parameter $a \approx 1.4$ nm and the Grüneisen parameter $\gamma \approx 2$, we obtain $N_s = 10A\bar{v}\hbar^2 / 7\alpha^2\gamma^2k_B^2 = 0.8$ μm^{-1}. This micrometer-scale N_s value is comparable to those reported for the ψ-Al-Pd-Mn[8] i-Al-Pd-Mn[22] and the decagonal d-Al-Mn-Pd[23]. Therefore, the stacking-fault-like structural defects may be considered as the source of the phonon scattering at low temperatures in the (T,d)-$Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6) samples. The parameters *B* and α define the phonon scattering by the umklapp processes in a phenomenological way. From the fitting procedure we get $\alpha = 1$, so the frequency and temperature dependence of the umklapp term is $\tau_{um}^{-1} \sim \omega T^3$. For all the samples, the hopping contribution κ_H becomes significant above 100 K. The activation energy E_a for all the samples is between 10 meV and 20 meV and is smaller by a factor of 2 than the E_a of ψ-Al-Pd-Mn[9]. This smaller E_a value reflects the considerably less steep $\kappa(T)$ increase at temperatures above 100 K for our samples compared to ψ-Al-Pd-Mn. On the other hand, the above E_a values correlate with the inelastic neutron[24] and x-ray[25] scattering experiments on i-Al-Pd-Mn quasicrystals, where dispersionless vibrational states were identified for the energies higher than 12 meV. In quasicrystals such dispersionless states indicate localized vibrations and are considered to be a consequence of the dense distribution of energy gaps in the phonon excitation spectrum. This prevents extended phonons from propagating through the lattice, whereas localized vibrations may still be excited. Therefore, localized vibrations also appear to be present in the giant unit cell of (T,d)-$Al_{73}Mn_{27-x}Fe_x$ samples, where their origin may be attributed to the cluster substructure.

4 CONCLUSION

We have investigated the thermal conductivity of (T,d)-$Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4, 6) samples, which show typical behaviour of $\kappa(T)$ for complex metallic alloys, i.e., relatively small values (thermal insulators), a change of the slope at about 50 K and an increase of the conductivity above 100 K. We have separated $\kappa(T)$ into the electron $\kappa_e(T)$ and lattice (phonon) $\kappa_l(T)$ parts, which both have small values. The electron part was determined by the spectral conductivity model, and it is much smaller than the lattice part. The reason is the very high electrical resistivity of all the samples, which reduces the electron thermal conductivity. The lattice's thermal conductivity is greatly reduced because of the enhanced umklapp processes of the phonon scattering (caused by a large lattice constant and, consequently, a small Brillouin zone) and by disorder in the structure.

Acknowledgement

This work was carried out within the activities of the 6th Framework EU Network of Excellence "Complex Metallic Alloys" (Contract No. NMP3-CT-2005-500140), and has been supported in part by the Ministry of Science, Education and Sports of Republic of Croatia through the Research Projects: 035-0352826-2848 "Thermal and charge transport in highly frustrated magnets and related materials"; 035-0352826-2847 "Modelling physical properties of materials with marked frustration or disorder" and 119-1191458-0512 "Low-dimensional strongly correlated conducting systems".

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