Journal of Alloys and Compounds, 189 (1992) 217–220
JALCOM 380

Thermal conductivity of $UCuP_2$ and $UCuAs_2$ single crystals

J. Mucha, H. Misiorek, D. Kaczorowski and A. Jeżowski
W. Trzebiatowski Institute for Low Temperature and Structure Research, Polish Academy of Sciences, 50-950 Wrocław (Poland)

(Received May 27, 1992)

Abstract

We report thermal conductivity measurements on single crystals of ferromagnetic $UCuP_2$ and $UCuAs_2$. The results obtained are discussed in the framework of various contributions to the total thermal conductivity of magnetically ordered solids.

1. Introduction

A new family of ternary uranium compounds with the general formula UTX_2, where T is a 3d- or 4d-transition metal and X is phosphorus, arsenic, antimony or bismuth, has attracted much interest in recent years because of its remarkable physical properties. For the most intensively studied $UCuP_2$ and $UCuAs_2$ we reported previously in a series of publications the structural [1, 2], magnetic [3], electrical transport [3–5] and magneto-optical [6, 7] data. In this communication we present the results of thermal conductivity measurements performed on single-crystal samples of both of the above pnictides.

2. Experimental details

Single crystals of $UCuP_2$ and $UCuAs_2$ were grown by the chemical transport method, as described previously [1, 2]. The crystals obtained had the form of thin plates oriented perpendicular to the crystallographic *c*-axis. The specimens for thermal conductivity measurements were cut from the largest pieces using a diamond-wire saw and had dimensions of 10 mm × 2 mm × 0.2 mm.

The thermal conductivity was measured over the temperature range 4.2–300 K using the axial stationary heat flow method. The absolute temperature as well as the temperature gradient along the sample were determined with a manganin–constantan thermocouple. An average error in the measured thermal conductivity coefficient λ was about $\pm 1.5\%$.

3. Results and discussion

The total thermal conductivity of a solid may be regarded as a sum of three contributions: an electronic component λ_e, a phonon component λ_{ph} and a magnon component λ_m [8]. Assuming that all the scattering mechanisms responsible for the heat transport in a metal are additive (the Matthiessen rule), the electronic contribution to the thermal conductivity can be expressed as follows

$$\frac{1}{\lambda_e} \equiv W_e = W_{e,i} + W_{e,ph} + W_{e,m} \quad (1)$$

The particular terms occurring in the above equation denote the thermal resistivity due to collisions of the conduction electrons with lattice imperfections, phonons and magnetic moments respectively. A similar formula can also be written for the lattice component of the thermal conductivity

$$\frac{1}{\lambda_{ph}} \equiv W_{ph} = W_{ph,i} + W_{ph,e} + W_{ph,ph} + W_{ph,m} \quad (2)$$

where the subsequent terms characterize scattering of phonons on impurities and/or defects, conduction electrons, lattice vibrations and magnetic moments respectively. The scattering of electrons and phonons on lattice imperfections is elastic and these mechanisms are the most important at low temperatures. In contrast, the electron–phonon and phonon–phonon interactions may have an elastic as well as an inelastic character and they are described by processes of the normal and Umklapp types. As shown in ref. 9, scattering of the conduction electrons on localized magnetic moments (the $W_{e,m}$ term in eqn. (1)) usually plays a substantial role in compounds exhibiting the Kondo effect and strong crystal field interactions (as in $UCuP_2$ and $UCuAs_2$ [3]).

0925-8388/92/$5.00

The third contribution to the total thermal conductivity, *i.e.* the magnon component λ_m, is expected to appear in magnetically ordered solids below their ordering temperature. It originates mainly from the interactions of spin wave excitations with electrons and phonons. The temperature variation of λ_m may be determined experimentally by measuring the thermal conductivity of the sample with and without an applied magnetic field. In a strong field the magnetic moments are "frozen" and the magnon contribution is given by the difference

$$\lambda_m = \lambda_{H=0} - \lambda_H \tag{3}$$

In the literature, indirect methods of finding λ_m are also described [10–13]. At low temperatures the magnon and phonon contributions to the total thermal conductivity may be of comparable magnitudes. With increasing temperature the value of the ratio λ_m/λ_{ph} usually strongly decreases and becomes close to zero in the vicinity of the magnetic phase transition.

The uranium compounds studied here, *i.e.* $UCuP_2$ and $UCuAs_2$, are semimetals with strongly damped free carrier concentrations [4–7]. Hence, for these pnictides the phonon contribution to the total thermal conductivity is expected to be significant. Moreover, owing to their ferromagnetic character [3] the magnon contribution should play some role at low temperatures. The measured thermal conductivity of $UCuP_2$ and $UCuAs_2$ is displayed in Fig. 1. In the paramagnetic region, the total thermal conductivity for both compounds decreases with decreasing temperature and the conductivity of the phosphide is twice as large as that of the arsenide. This feature agrees well with the results of the electrical resistivity measurements [3]: $\rho(UCuP_2) < \rho(UCuAs_2)$.

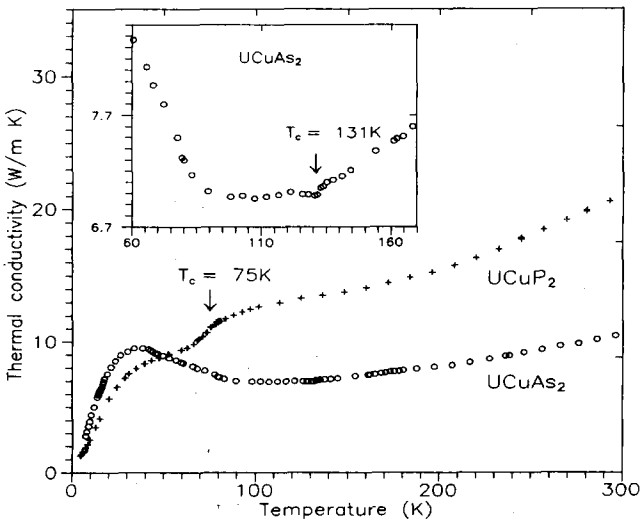

Fig. 1. Temperature dependence of the thermal conductivity of single-crystal $UCuP_2$ and $UCuAs_2$. The inset shows the thermal conductivity of $UCuAs_2$ in the vicinity of the magnetic phase transition at $T_C = 131$ K.

As seen from Fig. 1 and the inset to this figure, the magnetic phase transition manifests itself on the $\lambda(T)$ curves either as a very shallow minimum at $T_C = 131$ K for $UCuAs_2$ or as a kink at $T_C = 75$ K in the case of $UCuP_2$. In the ferromagnetic region, the thermal conductivity of the latter compound decreases continuously with decreasing temperature, showing only a shoulder at about 50 K. In contrast, the thermal conductivity of $UCuAs_2$ appears to be almost constant from T_C down to about 90 K, then goes through a broad maximum centred around 35 K, and finally rapidly decreases.

Assuming a relation between the thermal conductivity and the electrical resistivity ρ_0, given by the Wiedemann–Franz law, $\lambda\rho/T = L_0$ ($L_0 = 2.45 \times 10^{-8}$ W Ω K^{-2}), we derived the expected temperature variation of the electronic contribution to the total thermal conductivity for both the compounds investigated. In this procedure the electrical resistivity data from ref. 3 were used. The calculated $\lambda_e(T)$ dependences are shown in Figs. 2 and 3 for $UCuP_2$ and $UCuAs_2$ respectively.

At low temperatures the electrical resistivity for both pnictides was shown [3] to follow the equation

$$\rho(T) = \rho_0 + c_m T^2 \tag{4}$$

where ρ_0 is the residual resistivity and the second term describes scattering processes of the electron–magnon type. Therefore, with rising temperature, a maximum of $\lambda_e(T)$, occurring between the region of a linear increase and a hyperbolic decrease in the thermal conductivity, might be expected. Instead of this, only a smooth increase in λ_e with temperature is observed for both compounds studied. A reason for this feature seems to be large values of the residual resistivity ρ_0 which dominate the T^2 term over the whole temperature range concerned.

In the paramagnetic region the resistivity of $UCuP_2$ and $UCuAs_2$ varies according to the formula

$$\rho(T) = \rho_0 + \rho_0^{\infty} + c_{ph} T - c_K \ln T \tag{5}$$

where in addition to the residual resistivity there is also a temperature independent spin-disorder term ρ_0^{∞}, the linear phonon contribution and the logarithmic Kondo term. Then, the thermal conductivity in this range should be nearly proportional to T and is expected to show a tendency to saturation at high temperatures. In fact, such behaviour of $\lambda_e(T)$ in the paramagnetic region is observed for our samples (see Figs. 2 and 3).

The difference $\lambda - \lambda_e$ may be regarded as a measure of the lattice contribution λ_{ph} to the total thermal conductivity, enlarged in the ordered region by the magnon component λ_m. The sum $\lambda_{ph} + \lambda_m$ derived for $UCuP_2$ and $UCuAs_2$ is shown in Figs. 2 and 3 respectively. Assuming that at low temperatures the

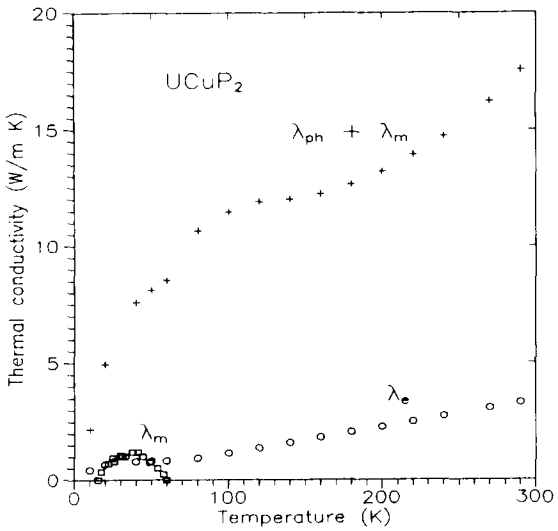

Fig. 2. Temperature variation of the electronic λ_e, lattice + magnon $\lambda_{ph}+\lambda_m$, and magnon λ_m contributions to the total thermal conductivity for $UCuP_2$.

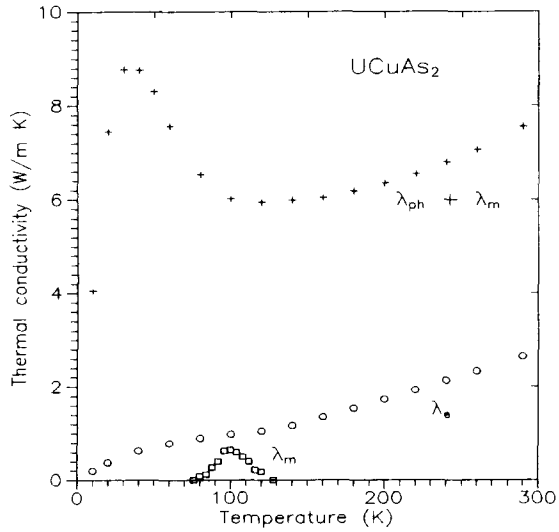

Fig. 3. Temperature variation of the electronic λ_e, lattice + magnon $\lambda_{ph}+\lambda_m$, and magnon λ_m contributions to the total thermal conductivity for $UCuAs_2$.

phonon contribution in the compounds studied varies with temperature as in non-magnetic isolators [8], we were able to estimate the magnitude of the magnon component λ_m. In this procedure graphical methods described in refs. 10–13 were used. The results are presented in Table 1 and in Figs. 2 and 3. As seen, in the case of $UCuP_2$, $\lambda_m(T)$ reaches its maximum at about 40 K where it is slightly larger than the electronic component λ_e. Above 60 K the magnon contribution in this compound is already close to zero. For $UCuAs_2$, λ_m appears to be significantly smaller in comparison with the phosphide. It only becomes measurable above 75 K, goes through a maximum at around 100 K and finally vanishes at $T_C = 131$ K.

TABLE 1. The magnon contribution to the thermal conductivity of single-crystal $UCuP_2$ and $UCuAs_2$ as estimated at several temperatures below the corresponding values of T_C

$UCuP_2$		$UCuAs_2$	
T (K)	λ_m/λ (%)	T (K)	λ_m/λ (%)
20	12.5	80	1.1
40	14.3	100	9.3
50	8.4	120	2.6

In the paramagnetic region the phonon contribution to the total thermal conductivity is determined mainly by phonon–electron and phonon–phonon interactions of the Umklapp type (see eqn. (2)). In this region λ_{ph} is expected to transform from an exponential decrease with increasing temperature to the T^{-1} dependence for $T \geqslant \Theta_D$ [14]. Instead, the phonon component in $UCuP_2$ and $UCuAs_2$ continuously increases with increasing temperature. This feature is not caused by heat radiation because almost the same results were obtained using two different sets of equipment. One of them was specially constructed for thermal conductivity measurements on small samples at high temperatures [15]. It is worthwhile noting that a similar increase in thermal conductivity at high temperatures has been observed for cerium Kondo-like compounds, such as $CeCu_2Si_2$ [16], $CeAl_3$ [17], $CeCu_6$ [18] or $Ce(Cu_{1-x}Al_x)_5$ [19]. Theoretically, a linear temperature dependence of λ_{ph} has been predicted by Zimmermann [20] in materials where the electronic mean free path is comparable with or smaller than the predominant phonon wavelength. The pnictides studied, being semimetals with rather larger values of the residual resistivity, belong to this class of compounds.

Finally, we should note here that the separation of the particular contributions to the total thermal conductivity of $UCuP_2$ and $UCuAs_2$, presented above, should be treated only semiquantitatively because of the crudeness of the models applied. Moreover, we only considered the main mechanisms responsible for the thermal resistivity of our samples, neglecting other scattering processes which may also strongly influence the heat transport in the pnictides investigated.

Acknowledgments

The authors are indebted to Professors W. Suski and R. Troć for their kind interest in this study.

References

1 J. Stępień-Damm, D. Kaczorowski and R. Troć, *J. Less-Common Met., 132* (1987) 15.

2 H. Noël, Z. Żołnierek, D. Kaczorowski and R. Troć, *J. Less-Common Met., 132* (1987) 327.
3 D. Kaczorowski, R. Troć and H. Noël, *J. Phys. Condens. Matter, 3* (1991) 4959.
4 D. Kaczorowski and J. Schoenes, *Solid State Commun., 74* (1990) 143.
5 N. Korner, J. Schoenes and D. Kaczorowski, *Helv. Phys. Acta, 62* (1989) 207.
6 J. Schoenes, P. Fumagalli, H. Rûegsegger and D. Kaczorowski, *J. Magn. Magn. Mater., 81* (1989) 112.
7 P. Fumagalli, J. Schoenes and D. Kaczorowski, *Solid State Commun., 65* (1988) 173.
8 R. Berman, *Thermal Conductivity in Solids,* Clarendon Press, Oxford, 1976.
9 A. K. Bhattacharjee and B. Coqblin, *Phys. Rev. B, 38* (1988) 338.
10 W. B. Yelon and L. Berger, *Phys. Rev. Lett., 25* (1970) 1207.
11 Y. Hsu and L. Berger, *Phys. Rev. B, 14* (1976) 4059.
12 A. Kumar, *Phys. Rev. B, 25* (1982) 3369.
13 J. A. Smirnov and W. J. Tamarczenko, *Elektronnaja tiepłoprowodnost w metalach i połuprowodnikach,* Nauka, Leningrad, 1972, in Russian.
14 G. Leibfried and E. Schlömann, *Nachr. Akad. Wiss. Göttingen, 2A, 4* (1954) 71.
15 A. Jeżowski, J. Mucha and G. Pompe, *J. Phys. D, 20* (1987) 1500.
16 W. Franz, A. Griessel, F. Steglich and D. Wohlleben, *Z. Phys. B, 31* (1978) 7.
17 H. R. Ott, O. Marti and F. Hulliger, *Solid State Commun., 49* (1984) 1129.
18 A. Amato, D. Jaccard, J. Flouquet, F. Lapierre, J. L. Tholence, R. A. Fisher, S. E. Lacy, J. A. Olsen and N. E. Phillips, *J. Low Temp. Phys., 68* (1987) 371.
19 E. Bauer, E. Gratz, G. Hutflesz and H. Müller, *J. Phys. Condens. Matter, 3* (1991) 7641.
20 J. E. Zimmermann, *J. Phys. Chem. Solids, 11* (1959) 299.