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Thermal Conductivity of $Ce_2Ru_3Ga_9$ Compound

M. Falkowski* and A.M. Strydom
Highly Correlated Matter Research Group, Physics Department, University of Johannesburg,
PO Box 524, Auckland Park 2006, South Africa

The Ce-based 2:3:9 series of compounds are known for strongly correlated $4f$-electron behaviour. Here, we report for the first time a study of the thermal conductivity $\kappa(T)$ in zero and 9 T magnetic field for $Ce_2Ru_3Ga_9$ across the temperature range 2 K $\le T \le$ 300 K. The zero-field temperature dependence of $\kappa(T)$ exhibits a pronounced maximum, characteristic for metals with large electronic mean free path and towards room temperature $\kappa(T)$ starts behaving in a manner usually attributed to the enhanced electron–phonon coupling. Based on the Wiedemann–Franz law the electronic and lattice contributions to the thermal conductivity were estimated. In high temperature region a distinct step-like anomaly at $T^* = 203$ K has been observed which signals a putative phase transition, probably of phononic or lattice origin. We furthermore discuss the effect of applied magnetic fields on the thermal transport in $Ce_2Ru_3Ga_9$.

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1. Introduction

$Ce_2Ru_3Ga_9$ is one of the representatives of the group of ternary compounds $Ce_2T_3X_9$ (T = Rh, Ru, Ir; X = Al, Ga), crystallizing in the orthorhombic $Y_2Co_3Ga_9$-structure with space group *Cmcm* [1–5]. The physical properties of the mentioned compounds indicate an important role of correlations between the Ce $4f$-electron orbital with conduction states and also on the electronic structure of valence band which depends on the constitution of T and X atoms. Previous experimental studies have shown that the nature of the ground state in these compounds remains controversial, i.e. exhibits coexistence of intermediate valent type of behaviour in magnetic susceptibility $\chi(T)$ and the Kondo interaction in electrical resistivity $\rho(T)$, suggest the existence of two energy scales [1–3, 5]. Kumar et al. [3] reported that for $Ce_2Ru_3Ga_9$ the temperature behaviour of $\chi(T)$ reveals a broad maximum which is usually attributed to fluctuating valence materials, while the shape of $\rho(T)$ is qualitatively similar to many hybridized $4f$ compounds [6] and has some common features with the previously studied $Ce_2T_3X_9$ (T = Rh, Ru, Ir; X = Al, Ga) compounds [1–3, 5]. The estimated Sommerfeld coefficient from specific heat yields a value of 125 mJ/(mol Ce K^2), showing a moderate mass enhancement near Fermi level due to the electron correlations. The strength of hybridization between $4f$ and conduction electrons has a large impact to destabilize the Ce moments giving rise to the Pauli paramagnetic and mixed valence behavior, and the anomalous thermal variation in resistivity together with high Sommerfeld coefficient value indicate strong electronic correlations associated with $4f$ states in $Ce_2Ru_3Ga_9$.

In this paper we report the experimental results of the thermal conductivity of $Ce_2Ru_3Ga_9$ measured across the temperature range 2 K $\le T \le$ 300 K, along with the response of $\kappa(T)$ to the applied magnetic field $B = 9$ T.

2. Experimental details

A polycrystalline sample of $Ce_2Ru_3Ga_9$ has been synthesized by arc-melting technique using high purity elements (99.99 wt.% or better) on water-cooled hearth in a ultra-high purity argon atmosphere with further *in situ* purification of argon. To improve homogeneity the ingot was inverted and remelted several times. The sample was annealed for two weeks at 600 °C in evacuated silica tube. Homogeneity and phase quality was examined by room temperature powder X-ray diffraction analyses of the sample which confirmed the single phase character (see Fig. 1). The additional electron-dispersive X-ray analysis showed no impurities of other elements and led to the following compositions: Ce: 13.5 at.%, Ru: 21.9 at.%, Ga: 64.6 at.%, confirmed the desired stoichiometry of the $Ce_2Ru_3Ga_9$.

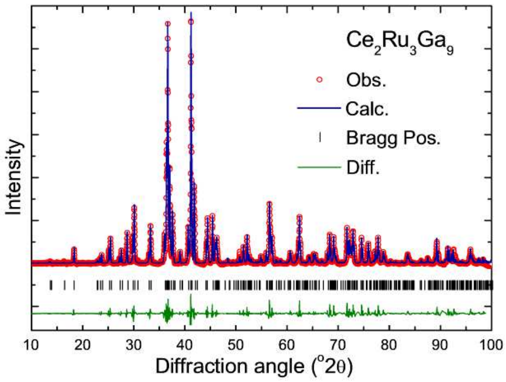

Fig. 1. Room temperature powder X-ray diffraction spectrum. The vertical bars successfully index the orthorhombic $Y_2Co_3Ga_9$-type structure (space group *Cmcm*) with the lattice parameters: $a = 12.939(7)$ Å, $b = 7.620(4)$ Å, $c = 9.705(6)$ Å. The solid line is the full-profile of the Rietveld refinement using General Structure Analysis System (GSAS) software.

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*corresponding author; e-mail: `falkowski@ifmpan.poznan.pl`

The thermal conductivity was measured over the temperature interval 2–300 K in zero and magnetic field $B = 9$ T using PPMS-9T apparatus (Quantum Design, San Diego). The thermal transport sample was cut in a bar-shaped form with approximate dimensions: $1.9\times1.3\times6.5\,\mathrm{mm}^3$. The thermometer and heater contacts were established to the sample by four gold-coated copper contact leads, using electrically and thermally conductive epoxy. The thermal transport was measured by applying a direct heat-pulse with steady state heat-flow technique performed during the course of a slow cooling process from room temperature to 2 K, maintaining a good vacuum ($\approx 10^{-5}$ Torr).

3. Results and discussion

Figure 2a shows zero-magnetic field temperature dependence of total measured thermal conductivity $\kappa(T)$ of $Ce_2Ru_3Ga_9$ up to 300 K, with its lattice $\kappa_\mathrm{L}(T)$ and electronic $\kappa_\mathrm{e}(T)$ components. Applying the Wiedemann–Franz law the $\kappa_\mathrm{e}(T) = L_0T/\rho$, with the Lorenz number $L_0 = \pi^2 k_\mathrm{B}^2/3\mathrm{e} = 2.45\times10^{-8}\ \mathrm{W}\,\Omega/\mathrm{K}^2$ and $\kappa_\mathrm{L}(T) = \kappa(T) - \kappa_\mathrm{e}(T)$ contributions have been estimated. In simple metals the $\kappa(T)$ is usually dominated by the free electron gas but the lattice heat conductivity, and consequent effects such as the scattering of phonons by structural defects and quasiparticles like other phonons or electrons should also be considered. We may clearly see from Fig. 2a that the lattice thermal conductivity dominates over electronic contribution in entire measured temperature range, indicating that phonons are main heat carriers. The overall character of total thermal conductivity of $Ce_2Ru_3Ga_9$ exhibits several interesting features. With increasing temperature $\kappa(T)$ is characterized by a strong increase reached at a maximum $T_\mathrm{max} = 37$ K, which frequently characterizes metals and compounds with large electronic mean free path, i.e. showing low scattering rates on static imperfections. However, the absolute value of $\kappa(T)$ is quite low compared with the thermal conductivity for good metals having an absence of free electrons and therefore which can reflect the presence of an additional contribution due to high heat conduction via phonons, reducing thermal conductivity.

With further increasing temperature $\kappa(T)$ starts behaving in manner usually ascribed to the enhanced electron–phonon coupling [7], until the appearance of a pronounced step-like anomaly at $T^* = 203$ K, which is also visible in $\kappa_\mathrm{L}(T)$ and specific heat data (not shown here) at the same temperature. However, we would like to note that we did not observe this anomaly in our measurements of thermopower, electrical resistivity and magnetic susceptibility. In order to be certain that the appearing anomaly is not directly caused by possible problems with contacts we performed the same measurement using two separate PPMS apparatus (using the same sample, with the same contact leads) and in result we obtained the similar effect. On the other hand, the possibility of low metallurgical quality of the sample, mainly related with micro-cracks cannot be excluded ultimately as the source of this singularity, but most likely the anomaly at T^* may signal a phase transition, probably of phononic origin. Nevertheless, to confirm this aspect, low-temperature X-ray or neutron diffraction measurements are needed to unambiguously determine the temperature relationship of this compound.

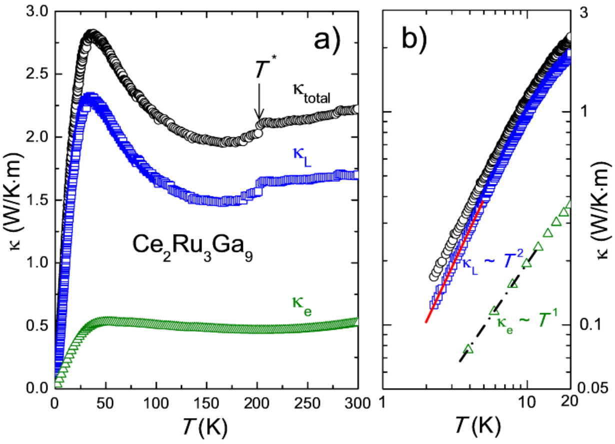

Fig. 2. (a) Temperature dependence of the total thermal conductivity $\kappa(T)$ measured in zero-magnetic field together with the electronic $\kappa_\mathrm{e}(T)$ and lattice $\kappa_\mathrm{L}(T)$ components estimated using the Wiedemann–Franz law. (b) Log–log plot of the low-temperature region with various temperature relations for respective contributions described in text.

In Fig. 2(b) we plot in log–log scale the low-temperature behaviour of respective thermal conductivity contributions for $Ce_2Ru_3Ga_9$. We noticed that below 5 K the lattice thermal conductivity follows $\kappa_\mathrm{L}(T) \approx T^2$ mainly due to dominance of phonon scattering from conduction electrons, which is the most efficient scattering process for phonons in low temperatures. For electronic thermal conductivity component the low-temperature dependence behaves in linear manner $\kappa_\mathrm{e}(T) \approx T$ up to 12 K indicates the dominance by the elastic electron scattering on static lattice imperfections.

The temperature dependence of the normalized Lorenz number L/L_0 up to 300 K for $Ce_2Ru_3Ga_9$ is displayed in Fig. 3. This general character of L/L_0 vs. T resembles the behaviour which is commonly found in other Ce-based compounds [8–10]. The major feature at intermediate temperatures is a well-defined peak at 15 K, followed by a shallow minimum at ≈ 150 K. The thermal anomaly at T^* is also clearly seen in L/L_0 vs. T, and is here expressed as a peak at 203 K. In pure metals at low-temperature region the ratio $L/L_0 \approx 1$, as a result of the predominance of electronic type of scattering mechanisms and electrons as charge carriers in thermal transport. The enhanced value of L/L_0 of investigated compound compared to the value of ratio $L/L_0 \approx 1$ may designate on phonons as the dominant heat carriers in thermal transport, thereby suggesting that the electron scattering of charge carriers play a role of minor importance. The deviation of L/L_0 from the theoretical Lorenz number is observable in the whole temperature range, especially at low-temperature where L/L_0 deviates considerably from the theoretical value. This higher value of L/L_0 could be due to the dominance of the $\kappa_L(T)$ with respect to the $\kappa_e(T)$ contribution. However, it should be noted that in strongly correlated materials treatment of electronic scattering phenomena is notoriously difficult in the absence of a quantitative theory.

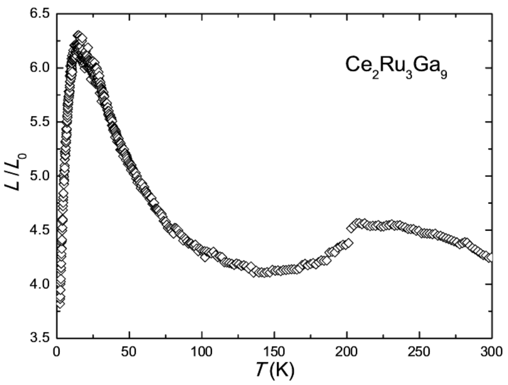

Fig. 3. Temperature dependence of the Lorenz number normalized to the free-electron number for $Ce_2Ru_3Ga_9$.

We also investigated the response of thermal conductivity to applied magnetic field, $B = 9$ T, over the whole accessible temperature range. Figure 4 shows the temperature dependence of total thermal conductivity in zero and 9 T magnetic field of $Ce_2Ru_3Ga_9$. At low temperatures, the $\kappa(T)$ is affected by the applied magnetic field up to approximately 10 K. The effects of magnetic field on $\kappa(T)$ are most pronounced over the temperature range 10 K $< T <$ 150 K, i.e. showing significant influence in that region where $\kappa_L(T)$ exhibits a maximum.

Inset in Fig. 4 shows the influence of magnetic field in the critical region where the anomaly at T^* is present. Apparently from the main graph of Fig. 4, the effect of magnetic field on the anomaly region is rather faint, however, in the inset Fig. 4 it is discerned that the maximum of the anomaly is slightly shifted towards lower temperature.

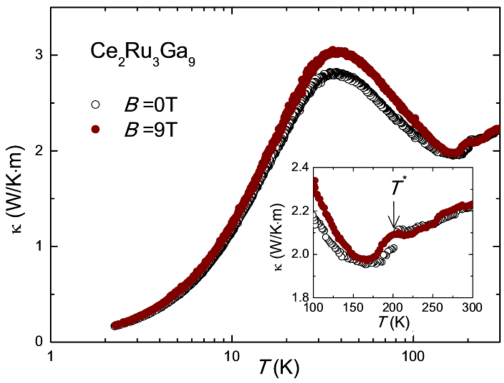

Fig. 4. Semi-log plot of the total thermal conductivity of $Ce_2Ru_3Ga_9$ measured in zero field (open circles) and 9 T (solid circles). Inset shows the influence of the applied magnetic field on the anomaly at T^*.

4. Conclusions

In summary, our study of thermal conductivity of $Ce_2Ru_3Ga_9$ compound revealed the following features:

1. The absolute value of $\kappa(T)$ is low in respect to the thermal conductivity for simple metals.

2. $\kappa_L(T)$ dominates towards $\kappa_e(T)$ in entire measured temperature range, indicating that phonons are main heat carriers.

3. In high temperature region a distinct step-like anomaly at $T^* = 203$ K appears, which changed the nature of the slope in $\kappa(T)$, $\kappa_L(T)$ and the ratio L/L_0. It cannot be excluded that the anomaly at T^* may signals the transition to a new structural phase.

4. L/L_0 vs. T resembles the behaviour usually observed in other Ce-based compounds, however, the enhanced value of L/L_0 supports that phonons are the dominant heat carriers in $Ce_2Ru_3Ga_9$.

5. The pronounced magnetic field effect has been observed in the temperature region where $\kappa(T)$ and consequently $\kappa_L(T)$ exhibits a maximum. Interestingly, we found a weak but resolvable effect of applied magnetic field on the putative lattice transition at $T^* = 203$ K. This observation warrants further investigation.

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