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# Thermal conductivity of the superconducting filled skutterudite compounds $\text{PrOs}_4\text{Sb}_{12}$ and $\text{PrRu}_4\text{Sb}_{12}$

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## Abstract

We report measurements of the thermal and charge conductivities of two single crystals of  $\text{PrOs}_4\text{Sb}_{12}$  and  $\text{PrRu}_4\text{Sb}_{12}$  over the temperature range of 1.2–30 K at zero magnetic field. For  $\text{PrRu}_4\text{Sb}_{12}$ , we find behaviour consistent with a simple metal and recover the Wiedemann–Franz law in the low temperature limit above  $T_c$ . For  $\text{PrOs}_4\text{Sb}_{12}$ , a simple model is used to separate out the electronic conductivity which shows an unusual temperature dependence upon entering the superconducting state. For both systems, the temperature dependence of the Lorenz number is shown to be consistent with scattering from  $\text{Pr}^{3+}$  ions in the presence of the crystalline electric field.

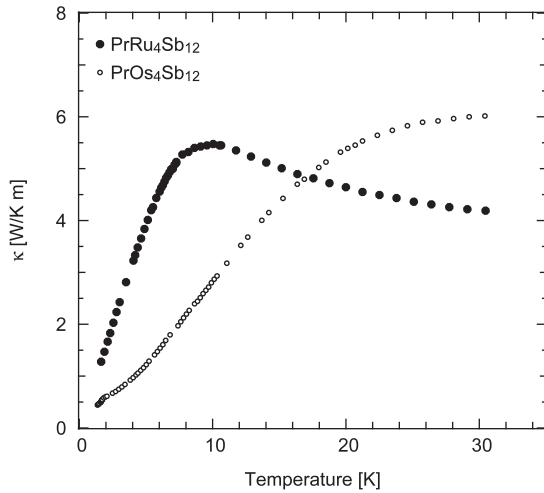
## 1. Introduction

The filled skutterudite compounds have attracted a great deal of attention from scientists during the past decade. In the lanthanide series of filled skutterudites, the crystal structure places a lanthanide ion inside an atomic cage of twelve nearest neighbour pnictogen ions. The hybridization of the localized f-states of the lanthanide ions with the ligand states of the pnictogen ions give rise to a variety of exotic correlated electron ground states.

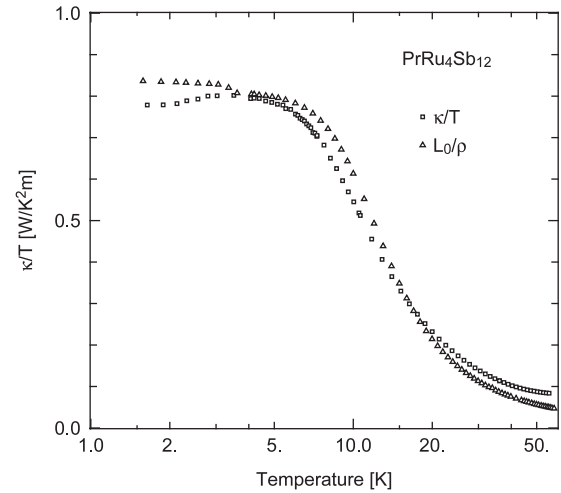
$\text{PrOs}_4\text{Sb}_{12}$  was found to be the first Pr-based heavy fermion superconductor with an effective mass,  $m^* \sim 50m_e$ , where  $m_e$  is the mass of a free electron [1]. Evidence for the heavy fermion state is mainly provided by the magnitude of the coefficient of the electronic specific heat,  $\gamma \sim 500 \text{ mJ mol}^{-1} \text{ K}^{-2}$  [2], suggesting strong correlations between electrons. The origin of this heavy fermion state might be either the interactions between the electric quadrupolar moments of  $\text{Pr}^{3+}$  ions and the charges of conduction electrons or the interaction between the magnetic moments of  $\text{Pr}^{3+}$  ions and spins of conduction electrons. The superconducting state in this compound that appears below  $T_c = 1.86 \text{ K}$  is most likely unconventional. Angle resolved magneto-thermal conductivity measurements [3] show two distinct superconducting phases

with different nodal structures. Also, specific heat measurements reveal a power law temperature dependence at low temperature in the superconducting state and two superconducting jumps at  $T_c$  [4, 5], although this feature is possibly sample dependent [6]. In contrast, penetration depth measurements based on  $\mu$ -SR experiments [7] and Sb nuclear quadrupole resonance measurements [8] are consistent with an isotropic superconducting gap. However, recent experiments suggest that the energy gap may undergo a transition from an isotropic gap to a gap with point nodes below a temperature  $T_{c3} \sim 0.6 \text{ K}$  [9, 11, 10, 12].

The isostructural compound,  $\text{PrRu}_4\text{Sb}_{12}$  has a smaller lattice constant and shows the properties of a BCS superconducting state below  $T_c \sim 1.1 \text{ K}$  [13]. The specific heat measurements shows a jump at  $T_c$  with an electronic coefficient of about  $\gamma = 59 \text{ mJ mol}^{-1} \text{ K}^{-2}$  [2, 15]. The  $\gamma$  value and the jump of specific heat at  $T_c$  yield the ratio  $\Delta C/\gamma T_c = 1.49$ , which is very close to 1.43 for conventional superconductors, indicative of weak coupling between the electrons mediated by phonons, most likely associated with an isotropic superconducting gap. Besides the specific heat measurements, measurements of the superfluid density exhibit exponential behaviour at low temperatures with an energy gap of the order of  $2\Delta = 3k_B T_c$  [16].



**Figure 1.** Thermal conductivity,  $\kappa$ , as a function of temperature for  $\text{PrRu}_4\text{Sb}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$  measured at temperatures below  $T = 30$  K.



**Figure 2.** Thermal conductivity of  $\text{PrRu}_4\text{Sb}_{12}$  divided by temperature,  $\frac{\kappa}{T}$ , and the electrical resistivity in thermal units,  $\frac{L_0}{\rho}$ , as a function of temperature.

In this paper, we present and discuss measurements of the thermal and charge conductivities of  $\text{PrRu}_4\text{Sb}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$  single crystal samples in the temperature range 1.2–40 K. The low residual resistivity of both samples,  $\rho_0 < 5 \mu\Omega \text{ cm}$ , and the sharp superconducting transition measured in  $\text{PrOs}_4\text{Sb}_{12}$  indicate the high purity level of these samples. As we will show, there is a strong contrast between the thermal transport of these systems.  $\text{PrRu}_4\text{Sb}_{12}$  is consistent with conventional metallic behaviour, while  $\text{PrOs}_4\text{Sb}_{12}$ , shows rather more complex temperature dependence consistent with a more strongly correlated electronic phase. The features in the heat transport of this compound will be explained based on a simple model for the separation between the lattice and electronic conductivity. We will also discuss the effects of the crystalline electric field on the electrical resistivity and Lorenz number.

## 2. Experimental details

Single crystal samples were grown using an Sb flux method. Further details of sample growth can be obtained from [17]. The samples were aligned using Laué x-ray backscattering and then were polished to cuboid shape with the long axis in the direction of the  $a$ -axis of the cubic crystal structure. The dimensions of the  $\text{PrRu}_4\text{Sb}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$  samples were  $2500 \times 190 \times 380 \mu\text{m}^3$  and  $1080 \times 185 \times 100 \mu\text{m}^3$  respectively.

Thermal conductivity was measured using a two-thermometer, one heater method. Four high purity silver wires were attached to the samples using indium solder. These wires are then attached to the heaters and thermometers and to thermal and electrical ground on the cryostat. The heat current is applied along the long axis of the samples and is therefore parallel with the  $a$ -axis of the crystal structure. The same four wires are also used for electrical resistivity measurements. The thermal conductivity mount used for these measurements was tested using a silver sample. The results of the thermal and charge conductivity measurements in silver were found

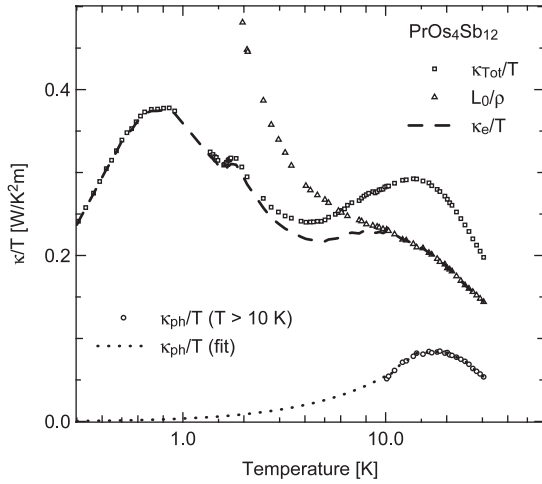
to agree with the Wiedemann–Franz law and recovered the Sommerfeld value of the Lorenz number to within 5%. All measurements were performed using a pumped  $^4\text{He}$  cryostat and in zero magnetic field.

## 3. Results and discussion

In figure 1, we present the results of our measurements of thermal conductivity below  $T = 30$  K on both materials  $\text{PrRu}_4\text{Sb}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$ . In comparison to other thermal conductivity measurements on filled skutterudite systems where the research was aimed at exploring the lattice conductivity [14], the values we obtain are typically an order of magnitude larger. As we shall show in the more detailed analysis to follow, the difference is due to a large electronic conductivity in these stoichiometric single crystal samples with low residual resistivities ( $< 10 \mu\Omega \text{ cm}$ ). In this temperature range, the electronic conductivity obscures the putative reduced phonon conductivity that results from the rattling of the rare-earth ion inside the pnicto-gen cage.

### 3.1. $\text{PrRu}_4\text{Sb}_{12}$

Figure 2 shows the temperature dependence of the thermal conductivity,  $\kappa/T$ , and the charge conductivity (in thermal units),  $L_0/\rho$ , of  $\text{PrRu}_4\text{Sb}_{12}$ , where  $L_0 = 2.44 \times 10^{-8} \Omega \text{ W K}^{-2}$  is the Sommerfeld value of the Lorenz number and  $\rho$  is the resistivity. The temperature dependence of the thermal conductivity is characteristic of a regular metallic compound. At low temperatures (below  $T \sim 5$  K), the dependence is linear in temperature ( $\kappa/T$  is constant in temperature) consistent with temperature independent elastic scattering of electrons by impurities. As the temperature is raised, inelastic scattering increases and the conductivity decreases. We explore what can be said about these inelastic processes using the Wiedemann–Franz law later in this paper. For now, we note that we recover the Sommerfeld value of the Lorenz number to within 5%



**Figure 3.** Thermal conductivity divided by the temperature and the electrical resistivity in thermal units of  $\text{PrOs}_4\text{Sb}_{12}$ . The data for temperatures below  $T = 1.2$  K are taken from [24]. Based on the model outlined in the main text, the dotted and dashed lines show the contributions due to phonons,  $\frac{\kappa_{\text{ph}}}{T}$ , and electrons,  $\frac{\kappa_{\text{e}}}{T}$ , respectively.

below 5 K (see figure 5), consistent with conventional metallic behaviour. The electrical resistivity is quantitatively consistent with that reported earlier [13]. The residual resistivity is measured to be  $3 \mu\Omega \text{ cm}$  demonstrating the high quality of these single crystal samples. Since the samples are from the same source as those measured and reported by Frederick *et al* [17], where the high resistivity of the  $\text{PrRu}_4\text{Sb}_{12}$  were reported as being anomalously high for a stoichiometric compound, this perhaps demonstrates that the earlier result was related to extrinsic issues that are not encountered here. The small anomaly at  $T \sim 4$  K is related to temperature control issues at the boiling point of helium in our pumped helium-4 cryostat.

### 3.2. $\text{PrOs}_4\text{Sb}_{12}$

In figure 3 we show  $\kappa/T$  and  $L_0/\rho$  for  $\text{PrOs}_4\text{Sb}_{12}$ . The data for temperatures below  $T = 1.2$  K are taken from measurements performed on the same single crystal sample in a dilution refrigerator and are reported in detail elsewhere [24]. They are included to allow for a more detailed discussion of the behaviour of the thermal conductivity immediately below  $T_c$ . In contrast to the data for  $\text{PrRu}_4\text{Sb}_{12}$ , the rich structure is evidence of more complex behaviour than is seen in a simple metal. In order to investigate these features in the thermal conductivity, it is necessary to separate the contribution to the total conductivity from electrons and phonons. In metallic compounds, the total thermal conductivity can be expressed as the sum of the contributions from the lattice and from conduction electrons:  $\kappa_{\text{tot}} = \kappa_{\text{e}} + \kappa_{\text{ph}}$ , where  $\kappa_{\text{e}}$  and  $\kappa_{\text{ph}}$  are the electronic and lattice thermal conductivity. Our goal is to establish a reasonable approximation for the phonon conductivity that will allow us to study the electronic contribution more closely.

The lattice contribution to the thermal conductivity can be estimated in the following way. We assume that the Wiedemann–Franz law is satisfied at temperatures above

$T = 10$  K, and so the electronic contribution,  $\kappa_{\text{e}}/T = L_0/\rho$ . Subtracting this from the total conductivity reveals the phonon conductivity above 10 K,

$$\kappa_{\text{ph}}/T (T > 10 \text{ K}) = \kappa_{\text{tot}}/T - L_0/\rho. \quad (1)$$

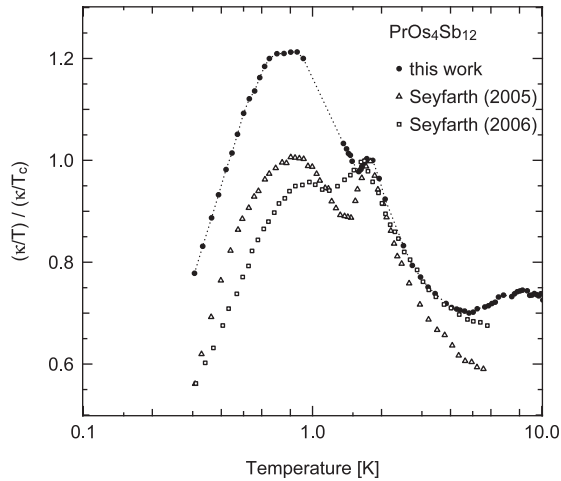
Based on this construction the peak in the conductivity at  $T \sim 13$  K is mostly due to phonons. For temperatures in this range, the most important scattering mechanisms involved in  $\kappa_{\text{ph}}$  are the Umklapp process and the phonon–electron collisions. Assuming a form of  $T^2 \exp(-\frac{\theta_{\text{D}}}{T})$  ( $T < \theta_{\text{D}}$ ), for the lattice conductivity affected by Umklapp processes [18], where  $\theta_{\text{D}}$  is the Debye temperature, and a  $T^2$  form for phonons scattered by electrons, we can fit the phonon conductivity above 10 K to the expression

$$\frac{\kappa_{\text{ph}}}{T} = \left( \alpha \frac{\exp(-\frac{\theta_{\text{D}}}{T})}{T} + \frac{\beta}{T} \right)^{-1} \quad (2)$$

where  $\alpha$  and  $\beta$  are fitting parameters. Using the Debye temperature which is reported to be 164 K for  $\text{PrOs}_4\text{Sb}_{12}$  [4], the fitting gives  $\alpha = 5180 \text{ K}^3 \text{ m W}^{-1}$  and  $\beta = 170 \text{ K}^3 \text{ m W}^{-1}$ . This is plotted as the dotted line in figure 3. The fit has been extrapolated to temperatures below 10 K where the Wiedemann–Franz law does not provide any knowledge of the electronic contribution. As can be seen in the figure, the resulting temperature dependence for the phonon contribution has a peak at  $T = 17$  K. On the low temperature side of the peak, the behaviour is governed primarily by the coefficient of the phonon–electron scattering term,  $\beta$ . From our fit, the value we obtain is consistent with the upper values obtained for pure metallic samples. For example, in niobium the value is  $380 \text{ K}^3 \text{ m W}^{-1}$  [19]. According to this scale, it would appear that phonon–electron coupling in this material is relatively strong as might be expected in a material with a structure renown for its thermoelectric properties. Furthermore, the absence of such a peak in  $\text{PrRu}_4\text{Sb}_{12}$  suggests that phonon–electron coupling in this material must be even larger. This line of reasoning is consistent with the observation of electron–phonon mediated superconductivity in  $\text{PrRu}_4\text{Sb}_{12}$ , and the possibility of a more exotic mechanism in  $\text{PrOs}_4\text{Sb}_{12}$ .

On the high temperature side of the peak, the behaviour is governed by Umklapp scattering. We note that a  $1/T$  temperature dependence also produces a reasonable fit. A crossover to this kind of temperature dependence is expected as one moves closer to the Debye temperature [18]. Consequently, it is difficult to conclude very much from this part of the model.

In the low temperature limit, well below 10 K, boundary scattering of phonons needs to be considered in addition to the electronic term discussed above. We estimate this using the kinetic formula,  $\kappa_{\text{ph}} = c_v l v / 3$ , where we use an approximate sound velocity  $v = 2000 \text{ m s}^{-1}$  and the specific heat  $c_v = 3.95 T^3 \text{ mJ mol}^{-1} \text{ K}^{-4}$  [4] and  $l$  is a characteristic crystal dimension  $\sim 100 \mu\text{m}$ . Hence, the lattice thermal conductivity due to scattering by the boundaries of the sample can be written as  $\frac{\kappa_{\text{ph}}}{T} = 8 T^2 \text{ mW K}^{-2} \text{ m}^{-1}$ . Nevertheless, even with this term included, the contribution due to phonons at or around



**Figure 4.** Thermal conductivity divided by temperature and normalized to the value at  $T_c$  for three samples of  $\text{PrOs}_4\text{Sb}_{12}$  with different purity levels. Closed circles are from this work, open triangles are from [20] and open squares are from [6].

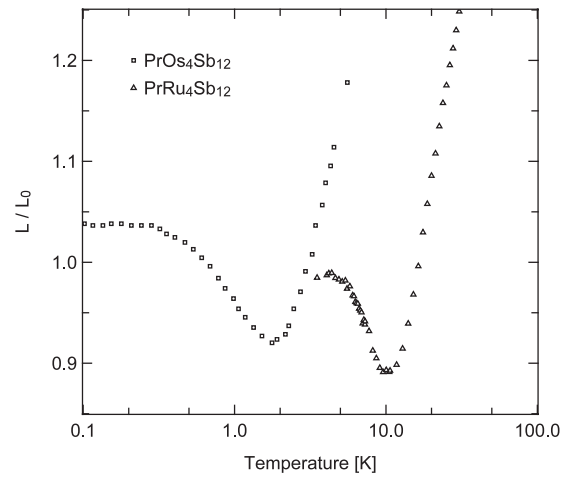
$T_c$  remains a very small fraction of the total conductivity, and therefore the vast majority is attributed to electrons.

Overall, this model provides a reasonable explanation for the peak in the thermal conductivity above  $T = 10$  K being attributed to phonons. Quantitatively, we note that our assumption that the Wiedemann–Franz law is satisfied means that we estimate a minimum phonon conductivity. However, based on the value of the Lorenz number measured by Seyfarth *et al* [20] (see figure 5) which approaches the Sommerfeld value around 10 K, this assumption seems justified.

The electronic thermal conductivity can be obtained by subtracting our estimated phonon contribution from the total conductivity. This is plotted as the dashed line in figure 3. By construction this electronic contribution agrees with the charge conductivity (in thermal units) above  $T = 10$  K.

As the temperature is reduced towards  $T_c$ , the electronic conductivity increases. This increase occurs at approximately the same temperature range that a ‘roll-off’ feature is observed in the electrical resistivity. This feature has been shown to result from loss of scattering of the conduction electrons by Pr ions in the crystalline electric field (CEF) [21]. When the temperature reaches  $T_c = 1.81$  K, the electronic conductivity initially decreases but then hits a minimum at  $T \sim 1.2$  K, and increases again to a maximum at 0.6 K. This behaviour results in two further peaks which we now discuss.

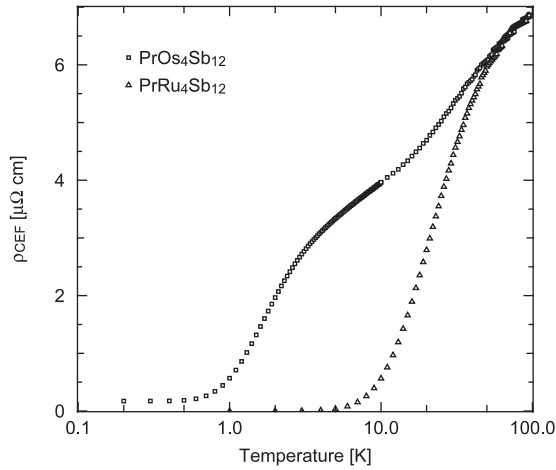
As the temperature is reduced below  $T_c$ , two mechanisms control the changes in  $\kappa_e/T$ ; the number of heat carriers and the scattering rate. As electrons condense, they no longer contribute to the thermal conductivity. If there is no change in the scattering rate, the thermal conductivity will decrease. This is what is observed in conventional s-wave superconductors, for example in aluminium [22]. However, if the dominant scattering mechanism is electronic, then as electrons condense, the scattering rate will also decrease. The thermal conductivity that results will depend upon which effect dominates; the loss of carriers or the loss of scattering. In high- $T_c$  superconductors, the loss of scattering dominates and results in an increase in



**Figure 5.** Temperature dependence of the Lorenz ratio of  $\text{PrRu}_4\text{Sb}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$ . Data for the latter are taken from [20].

the thermal conductivity below  $T_c$  [23]. To our knowledge, all superconductors measured so far show either an increase or a decrease in electronic thermal conductivity, but never a decrease followed by an increase. A peak well below  $T_c$  can also be attributed to phonons whose scattering rate decreases with the condensation of electrons. In this case the magnitude of the conductivity would appear to rule out the possibility that this peak results from phonons. In the absence of any electronic scattering of phonons the maximum conductivity is limited by scattering from the boundaries of the sample. As estimated earlier, this conductivity can be at most  $8 \text{ mW K}^{-2} \text{ m}^{-1}$  at  $T = 1$  K which is only 2% of the total value.

Further evidence for the electronic origin of this large peak below  $T_c$  is found in figure 4. This plot shows the normalized electronic thermal conductivity divided by the temperature for the sample from this measurement and two other samples [20, 6]. For a peak that results from a loss of electronic scattering, the conductivity will rise until capped by another scattering mechanism namely impurity scattering. In this case the peak height will be related to the level of impurity scattering. A theory to explain this observation in high- $T_c$  cuprates was developed by Hirshfeld and Putikka [25]. Although no detailed calculation has been performed in this case, qualitatively we find that the peak below 1 K does vary with the purity level of the sample in an appropriate way. Basing the purity level on the magnitude of the normal state conductivity, we find that the higher the normal state conductivity, the more pure the sample and hence the higher the peak. The values for the residual normal state conductivity measured by suppressing superconductivity with a magnetic field are 8, 2.3, 2.0  $\text{mW K}^{-2} \text{ cm}^{-1}$  for this sample [24, 6] and [20] respectively. In detail, this theory only allows for an increase in the thermal conductivity below  $T_c$ , so the initial decrease observed in this case remains an outstanding issue. It is possible that the change in behaviour is related to the double transition observed for example in specific heat [5] and the possibility of multiple superconducting phases. Although we have not measured the specific heat of this particular sample, when compared to the separation in temperature of the



**Figure 6.** The contribution to the electrical resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  and  $\text{PrRu}_4\text{Sb}_{12}$  caused by the scattering of conduction electrons by  $\text{Pr}^{3+}$  ions in the presence of the crystalline electric field. The scattering increases appreciably at  $T \sim 0.6$  K for  $\text{PrOs}_4\text{Sb}_{12}$  and  $T \sim 6$  K for  $\text{PrRu}_4\text{Sb}_{12}$ .

two jumps in specific heat which is  $\sim 50$  mK [5], the difference in temperature between the peak (at  $T_c$ ) and the minimum in the thermal conductivity is  $\sim 250$  mK and therefore much larger.

### 3.3. Temperature dependence of Lorenz number

The normalized Lorenz number as a function of temperature for both compounds has been plotted in figure 6. The data for  $\text{PrOs}_4\text{Sb}_{12}$  is for the field-induced normal state and is taken from reference [20]. The data for  $\text{PrRu}_4\text{Sb}_{12}$  is shown for temperatures above  $T_c$ . At low temperatures both compounds show a plateau at a value close to the Sommerfeld value of the Lorenz number ( $L_0$ ) consistent with the Wiedemann–Franz law. The temperature dependence for both systems is qualitatively similar. As the temperature is increased  $\frac{L}{L_0}$  decreases and results in a minimum before the Lorenz number rises above 1 due to the presence of a non-negligible phonon conductivity at high temperatures. The decrease from  $L_0$  occurs at  $T \sim 0.6$  K in  $\text{PrOs}_4\text{Sb}_{12}$  resulting in a minimum at  $T \sim 1.6$  K. For  $\text{PrRu}_4\text{Sb}_{12}$  the decrease starts at  $T \sim 6$  K with the minimum at  $T \sim 12$  K. Such a reduction occurs due to the increase of a scattering mechanism that scatters heat more effectively than charge. In simple metallic systems this is usually scattering due to phonons. If the minimum is due to the effect of phonons, it should reflect the Debye temperature which is 232 K for  $\text{PrRu}_4\text{Sb}_{12}$  and 186 K for  $\text{PrOs}_4\text{Sb}_{12}$  [2]. Thus, both the low temperatures of these minima and the order of magnitude difference in the temperatures at which they occur are not consistent with this idea. Instead, we suggest that the origin in each case lies in the scattering associated with the splitting of the  $\text{Pr}^{3+}$  states due to the crystalline electric field (CEF). This type of scattering, either magnetic exchange or aspherical coulomb scattering or both, has been used to explain the temperature dependence of the electrical resistivity in both  $\text{PrOs}_4\text{Sb}_{12}$  [21] and  $\text{PrRu}_4\text{Sb}_{12}$  [13]. In each case the interest was in the temperature at which the scattering started to decrease as the thermal energy ( $k_B T$ ) dropped below the

splitting between the ground and first excited state. Here, we are interested in the temperature at which the scattering starts to increase and could, in principle, lead to a greater reduction in thermal over charge conductivity and give rise to a decrease in the Lorenz ratio. We repeat the calculations used for  $\text{PrOs}_4\text{Sb}_{12}$  [21]<sup>3</sup> and  $\text{PrRu}_4\text{Sb}_{12}$  [13]<sup>4</sup>, and have plotted the contribution to the resistivity from this crystalline electric field scattering in figure 6. For  $\text{PrOs}_4\text{Sb}_{12}$ , both the magnetic and electric interactions between the charge and spin of electrons and magnetic and quadrupolar moments of  $\text{Pr}^{3+}$  have been taken into account, whereas for  $\text{PrRu}_4\text{Sb}_{12}$  just the magnetic interaction has been considered in the calculation of CEF effects. The ‘roll-off’ feature can be seen in the electrical resistivity of both compounds at temperatures which are an order of magnitude different,  $T \sim 8$  K for  $\text{PrOs}_4\text{Sb}_{12}$  and  $T \sim 70$  K for  $\text{PrRu}_4\text{Sb}_{12}$ . Concentrating on the low temperature dependence, we note that in  $\text{PrOs}_4\text{Sb}_{12}$  the scattering picks up at  $T \sim 0.6$  K, while in  $\text{PrRu}_4\text{Sb}_{12}$  it does so at  $T \sim 6$  K. Both temperatures are exactly consistent with the point at which the Lorenz number begins to decrease from 1, which is quantitatively consistent with our explanation.

## 4. Conclusion

In conclusion, we have measured that charge and heat transport properties of two superconducting filled skutterudite materials,  $\text{PrRu}_4\text{Sb}_{12}$  and  $\text{PrOs}_4\text{Sb}_{12}$ . In  $\text{PrRu}_4\text{Sb}_{12}$  we find the properties are consistent with a conventional metallic system and recover the Sommerfeld value of the Lorenz number in the low temperature limit above  $T_c$ . The temperature dependence of the thermal transport in  $\text{PrOs}_4\text{Sb}_{12}$  is more complex and a simple analysis to separate electronic and phononic contributions suggests that phonon–electron coupling in this system is weaker than in  $\text{PrRu}_4\text{Sb}_{12}$ . At our lowest temperatures, we can access the superconducting state in  $\text{PrOs}_4\text{Sb}_{12}$ . Here too we find exotic temperature dependencies with two peaks in the electronic thermal transport; one at  $T_c$  and another at  $\sim 0.3T_c$ . Current theoretical models are not able to account for two peaks, although we speculate that the behaviour may be related to the double jump observed in the specific heat [4, 5]. Finally, we explore the temperature dependence of the Lorenz number,  $L$ , in these two systems. A decrease in  $L$  from the Sommerfeld value as the temperature is increased is attributed to the onset of scattering from  $\text{Pr}^{3+}$  moments in the presence of the crystalline electric field.

## Acknowledgments

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<sup>3</sup> Taking parameters from [21], in  $\text{PrOs}_4\text{Sb}_{12}$  a mixture of magnetic and Coulomb scattering is modelled with a ratio  $r = 0.46$ , for a  $\Gamma_5$  triplet first excited state lying 6 K above the  $\Gamma_3$  doublet ground state. The CEF parameter is  $x = -0.7225$ .

<sup>4</sup> Taking parameters from [13], in  $\text{PrRu}_4\text{Sb}_{12}$  only magnetic scattering is modelled for a  $\Gamma_4$  triplet first excited state lying 70 K above the  $\Gamma_1$  singlet groundstate. The CEF parameter is  $x = -0.7$ .

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