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LETTER TO THE EDITOR

Very low temperature thermal conductivity in the layered perovskite superconductor Sr_2RuO_4

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Abstract. We present thermal conductivity measurements on the layered perovskite superconductor Sr_2RuO_4 down to very low temperatures (20 mK). Our data show a large residual term in κ/T as $T \rightarrow 0$ K which depends on the sample quality. We do not observe any sign of the predicted crossover at low temperatures or of a second phase transition at least above 20 mK.

The recent discovery of superconductivity in the layered perovskite Sr_2RuO_4 [1] has led to intensive theoretical and experimental work. Up to now, Sr_2RuO_4 is the only known layered perovskite *without* copper–oxygen planes which is a superconductor. The understanding of the superconductivity in this compound could lead to advances in the search for an explanation of the properties of high T_c superconductors, which are mostly layered perovskites *with* copper–oxygen planes.

There are important differences in the physical properties of the high T_c perovskites and this compound. In Sr_2RuO_4, the normal phase properties follow clearly the behaviour expected for a strongly anisotropic Fermi liquid at low temperatures. For example the resistivity follows the law $\rho = \rho_0 + AT^2$ for $T < 20$ K and is at least 400 times larger for the current perpendicular to the plane than for the current in the plane [2]. The Fermi surface is now completely determined by de Haas–van Alphen measurements and its form well reproduced by band structure calculations [3]. These measurements, as well as specific heat [4] and NMR measurements [5], show a renormalization of the effective masses by a factor of 3–4, whose origin is still controversial. The determination of the whole Fermi surface was possible due to the metallic nature of the ground state of this compound and the high quality of the available single crystals which can have residual resistivities lower than 1 $\mu\Omega$ cm [2].

Superconductivity appears below 1 K, but in spite of the good quality of the samples, the superconducting properties remain unclear. First specific heat (down to $T_c/3$ [4]) and NQR (down to $0.15T_c$ [5]) measurements revealed an anomalously large density of states in the superconducting phase at low temperature and triggered theoretical studies [8] about

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the possibility of an unconventional, *p or d wave*, superconducting order parameter in this compound. More recently, Mackenzie *et al* [6] measured the dependence of the critical temperature on the sample quality, and found a strong suppression of superconductivity by *non-magnetic* impurities, in favour of an unconventional, non-s-wave, order parameter. In this paper we present thermal conductivity measurements on samples similar to those of Mackenzie *et al* [6] down to very low temperatures (20 mK, $0.03T_c$). Our measurements give the first experimental data sensitive to the density of states in the superconducting state of Sr_2RuO_4 down to such low temperatures.

We measured the resistivity of four samples, which were thin single-crystalline slices as described in [6]. The contacts to the samples are also described in [6] and are ohmic with a resistance of the order of $R_{contact} \sim 10$ mΩ at very low temperatures, which is a rather low value and makes the measurement of the resistivity easy by standard AC techniques. Nevertheless, the contact resistance $R_{contact}$ is not sufficiently low, or equivalently the thermal conduction of the contact not sufficiently high, to make thermal conductivity measurements easily.

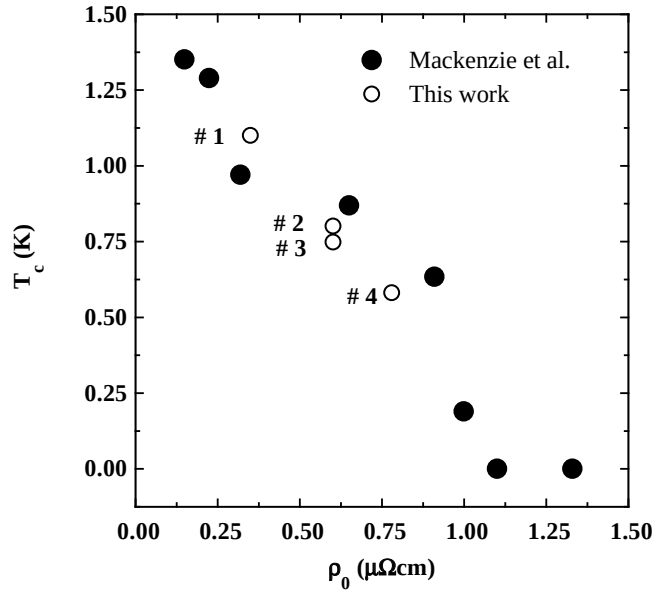

Figure 1. The residual resistivity as a function of the critical temperature, compared to the results of [6].

The very low residual resistivity of the samples, unusual for layered perovskites, leads to a very low resistance and therefore to very *high* values of the thermal conductivity of the samples, in contrast to the *low* value of the thermal conduction *between* the sample and the cryostat (given by $R_{contact}$). This means that upon applying power to one end of the sample the temperature gradient is greatest *between* the sample and the cryostat, and not *within* the sample. In sample 1 (see figure 1), $\kappa_{sample}/\kappa_{sample-cryostat} \gg 100$, that is, the temperature gradient is at least two orders of magnitude larger between the cryostat and the sample, than within the sample. In sample 4, the situation was better, due to a higher residual resistivity, but the measurement was still not possible. In samples 2 and 3, the geometry was more favourable (that is, longer samples with a smaller cross section) and the residual resistivity larger, which implies $\kappa_{sample}/\kappa_{sample-cryostat} \gg 10$. We could then perform the

measurement using standard techniques described in [9]. Note that, unfortunately, it was not possible to measure the samples with the lowest resistivities and the highest critical temperatures. The way to solve this problem would be to make contacts of even lower resistance $R_{contact} \ll 10$ mΩ, or to measure samples of higher resistance, ideally very long needles, but both points are non-trivial and need further experimental work.

In the normal phase of Sr_2RuO_4, and below 1 K, the resistivity is controlled by elastic scattering between electrons and defects or impurities ($\rho = \rho_0$). Inelastic collisions which give the Fermi liquid AT^2 term in the resistivity are important at higher temperatures and can be neglected below 1 K. Before discussing the thermal conductivity measurements, we note that we confirm the relation between the residual resistivity and the critical temperature found by Mackenzie *et al* [6] on the same type of sample. In figure 1 we plot the critical temperature as a function of the residual resistivity ρ_0 compared to the data of Mackenzie *et al* [6]. It is instructive to note that similar behaviours i.e. superconductivity which is strongly reduced with disorder, were found long ago in classical, s-wave superconductors, such as the A15 family (e.g. T_c of Nb_3Ge drops from 22 K to about 5 K as ρ_0 increases from 40 $\mu\Omega$ cm to 130 $\mu\Omega$ cm) [7]. Several explanations were given within classical s-wave superconductivity which were related to changes in the normal state properties as a function of disorder; nevertheless this type of theory cannot be applied in a simple way to Sr_2RuO_4 as the physics of this compound differs strongly from that of A15 superconductors. Indeed, even an extremely small amount of disorder is sufficient to suppress completely superconductivity in Sr_2RuO_4 (a residual resistivity of about ~ 1 $\mu\Omega$ cm corresponding to a rather large electronic mean free path of 500 Å, compared to several hundred $\mu\Omega$ cm in A15 compounds) and no significant changes in the normal state density of states are observed as a function of purity. Therefore, it is reasonable to think of another scenario to explain the results of figure 1, such as an unconventional (d- or p-wave) order parameter. By contrast to usual s-wave superconductors, where non-magnetic impurities lead only to a very small reduction of the critical temperature, even small amounts of non-magnetic impurities act as pair breakers in unconventional superconductors, and reduce considerably T_c (see e.g. [11]). Based on susceptibility measurements and x-ray analysis, Mackenzie *et al* show that the impurities in these samples are non-magnetic, so the strong reduction of T_c with increasing impurity concentration (proportional to the residual resistivity) shown in figure 1 favours an unconventional order parameter. Nevertheless, the dependence of the critical temperature on the residual resistivity (or defect concentration) gives a limited amount of information, as it does not show the density of states which is expected to appear in the case of strong pair breaking. It is only the hallmark of a vanishing average of the order parameter on the Fermi surface [12].

Our thermal conductivity measurements (on samples 2 and 4, see also figure 1) are shown in figure 2 plotted as κ/T as a function of T. In the normal phase, below 1 K, $\kappa \sim T$ and is larger for the sample with the lowest residual resistivity, as expected in normal metals. In the superconducting phase, below T_c, κ/T drops strongly and has a large zero temperature extrapolation in both samples. We first discuss the linear temperature behaviour of κ we find in the normal phase.

This is expected of the thermal conductivity of simple metals at very low temperatures, where phonons are negligible and the thermal transport is driven by electrons scattered elastically by impurities ($\rho = \rho_0$). In this case, the Wiedemann–Franz law is expected to be valid, that is, the Lorentz number $L = \kappa/T\rho$ is expected to be close to $L_0 = 2.44 \times 10^{-8}$ W Ω K^{-2}. In figure 3, we plot the Lorentz number L, normalized by L_0, as a function of temperature. We find clearly a value slightly higher (by 20%) than L_0, which could mean that a small part of the thermal conductivity is due to some other heat conduction

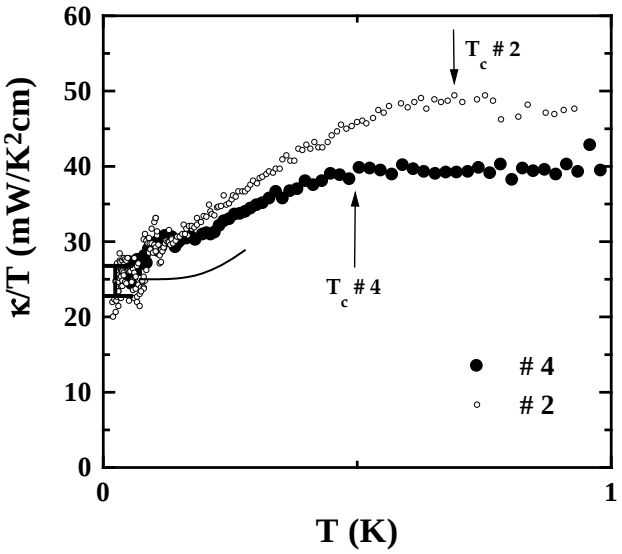

Figure 2. The thermal conductivity κ/T as a function of temperature for two different samples (see also figure 1 for the numbering). The thermal conductivity of the normal phase is clearly proportional to the temperature, as expected for simple metals. In the superconducting phase, κ/T drops and shows large values. The arrows show the critical temperatures observed by resistivity (see figure 1 for the numbers) and the line shows an exponential dependence which is clearly not followed by our data. The error bars of the measurements are given by the scatter of the data and are much more important below 100 mK, as shown by the bar in the figure.

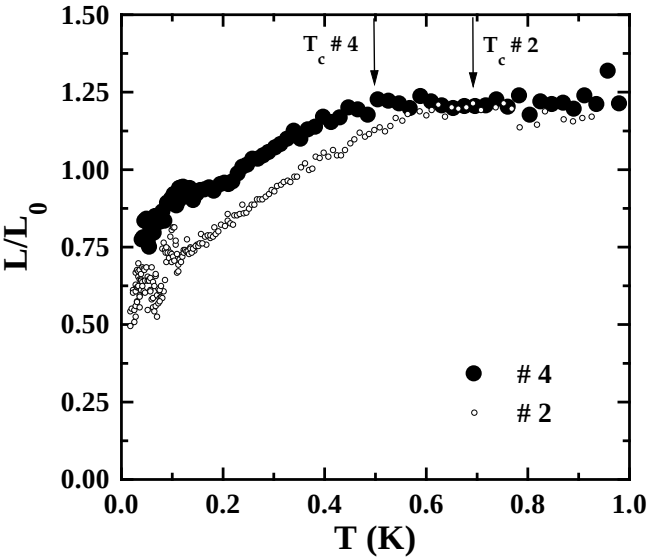

Figure 3. The Lorentz number $L = \kappa\rho/T$ as a function of temperature for the measured samples. The arrows show the critical temperatures observed by resistivity (see figure 1 for the numbers).

mechanism, such as phonons. Note nevertheless that we find roughly the same Lorentz number for both measured samples, which have significantly different residual resistivities. That is, the dependence on the impurity concentration of κ/T in the normal phase is the same as that of the residual resistivity ρ_0: it cancels out when plotting the Lorentz number $L = \kappa\rho/T$. In the case of a small contribution to κ of other, non-electronic, thermal conduction mechanisms such as phonons, this would not be the case. We can therefore rule out an important contribution of phonons to the thermal conductivity in the normal phase of this compound below 1 K.

In the superconducting phase, the thermal conductivity drops, as expected in all superconductors. Nevertheless, we find roughly $\kappa \sim aT + bT^2$, which is far from the behaviour of pure s-wave superconductors ($\kappa/T \propto \mathrm{e}^{-\Delta/kT}$).

The large zero temperature extrapolation of κ/T is clearly related to the pair breaking effect of defects or impurities, as it depends strongly on the sample quality. The very low temperature we could reach makes it possible to obtain reliable values of the residual term $\kappa/T(T \rightarrow 0\ \mathrm{K})$, by contrast to earlier work. We clearly observe (figures 2 and 3) that the better the sample and the higher the critical temperature, the lower the residual term observed by thermal conductivity, as expected in the case of a strong pair breaking effect of impurities. This result can be compared to several theoretical predictions. Machida *et al* [8] predict a triplet superconducting pairing function, and an intrinsic residual density of states of roughly half the value of the normal phase, in analogy to the superfluid phase A1 of ^{3}He under magnetic fields, in which only half of the excitations are gapped. By contrast, our measurements indicate that the large residual density of states shown by the experiments is, to a large extent, due to the pair breaking effect of apparently non-magnetic impurities. Nevertheless we cannot definitely exclude the theory of Machida *et al*, as, in pure samples, the residual density of states might still stay large. Another theory, due to Agterberg *et al* [8], predicts an exotic type of triplet superconductivity with two, well developed gaps associated with different sheets of the Fermi surface. These authors predict high values of the thermal conductivity down to $0.1T_c$, and a crossover to an activated behaviour at lower temperatures. We do not observe such a behaviour in the measured samples, even at the lowest temperatures ($0.03T_c$), which is against the theory of Agterberg *et al*. Other models note the possibility of different phase transitions within the superconducting state, in analogy with the different superconducting phases found in the heavy fermion superconductor UPt_3, but we have not observed such phase transitions in this compound. We note nevertheless that the pair breaking effect of impurities might destroy the crossover behaviour predicted by Agterberg *et al*, as well as evidence for different phase transitions, and cannot therefore rule out completely these possibilities.

In order to discuss the bT^2 term in κ we need to remember early theoretical work on thermal conductivity of unconventional superconductors (see e.g. [10]). These authors found that the scattering phase shift δ is an important parameter to understand $\kappa(T)$ in unconventional superconductors. If the scattering is in the unitary limit ($\delta = \pi/2$), $\kappa(T)$ is quantitatively related to the density of states and shows power laws depending on the position and form of the line or point nodes of the superconducting gap, but if the scattering phase shift is near the Born limit $\delta = 0$ no clear predictions can be made. In clean samples, the observation of a power law at low temperatures in κ is a strong indication for zeros in the superconducting gap and scattering with δ near $\pi/2$ [10, 13]. But in samples with a large amount of impurity scattering, such as the ones we have measured, we cannot relate the observed law to the superconducting gap structure without a careful analysis of impurities. This situation is similar to that of the first thermal conductivity experiments on 'poor quality' crystals of the superconductor UPt_3, where similar behaviours were observed.

But later on, further measurements on high quality crystals showed a T^3 power law over a large temperature range dominated by a gap structure with a line of zeros [13].

In conclusion, the pair breaking effect of non-magnetic impurities is, at least partly, responsible for the large values of the residual term found in the thermodynamic and transport measurements sensitive to the density of states. As shown by the authors of [6], the impurities in our samples are non-magnetic. Therefore, the large amount of gapless excitation and its dependence on the impurity concentration is also in favour of an unconventional order parameter. We do not find signs for a crossover behaviour, nor for a second phase transition down to 20 mK. Although our thermal conductivity data do not give more precise indications on the nature of the order parameter, we give reliable information about the dependence of the residual density of states in the superconducting state on the impurity concentration in this compound. Future theoretical developments on the pair breaking effect of non-magnetic impurities under the models advanced by the authors of [8] will be useful. Concerning the experimental developments, more measurements in cleaner samples are necessary to give conclusions about the symmetry of the order parameter in this compound.

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