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Quasi-particle density in Sr_2RuO_4 probed by means of the phonon thermal conductivity

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Abstract

The thermal conductivity of Sr_2RuO_4 along the least conducting direction perpendicular to the RuO_2 plane has been studied down to 0.3 K. In this configuration the phonons remain the dominant heat carriers down to the lowest temperature, and their conductivity in the normal state is determined by the scattering on conduction electrons. We show that the phonon mean free path in the superconducting state is sensitive to the density of the quasi-particles in the bulk. An unusual magnetic field dependence of the phonon thermal conductivity is ascribed to the anisotropic superconducting gap structure in Sr_2RuO_4 .

1. Introduction

Sr_2RuO_4 first attracted a lot of interest as a superconductor which has the layered perovskite structure without copper [1]. Later on it has become clear that despite the crystal structural resemblance to the high- T_c cuprate superconductor $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, the electronic properties of these two systems are quite different [2]. The normal state of Sr_2RuO_4 is well described by Landau Fermi-liquid model [3], and its superconductivity is characterized by the spin-triplet pairing and spontaneous breaking of the time-reversal symmetry [4–7].

Study of the thermal conductivity can be very useful for probing the quasi-particle spectrum in the superconducting state [8]. These measurements played an essential role in locating the position of the superconducting gap nodes in high- T_c cuprates [9, 10] and in heavy-fermion superconductors [11, 12]. Of note, in these systems the thermal conductivity is mostly determined by quasi-particle excitations and is electronic in origin, as confirmed in the cuprates by measurements of the Righi–Leduc effect (thermal Hall effect) [13]. Detailed experimental

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characterization of the electronic thermal conductivity stimulated intensive theoretical studies on the subject [14–17] and at present there is a reasonable understanding of the *electronic* thermal conductivity in unconventional superconductors. The thermal conductivity of Sr_2RuO_4 with the heat flow along the conducting plane was also shown to be electronic in origin [18, 19], and it is well described with a model for unconventional superconductors with line nodes in the superconducting gap [19–21].

It is significant that Sr_2RuO_4 has a quasi-two-dimensional electronic structure [3, 22] and, consequently, all of the electronic transport properties should be strongly anisotropic. Therefore it is of interest to extend the measurements to the configuration with the heat flow perpendicular to the plane. Here the low electronic conductivity provides a rare opportunity to study the *phonon* thermal conductivity in unconventional superconductors.

In this paper, we report measurements and analysis of the thermal conductivity along the least conducting direction (the [001] crystal axis) in the normal, superconducting and mixed states of Sr_2RuO_4 . Through the analysis of the thermal conductivity in magnetic fields and the comparison of the results for the heat flow directions parallel and perpendicular to the plane, we separated the phononic and electronic contributions and characterized their field and temperature dependences.

We revealed that the temperature and field dependence of the phonon thermal conductivity reflects an increased quasi-particle density in the bulk of unconventional superconductors. A sublinear field dependence of the phonon thermal resistivity is ascribed to the presence of nodes in the superconducting gap, which is consistent with other relevant experiments; specific heat [23], NMR [24], magnetic penetration depth [25] and ultrasonic attenuation [26].

2. Experimental methods

The single crystals of Sr_2RuO_4 used in this study were grown in an infrared image furnace by the floating zone method [27]. In the crystal growth procedure, an excess of Ru is charged to create the Ru self-flux in order to compensate for Ru metal evaporation from the surface of the crystal rod. Therefore, inclusions of Ru metal are frequently found in the central part of the crystal boule. Since these inclusions are responsible for the formation of the so-called 3K-phase [28], to obtain single-phase samples we cut them from the surface part of the boule. The absence of 3K-phase was checked by AC susceptibility measurements. We selected a high-quality sample with $T_c = 1.42$ K, close to the extrapolated impurity-free value $T_{c0} = 1.50$ K [29, 30]. The sample was cut into a rectangular slab elongated in the [001] direction with dimensions $1.5 \times 0.5 \times 0.5$ mm³.

The thermal conductivity was measured by the usual one-heater–two-thermometers method with three 2 k Ω RuO_2 resistance chips [32]. The direction of the heat current was set along the [001] crystal axis perpendicular to the conducting plane. (Below, for the thermal conductivity perpendicular to the plane, we use the notation κ_{001} , with the subscript showing the direction of the heat flow.) The end of the sample was attached directly to the cold finger of a miniature vacuum cell [33]. The cell was placed in a double-axis sample rotator in a ³He refrigerator inserted into a 17 T superconducting magnet. The rotator enabled precise cell orientation with a relative accuracy of better than 0.1°. Simultaneous electrical conductivity measurements were performed using the same thermal/electrical contacts.

3. Results and discussion

3.1. Temperature dependence of thermal conductivity

In figure 1 we show the dependence on temperature (T) of κ_{001}/T for Sr_2RuO_4 in magnetic fields along the [100] direction. κ_{001}/T shows a shoulder on entering the superconducting state,

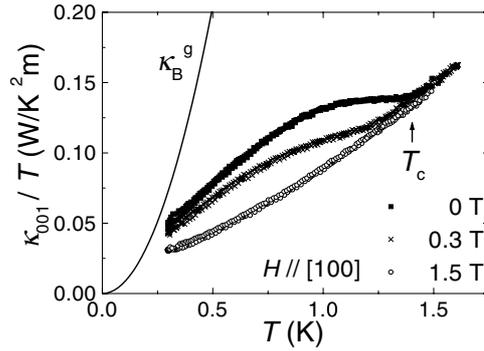


Figure 1. The temperature dependence of the thermal conductivity perpendicular to the highly conducting plane, κ_{001} , of Sr₂RuO₄ in magnetic fields of 0, 0.3 and 1.5 T (normal state) along the [100] direction. A solid curve shows the calculated phonon thermal conductivity in the boundary scattering mode.

in contrast to the in-plane thermal conductivity κ_{100} , of the same high-quality ($T_c = 1.44$ K) crystals, which shows an immediate decrease below T_c [19].

To obtain an appropriate understanding of these results, the identification of the dominant heat carriers and the scattering mechanisms is important. The thermal conductivity κ is a sum of contributions of electrons and phonons, $\kappa = \kappa^e + \kappa^g$, and in the superconducting state both of these quantities change in a complicated way. In the normal state, however, throughout the whole temperature range studied we are already in the residual resistivity range; therefore, the electronic component κ^e/T should remain constant as T changes.

The simplest way to estimate κ^e is to calculate it from the value of the electrical resistivity ρ . The two quantities are related by the Wiedemann–Franz (WF) law, $\rho \times (\kappa^e/T) = L_0$, where L_0 is a Lorenz number: $2.45 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$. This law was shown to be satisfied in Sr₂RuO₄ [19], in contrast to the electron-doped cuprate superconductors [31]. To exclude large errors in determination of the sample geometry, we compare measurements of both quantities made on the same electrical/thermal contacts. Using the resistance value in a field of 1.5 T along the [100] direction, the electronic contribution to κ_{001} in the normal state is evaluated as $\kappa_{001}^e/T = 9.9 \text{ mW K}^{-2} \text{ m}^{-1}$, which amounts to 7% of the total κ_{001} in the normal state at 1.5 K and 32% at 0.3 K. The validity of the WF law is based on the hypothesis that the electronic conduction is determined by the elastic scattering process. The inelastic processes reduce the thermal current more effectively than the charge current, giving a WF ratio smaller than L_0 . Thus this method of estimation provides the upper bound of the actual electronic contribution.

An alternative way of estimating κ^e uses the assumption that the relative changes of the electrical conductivity ρ^{-1} and of κ^e with field are the same [34]. In this way, using the data for magnetothermal resistance and magnetoresistance in the normal state with the field along the [100] direction, we obtain a consistent value $\kappa_{001}^e/T = 9.5 \text{ mW K}^{-2} \text{ m}^{-1}$, 7% of the measured κ_{001}/T at 1.5 K and 30% at 0.3 K.

As we see above, in the normal state, the thermal conductivity is mainly phononic in origin. This is moreover true for the superconducting state, since here, due to the dominance of impurity scattering for conduction electrons, the electronic contribution κ^e/T shows a notable additional decrease as compared to the normal state [19]. Therefore we can conclude that the enhancement of κ_{001} below T_c is due to the phonon mean free path increase, the cause of which will be discussed below.

Possible scattering mechanisms in the phonon heat conduction include the scattering on the sample boundaries, conduction electrons in the normal state, thermally excited quasi-particles and vortex cores in the superconducting state. As can be seen in figure 1, above T_c , κ_{001}/T changes linearly with temperature ($\kappa_{001}/T \propto T$), which shows that the κ^g is limited mainly by scattering on conduction electrons. Thus the hump of κ_{001}/T observed below T_c is interpreted as a result of the condensation of electrons into Cooper pairs which do not contribute to the scattering of phonons. For the conventional superconductors, Bardeen, Rickayzen and Tewordt (BRT) calculated the increase of κ^g due to the enhancement of the phonon mean free path caused by the formation of the Cooper pairs below T_c [35], assuming an exponential decrease of the quasi-particle density. According to their theory, an almost ten times increase of κ^g is expected at the temperature of $0.5 T_c$ [35]. It can be seen that the actual increase of the thermal conductivity is far lower than this expectation, and the large difference indicates the existence of additional scatterers such as grain/sample boundaries or excess quasi-particles resulting from the existence of nodes in the gap.

The role of the boundary scattering can be evaluated in the following way. In the boundary scattering regime, the phonon thermal conductivity is described by the asymptotic formula $\kappa_B^g = \frac{1}{3}C\bar{v}l \propto T^3$, where $C = \beta T^3$ is the phonon specific heat per unit volume, \bar{v} is the mean sound velocity and l is the mean free path of the heat carriers limited by the sample dimension. The mean sound velocity along the [001] direction of the tetragonal crystal is described by the relation $\bar{v} = v_L \frac{2s^2+1}{2s^3+1}$, where $s = v_L/v_T$ is the ratio of the longitudinal and transverse sound velocities [36]. The mean free path of phonons in the boundary scattering regime is $l_B = \frac{2d}{\sqrt{\pi}}$, where d is the mean width of the sample cross-section [36]. If we take a phonon specific heat coefficient of $\beta = 0.197 \text{ mJ K}^{-4} \text{ mol}^{-1}$ [23], sound velocities $v_L = 3900 \text{ m s}^{-1}$ and $v_T = 1300 \text{ m s}^{-1}$ at low temperatures [37] and $d = 0.5 \text{ mm}$, we obtain the phonon thermal conductivity in the boundary scattering regime as $\kappa_B^g = 0.8T^3 \text{ W K m}^{-1}$. This curve is plotted in figure 1 as a solid curve, which lies well above the actual data over the whole temperature range. This means that the phonon thermal conductivity is strongly limited by the large number of the thermally excited quasi-particles in the vicinity of nodes. Actually the observed height of the hump below T_c is eight times lower than the prediction based on scattering on the sample boundaries.

3.2. Field dependence of thermal conductivity

Now we discuss the field dependence of the phonon thermal conductivity. For the separation of the κ^e and κ^g , we analyse the different field dependences expected for two kinds of heat carrier in comparison with the known field dependence for κ^e [19].

The field dependence of κ_{001}/T at 0.3 K in the magnetic field applied along the [001] direction is shown in figure 2(a). On field increase from 0 T, κ_{001}/T at first remains nearly constant, then shows a sharp drop, takes a minimum and then starts to increase rapidly towards H_{c2} . Finally, κ_{001}/T flattens above H_{c2} in the normal state. The plateau observed at low fields corresponds to the complete Meissner state with $H_{c1} \sim 12 \text{ mT}$ at⁷ 0.3 K.

Since the effect of magnetic fields on the phonon thermal conductivity comes only from the creation of additional scatterers, κ^g decreases monotonically with fields. The field dependence of κ^e of Sr_2RuO_4 is more complicated. At high temperature, κ^e shows an initial decrease with field, and then increases towards H_{c2} [19]. However, at low temperatures, κ^e increases monotonically with field [19]. Thus the rapid increase of κ_{001}/T near H_{c2} comes from the

⁷ In our study, the sample is elongated along the [001] direction and the demagnetization factor is close to unity. Thus H_{c1} obtained here is more reliable than in our previous work [19] although this is twice larger than the estimate given by Akima *et al* [38].

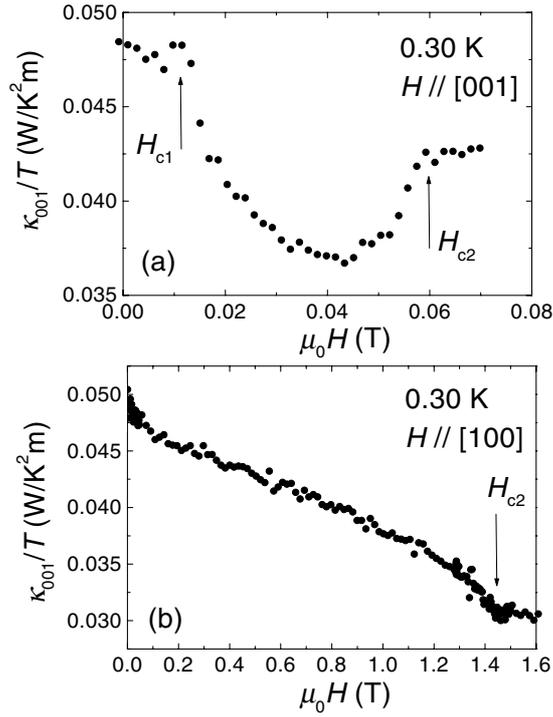


Figure 2. The magnetic field dependence of κ_{001}/T at 0.30 K; (a) in magnetic fields along the [001] direction; (b) in magnetic fields along the [100] direction.

increase of κ^e against the slowly decreasing background of κ^g . This allows us to decompose the field dependence into electronic and phononic components. We assume that the electronic part of κ_{001} has the same field dependence, except for a difference in amplitude, as κ_{100} which is purely electronic in origin. Then the field dependence of κ_{001}^g can be determined by subtracting the scaled κ_{100} (i.e. κ_{001}^e) from the total κ_{001} as shown in figure 3. Here the scaling factor is used as an adjustable parameter, chosen so as to produce coincidence of the scaled κ_{100} - and κ_{001} -curves near H_{c2} . As shown in the figure, in the superconducting state, the phonon contribution is strongly dominant, accounting for 88% of κ_{001}/T at 0 T and 0.3 K. Although efficient, this procedure is, strictly speaking, not correct, because we neglected the difference in magnetoresistance and hence magnetothermal resistance between κ_{100}^e and κ_{001}^e . However, since the magnetic field necessary to suppress the superconducting state in the $H \parallel [001]$ configuration is small, this correction is not of great importance here.

In contrast, in magnetic fields parallel to the plane, the magnetoresistance becomes large for the current perpendicular to the plane even just above H_{c2} [39]. In our case the resistance at H_{c2} in a perpendicular field is approximately 1.7 times smaller than in a parallel field. This magnetoresistance accounts for the smaller normal-state electronic thermal conductivity in parallel fields (30–32% of the total conductivity at 0.3 K; see section 3.1) as compared to the perpendicular field case, in which case κ^e constitutes about a half of the total normal-state conductivity (figure 3).

The dependence of κ_{001}/T on magnetic fields $H \parallel [100]$ (along the plane) at 0.3 K is shown in figure 2(b). Contrary to the perpendicular field case, the κ_{001}/T curve does not have the minimum, resulting from increase of the electronic conductivity towards H_{c2} . Instead, the

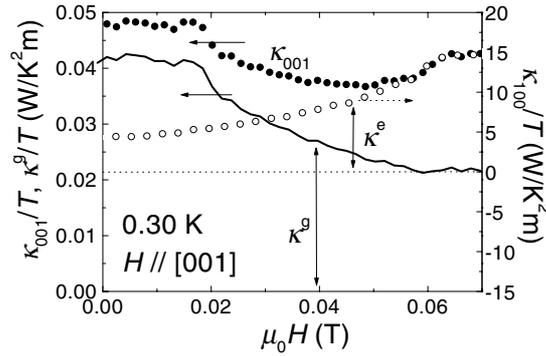


Figure 3. The magnetic field dependence of κ_{001}/T in magnetic fields along the [001] direction at 0.30 K (closed symbols). Open symbols represent the field dependence of the thermal conductivity with the heat flow along the conducting plane κ_{100} , measured in the same field orientation and temperature, scaled to fit the increase of κ_{001} near H_{c2} . The solid curve shows the difference, representing the field dependence of the phonon conductivity κ_{001}^g .

conductivity decreases gradually and then shows a rapid *decrease* towards the normal-state value in a narrow field range in the vicinity of H_{c2} . We point out here that the absence of the upturn at H_{c2} in parallel fields at low temperatures, despite a notable increase of κ^e [19], is very unusual. This implies that:

- (i) phonon scattering is strongly suppressed in the superconducting state at low fields;
- (ii) some additional mechanism for phonon scattering appears close to H_{c2} , which overcomes a rapid increase of κ^e .

The latter fact cannot find any explanation in the standard vortex scattering scenario. As is clear from the comparison with figure 3, the absence of the upturn rules out the application of the procedure that we used in the perpendicular field case for extracting $\kappa^g(H)$ in the superconducting state.

As we already mentioned above, the field dependence of the phonon thermal conductivity is totally determined by the variation of the scattering rate. The scattering in the superconductor is governed by the vortex cores and delocalized quasi-particles in the bulk, the latter acting similarly to conduction electrons in the normal state. For a superconductor with an anisotropic gap with line nodes, the quasi-particles in the bulk are generated by a magnetic field, as was first pointed out by Volovik [40]. This effect is caused by a supercurrent flow around a vortex and associated local Doppler shift of the quasi-particle spectrum. The shift causes an influx of the quasi-particles from the core into the bulk along the directions of the nodes in the superconducting gap. This increase of the quasi-particle density in fields is indeed observed in specific heat measurements of Sr_2RuO_4 [23], giving strong evidence for nodal structure of the superconducting gap. At low temperatures the density of delocalized QP is negligible in conventional type-II superconductors with the isotropic gap; therefore the phonon scattering in magnetic fields between H_{c1} and H_{c2} proceeds predominantly on the vortex cores. In this case the phonon thermal resistivity $W(H)$ increases linearly with H for $H > H_{c1}$, being proportional to the number of vortices. It can be expressed [41] as

$$W(H) = \frac{1}{\kappa^g} = W_0 + W_n H/H_{c2}, \quad (1)$$

where W_0 and $W_0 + W_n$ represent the phonon thermal resistivity in the Meissner and normal states, respectively.

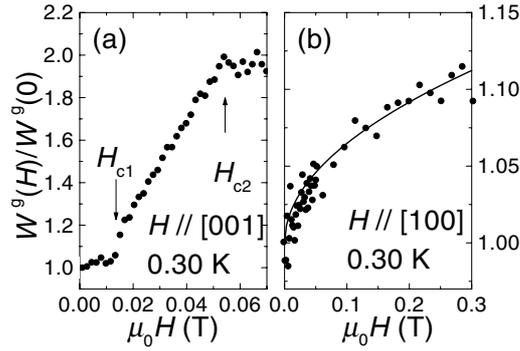


Figure 4. (a) The dependence of the phonon thermal resistivity on magnetic fields perpendicular to the plane ($H \parallel [001]$), extracted in the way shown in figure 3. (b) The low-field part of the field dependence of the thermal resistivity in magnetic fields along the [100] direction within the plane (symbols). The line represents a least-squares fits of the data with a \sqrt{H} -function.

Figure 4(a) shows the field dependence of the phonon thermal resistivity denoted, as $W^g(H)$, in the magnetic field $H \parallel [001]$. The phonon part was extracted as shown in figure 3. $W^g(H)$ increases linearly with field and shows the dominance of the vortex core scattering. In this field configuration, the coherence length is large ($\xi_{ab} \sim 660 \text{ \AA}$) [38], giving a large cross-section of the vortex core for the phonon scattering even at low fields. Thus the delocalized quasi-particles, despite their density being large as seen in the specific heat measurement [23], do not effectively influence the phonon transport.

In figure 4(b) we plot the thermal resistivity $W(H)$ at 0.3 K as a function of magnetic field parallel to the plane ($H \parallel [100]$). Here $W(H)$ is obtained directly from $1/\kappa_{001}$ and consequently contains a small electronic contribution. However, for fields $H < H_{c2}/2$, the electronic contribution slightly varies with field [19], and should give small and smooth offset of the zero line; hence the *shape* of $W(H)$ can well represent that of $W^g(H)$. As can be seen from the figure, $W(H)$ shows a notable sublinearity and at low fields exceeds the H -linear increase expected for scattering on the vortex lattice. This shows that the vortex scattering is not dominant in this configuration, which can be understood if we recall that the vortex core for parallel field is approximately 20 times smaller ($\xi_c \sim 33 \text{ \AA}$) [38] than in the perpendicular field. In the situation when the vortex scattering is weak, the delocalized quasi-particles induced by magnetic fields can become the main scatterers of phonons.

A quantitative description of the role of the delocalized quasi-particles in the phonon transport is very difficult to obtain at present, since this is almost unexplored theoretically. In a naive discussion, however, we can assume that the thermal resistivity is proportional to the number of scatterers, i.e. the delocalized quasi-particles. This density was calculated by Volovik for an isolated single-vortex core in a d-wave superconductor [40] and it is proportional to \sqrt{H} at very low temperatures. Thus we may consider that the phonon resistivity should be proportional to \sqrt{H} . The curve in figure 4(b) is the closest fit of the data with a \sqrt{H} -function. While this fit reproduces our result well, the above discussion is oversimplified and a proper theory for the phonon transport in the unconventional superconductor is strongly desired. Very recently, Won and Maki [42] calculated the field dependence of the phonon thermal resistivity in the nodal superconductor, assuming dominance of the longitudinal phonons in the heat transport at low temperatures and the validity of the Matthiessen rule for rates of scattering on sample boundaries and on quasi-particles. They derived a sublinear field dependence of the form $H \ln H$ due to the delocalized quasi-particles [42], which is not very distinct from the

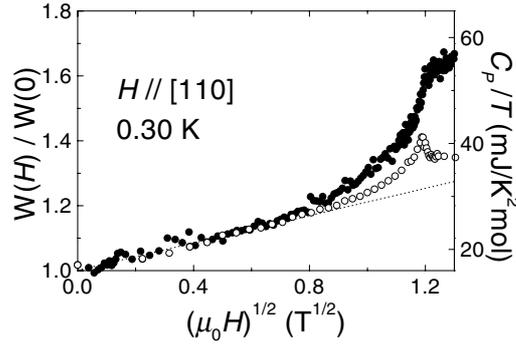


Figure 5. The field dependence of the thermal resistivity $W(H)$ (closed symbols) and specific heat (open symbols) from [23] in magnetic fields along the [110] direction within the plane at 0.30 K. The horizontal axis is scaled as \sqrt{H} .

\sqrt{H} -dependence at low fields and seems consistent with our results within the experimental error.

To check the relation between the phonon resistivity and the quasi-particle density, we compare in figure 5 the field dependence of the thermal resistivity and that of the specific heat given by Nishizaki *et al* [23] representing the quasi-particle density, both taken at 0.3 K in the same experimental configuration with in-plane fields. Note that the horizontal axis is scaled as \sqrt{H} . It can be seen that at low fields both quantities show similar sublinear dependence and match well, supporting our assumption on the phonon scattering on the delocalized quasi-particles. Despite this similarity, the two curves can never coincide completely, since corrections due to the electronic contribution and additional scattering on the vortex lattice are essential for the thermal conductivity at high fields. When scaled to overlap in the low-field region, the thermal resistivity appears to be higher than the specific heat curve. The electronic contribution κ^e , which can become important near H_{c2} , should cause a decrease of the resistivity and thus cannot be the reason for this deviation. Therefore we conclude that additional scattering on the vortex lattice is important as well.

Additional support for the role of scattering on field-induced quasi-particles in Sr_2RuO_4 in parallel fields comes from the unusual shape of the $\kappa(H)$ curve near H_{c2} at low temperatures. As can be deduced from equation (1), in fields close to H_{c2} the decrease of the phonon contribution due to scattering on the vortex lattice saturates to a value equal to that in the normal state. Therefore, as in the perpendicular field case, the increase of κ^e towards H_{c2} over this flat background gives an increase of total κ . However, the situation in Sr_2RuO_4 in magnetic fields parallel to the plane is notably different. Due to a multi-component order parameter, the superconducting state of Sr_2RuO_4 is characterized by the existence of at least two superconducting phases [43]. The main superconducting state occupies almost all of the H - T domain of the superconductivity, while the second superconducting state exists at low temperatures (below 0.8 K) at precisely aligned (within $\pm 3^\circ$ close to the plane) fields in close proximity to H_{c2} [19, 23, 44].

A phase transition from a low-field superconducting phase to a high-field one is accompanied by a release of a large density of quasi-particles. This release of QP gives a rapid increase of electronic thermal conductivity, which in the normal situation should give an upturn in $\kappa(H)$. However, since the release of the quasi-particles is similarly important for the phonon scattering, and the phonon contribution κ^g is much larger than κ^e , the decrease of κ^g dominates over the increase of κ^e and gives a downturn in $\kappa_{001}(H)$.

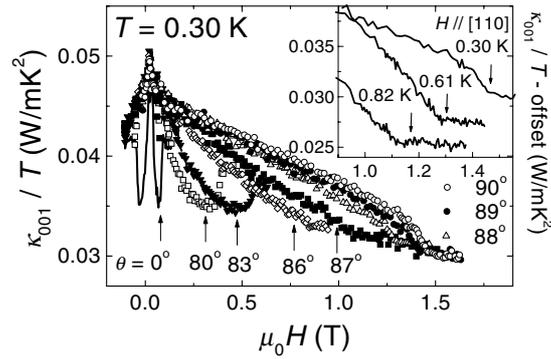


Figure 6. Transformation of the field dependence of κ_{001}/T at the base temperature (0.30 K) with the direction of the field. The numbers show the angle of field inclination to the conducting plane, being 90° for parallel alignment; the [110] direction. Inset: the magnetic field dependence of κ_{001}/T at several temperatures with H applied parallel to the conducting plane ([110]). Note that some offset is subtracted for each curve. Arrows show H_{c2} for each curve.

To see whether the unusual curvature is indeed related to the formation of the second phase, we show in the inset of figure 6 the $\kappa(H)$ dependence at 0.82 K, above the domain of the second superconducting phase. As can be seen, the curve shows the usual shape with a small upturn in $\kappa(H)$ near H_{c2} , despite the notably smaller electronic contribution to the total thermal conductivity at this elevated temperature. Simultaneously, as shown in the main panel of figure 6, the downward curvature appears in the same range of field inclinations (within $\pm 3^\circ$ to the plane) where the second superconducting state exists and where rapid release of quasi-particles is observed in specific heat and electronic thermal conductivity measurements [19, 23]. These observations clearly show that the downturn is determined by the formation of a second superconducting phase, and is determined by a rapid increase of scattering on quasi-particles near H_{c2} .

4. Conclusions

The thermal conductivity perpendicular to the conducting plane of Sr₂RuO₄ down to the lowest temperatures is dominated by phonon contributions, with scattering by conduction electrons or quasi-particles as the dominant scattering mechanism. The shoulder in the temperature dependence of the thermal conductivity in the superconducting state is related to the enhancement of the phonon mean free path due to the condensation of normal electrons below the superconducting gap, and its height is substantially lower than the expectation for a superconductor with a nodeless gap. In the mixed state the phonon heat transport is determined by the scattering on the vortex cores and on the bulk quasi-particles outside the core. The latter contribution is important in the configuration where the magnetic field is parallel to the plane, where the core radius is small. These results imply that the phonon thermal conductivity is a useful probe for characterization of the unconventional superconducting state, complementary to the electronic thermal conductivity or specific heat [45].

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