Thermal conductivity of superconducting Sr_2RuO_4 in oriented magnetic fields

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We report in-plane thermal conductivity along [100] direction, κ_{100}, of high quality single crystals ($T_c = 1.44$ K) of unconventional superconductor Sr_2RuO_4. Measurements were performed as a function of temperature T and magnetic fields H of various orientations. Linear decrease of κ_{100}/T in T in zero field is found below 1 K, in contrast to nearly constant value in the normal state. Field dependence of κ_{100}/T at 0.3 K in H perpendicular to the plane is qualitatively similar to that for a line-node superconducting state in the low temperature limit. The dependence gives $\mu_0 H_{c1} \approx 8$ mT and $\mu_0 H_{c2} \approx 60$ mT. In the in-plane fields, the field dependence at low temperatures shows a notable difference from the perpendicular field case. The temperature and field domain of this anomalous behavior is consistent with the existence of another superconducting phase, as proposed recently.

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I. INTRODUCTION

The discovery of superconductivity in Sr_2RuO_4,[1] a Ru analog of the parent high-T_c superconductor La_2CuO_4, stimulated notable interest to this material. As early as in 1995, Rice and Sigrist pointed out that the material may be a spin-tripled superconductor.[2] Series of experiments performed recently indeed showed unconventional character of the superconductivity in Sr_2RuO_4.[3] Muon spin rotation experiments revealed appearance of spontaneous magnetic field on entering the superconducting state, evidencing a superconductor with the broken time-reversal symmetry.[4] NMR studies showed the lack of a Knight-shift change on entering the superconducting state, as is expected for a spin triplet superconductor in which the spins of the Cooper pairs are oriented within the plane.[5]

However, the nature of the superconducting state in Sr_2RuO_4 is still a point of intense theoretical debate.[6] To some extent it is related to a contradictory experimental situation. Early experiments were performed on samples of insufficiently high crystal quality, showing large residual density of states in zero temperature T limit.[7] Later studies of dependence of the transition temperature on impurity[8] and defect[9] concentrations indicated that the impurity and defect free material has T_c of the order of 1.5 K, so-called intrinsic superconducting T_c. On sample quality improvement the residual density of states is notably decreased and seems to go to zero in the best samples.[10]

Experimentally, several key points should be addressed in relation to theoretical model of superconductivity in Sr_2RuO_4. These include the issues on the nodes in the superconducting gap, their location with respect to the Fermi surface and external magnetic field, and multiple superconducting phases.[11] The first question was addressed recently by the studies of NQR,[12] penetration depth,[13] and specific heat.[10] Most recent experiments seem to be consistent with a superconducting state with line nodes, as evidenced by low temperature exponents of various physical properties. Nevertheless, this conclusion is not well experimentally verified yet at very low temperatures. It may be possible to explain the observed quasiparticle spectrum within the nodeless model by inclusion of band-dependent gap. Furthermore, nothing is known on location of the nodes (if any) on the Fermi surface. The presence of multiple superconducting phases was proposed theoretically for a nodeless superconducting state.[14] In high enough magnetic fields, oriented along high symmetry directions within conducting plane, the nodeless gap is transformed into the one with the nodes, located in the directions perpendicular to the external magnetic field.[14] A phase transition within superconducting state was indeed observed in ac susceptibility and specific heat studies,[15] but only for magnetic field along [110] direction within conducting plane and at low temperatures. Hence, further experimental studies on the subject are essential.

Thermal conductivity κ is known as a powerful tool for study of the order parameter in unconventional superconductors.[16,17] Up to now the study of thermal conductivity in Sr_2RuO_4 was limited to low-quality samples (T_c below 1 K).[18] The results showed large residual term in κ/T (of the order of half of its value at T_c for $T \to 0$), the origin of which is thought to be an impurity induced pair breaking. No studies on magnetic field H effect were reported. In view of the sensitivity to impurities, it is essential to extend the study to high quality single crystals. In this article we report study of thermal conductivity in the high quality single crystals of Sr_2RuO_4 with the T_c of 1.44 K as a function of the magnetic field strength and the orientation with respect to the crystal and heat flow. We find a general agreement of temperature and field (oriented perpendicular to the plane) dependence of the thermal conductivity with those expected for a superconducting state with line nodes. We give evidence for an unusual dependence of thermal conductivity on a magnetic field parallel to the conducting plane. We show that this behavior appears in the H-T domain close to that of second superconducting phase found in ac susceptibility and specific heat studies,[15] but is observed in all the field directions within the highly conducting plane. Contrary to the expectation for the formation of the high-field state with a gap node direction following that of the magnetic field,[19,14] we find no increase in κ in transverse field configuration, as compared to the longitudinal configuration.

II. EXPERIMENT

Single crystals of Sr_2RuO_4 were grown by a floating-zone technique with Ru self-flux.[20] For our studies we selected crystals with T_c above 1.4 K, close to the intrinsic T_c of 1.5 K.[8] Measurement of thermal conductivity on high quality crystals is technically difficult, since κ of the material is very high. Therefore the resistance of the contacts (typically about 1 mΩ) is essentially higher than the resistance of the bulk of any typical sample. We improved bulk-to-contact resistance ratio by selecting very long and thin slabs, below 100 μm thick. The best crystal, for which detailed studies as a function of field orientation were performed, had $T_c = 1.44$ K and the size $2\times0.5\times0.07$ mm^3. Its long side, i.e., the direction of the heat flow, coincided with [100] crystallographic direction. Similar studies were performed on a lower quality sample ($T_c = 1.37$ K, not shown[21]). The data for the two samples are qualitatively consistent.

The samples were mounted directly on the cold finger of a miniature local vacuum cell.[22] Measurements were done according to the steady state one heater–two thermometers technique with RuO_2 flat chips resistors (model RK73K1EJ, KOA) as a heater and thermometers. The thermometers were calibrated both under zero and a set of low magnetic fields against a calibrated RuO_2 thermometer (Model RO600A2, Scientific Instruments, Inc.). Measurements of the electrical and thermal conductivity were performed on the same electrical/thermal contacts, but in two separate thermal runs. Simultaneous measurements were not possible because of the very low sample resistance of 30 ± 3 $\mu\Omega$ and use of high-resistance Pt-W wire to make electrical contacts to the sample in order to avoid thermal leak. Use of the high resistance wire limited affordable current density in resistivity measurements due to overheating, making them completely impossible in vacuum. The useful current density was still limited even in the ^{3}He liquid ambient, causing poor accuracy of the resistance measurement and hence of a Wiedemann-Franz ratio determination. In the normal state (in the magnetic field of 1.5 T parallel to the conducting plane) the ratio was found to be 1 ± 0.1 L_0 in very good agreement with the Lorenz number $L_0 = 2.44\times10^{-8}$ WΩK^{-2}. This agreement indicates that the phonon contribution in the normal state should not exceed the experimental error of 10%. Similar conclusion can be made from the slight temperature dependence of κ_{100}/T found in the normal state. It is in line with the measurements on the lower quality single crystals.[18]

As regards the superconducting state, it should be noted that the phonon contribution at these temperatures usually follows a T^3 dependence and thus should decrease more rapidly with temperature than the T^2 dependence observed in the zero field. We estimate an upper bound of phonon contribution in the superconducting state in the following way. Assuming a 10% phonon contribution at 1.5 K (the upper bound of the experimental error) and the T^3 dependence of the phonon thermal conductivity we come to a negligible contribution of 2% at 0.3 K. The absolute value of thermal conductivity is difficult to determine precisely, mainly due to a gross error in the contact and sample geometry determination. Based on the actual sample dimension, κ_{100}/T was found to be 12 ± 3 Wm^{-1}K^{-2} at 1.5 K. This value is a little low compared to the estimation based on the resistivity value expected for the material of this quality,[8,18] though it is not far from the interval determined by the experimental scatter of the resistivity for different samples.

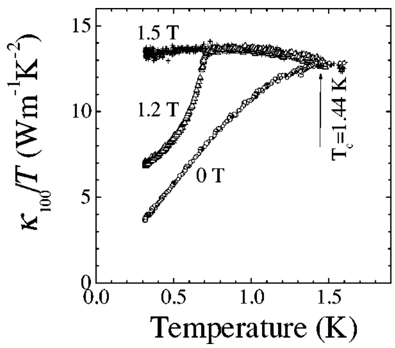

FIG. 1. Temperature dependence of the in-plane thermal conductivity, κ_{100}/T, of Sr_2RuO_4 in zero field, 1.2 T and 1.5 T magnetic field oriented along [010]. The heat flow is along the [100] direction. The field of 1.2 T is on the boundary for anomalous thermal conductivity behavior. The data at 1.5 T corresponds to the normal state.

The cell with a sample was placed in a double axis goniometer in ^{3}He refrigerator insert of a 17 T superconducting magnet. Orientation of the field parallel to the plane was made by determining the angular position of the maximum of H_{c2} from $\kappa_{100}(H)$ dependence. $\kappa_{100}(H)$ was measured in 1.3 to 1.5 T range at 0.3 K with a step of 0.1° in $\pm0.5°$ inclination range. This procedure gave an accuracy of the alignment of the order of $\pm0.1°$ with respect to the conducting plane. It was repeated for each of the three in-plane field orientations discussed below. $\kappa_{100}(H)$ curves in the close vicinity of H_{c2} does not change much in this range of inclinations, contrary to the ac susceptibility measurements, in which a complicated behavior is observed.[15] We performed measurements in four different experimental configurations. In the first one, the field was oriented along the [001] direction, perpendicular to both the conducting plane and the heat flow. In the second one, the field was oriented along the [100] direction, i.e., along the heat flow. In the third configuration the field was oriented along the [010], equivalent crystallographic direction, but perpendicular to the heat flow. And finally the field was oriented along the [110] direction in the conducting plane, 45° from the heat flow.

The measurements were made in both field-cooled and zero-field-cooled states. We did not detect any difference between these sets of data beyond experimental scatter. Similarly, the data measured at fixed field on temperature variation were consistent with those measured at fixed temperature on field variation.

III. RESULTS

In Fig. 1 we show the temperature dependence of thermal conductivity of Sr_2RuO_4 in zero field, and the magnetic fields of 1.2 T and 1.5 T oriented along the [100] direction in the superconducting plane. First of all we point out that the thermal conductivity in the normal state is nearly temperature-independent. A slight variation of the κ_{100}/T may be due to slight resistivity decrease on cooling. Thus we conclude that the sample is in the temperature range in which the carrier scattering is determined by the impurities, i.e., in a residual resistivity range.

This temperature-independent scattering makes the behavior of thermal conductivity in Sr_2RuO_4 drastically different from the more familiar case of high-T_c cuprates. In the cuprates the scattering in the normal state is electronic in origin, although its detailed mechanism is not known. Because of the electronic nature of scattering, two effects appear simultaneously on entering the superconducting state. The decrease in the quasiparticle density leads to a decrease in the density of electronic carriers, and hence thermal conductivity. On the other hand the decrease of the quasiparticle density leads to the mean-free-path increase due to the decrease of density of scatterers, both for electrons and phonons. In the cuprates the mean-free-path increase for electronic carriers dominates,[23] leading to a pronounced peak in the thermal conductivity. On contrary, the mean-free-path in Sr_2RuO_4 is determined by the scattering by crystal imperfections (impurities and defects) and is of the order of the interscatterer distance. Therefore, on entering the superconducting state the decrease of the electron-electron scattering does not lead to the mean free path change. As a result, to a good approximation we can consider the mean free path as a constant, with all the κ_{100}/T variation in the superconducting state coming from the quasiparticle density variation.

As can be seen from Fig. 1, in the superconducting state κ_{100}/T decreases below T_c and shows a gradual increase of slope below around 1 K. This slope change becomes less notable with sample quality improvement (compare with the results in Refs. 18 and 21). It is unlikely that the small slope change is caused by the mean-free-path variation of the electronic carriers, since it becomes essentially shorter and thus even less temperature-dependent in low quality samples. The slope change may either reflect a small residual contribution of the phonons, as indicated by diminishing of the feature with electronic thermal conductivity increase in high quality samples, or signal some transformation within the superconducting state, as indicated by an anomaly in the temperature dependence of H_{c2} anisotropy.[15] Below 1 K the κ_{100}/T decreases linearly, extrapolating to a value a little higher than zero at the origin. The linear variation of κ_{100}/T in the constant mean free path condition is consistent with the line node state, discussed from the recent results of specific heat[10] and NQR.[12] To make this statement definite, measurements of the thermal conductivity at lower temperatures are essential. We point out, however, that in the high quality crystals the linear κ_{100}/T is observed in a rather broad temperature range, not just in the low T limit.[24]

Of special note is a temperature dependence of κ_{100}/T in the field of 1.2 T. This field corresponds to the boundary of the appearance of the second superconducting phase in ac susceptibility and specific heat measurements.[15] The dependence shows the main feature specific to the second phase. On entering the superconducting state κ_{100}/T shows almost vertical decrease, followed by a more usual linear dependence.

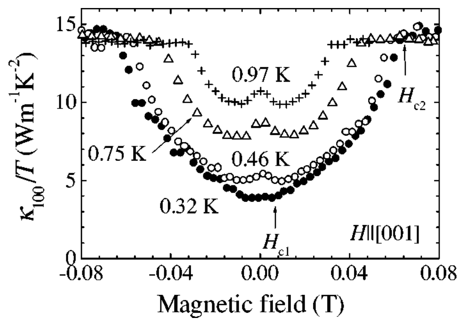

FIG. 2. Field dependence of κ_{100}/T in the magnetic field along the [001] direction, perpendicular to the conducting plane.

In Fig. 2 we show a dependence of κ_{100}/T on the magnetic field perpendicular to the plane at several representative temperatures. At high temperatures, κ_{100}/T shows small decrease on first vortex entering into the superconducting state. This feature is not sharp enough to make an exact determination of the H_{c1} for the perpendicular field. Assuming that the plateau in the vicinity of zero field in the 0.3 K curve represents the range of the fields which do not penetrate into the sample, we come to a value of 8 mT, in a reasonable agreement with the previous H_{c1} estimation.[25] The feature at H_{c1} becomes less notable at lower temperatures, indicating that the quasiparticle scattering becomes less important, in line with Vekhter and Huntington model.[26] On further field increase, the thermal conductivity increases more rapidly on approaching H_{c2}. The field dependence of the thermal conductivity in the perpendicular field can be reasonably understood in the framework developed for the high-T_c cuprates,[27,26] if we take its low temperature limit. In this model the dependence is determined by a competition between the quasi-particle density increase on field increase and a mean-free-path decrease due to a decrease in the intervortex distance. The model assumes that at high temperatures the scattering term is more essential, giving the thermal conductivity decrease on field increase. In the low temperature limit, the scattering becomes less important. As a result, the variation of thermal conductivity is mainly determined by the quasi-particle density increase, giving thermal conductivity increase with field. The numerical calculation of this model for low temperature regime[28] reproduces the main features of our data, namely, gradual thermal conductivity increase above H_{c1}, followed by a rapid increase on approaching H_{c2}.

This field dependence is, however, in a striking contrast with the behavior in a magnetic field parallel to the superconducting plane, Figs. 3(a)–3(c). We show the dependence for three in-plane orientations of the field at different temperatures. The main feature of this dependence, irrespective of the field orientation, is a sharp κ_{100}/T increase in the vicinity of H_{c2} at low temperatures. This behavior can be characterized by the field of the slope change, H_2 (see definition below in Fig. 5), and the slope of $\kappa_{100}/T(H)$ field dependence at H_{c2}, $d(\kappa_{100}/T)/dH\|_{H_{c2}}$. Such variation of the derivative with temperature is shown in Fig. 4. It can be seen that for all three configurations the slope changes non-

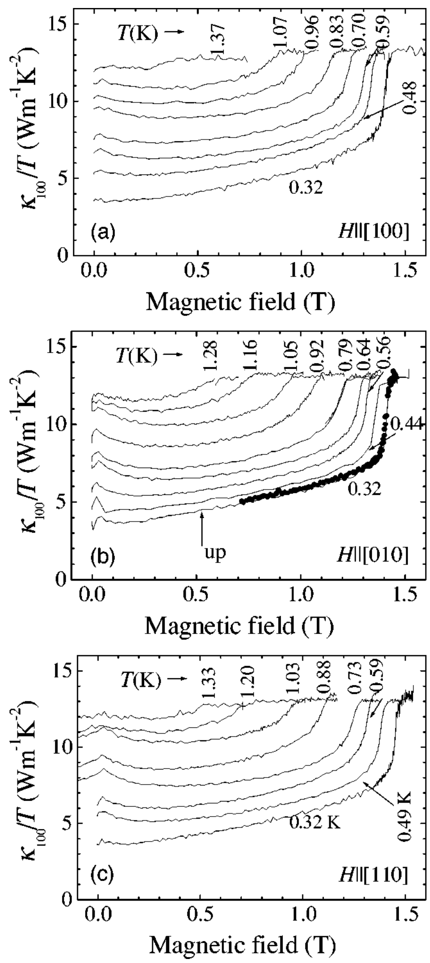

FIG. 3. Field dependence of κ_{100}/T in the magnetic field H parallel to the conducting plane. (a) H is in the [100] direction, parallel to the heat flow; (b) H is in the [010] direction, perpendicular to the heat flow. For 0.32 K the full line represents the data on increase in H, and the solid dots on decreasing H sweep; (c) H is along [110].

monotonically with temperature, indicating a special point on the $H_{c2}(T)$ line at approximately 0.8 K and 1.2 T (Fig. 4). This point is in a very good agreement with the results of ac susceptibility and specific heat measurements, showing realization of a second superconducting phase in the parallel magnetic field.[15] However, as can be seen from Fig. 3, con-

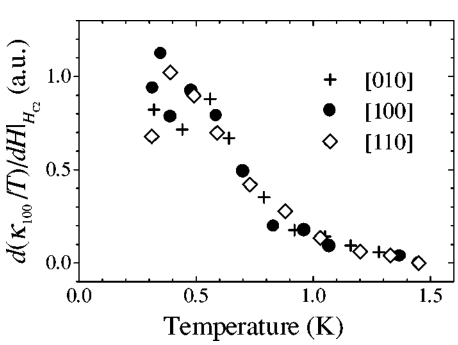

FIG. 4. The slope of κ_{100}/T vs H curve at H_{c2} for three in-plane orientations of the field.

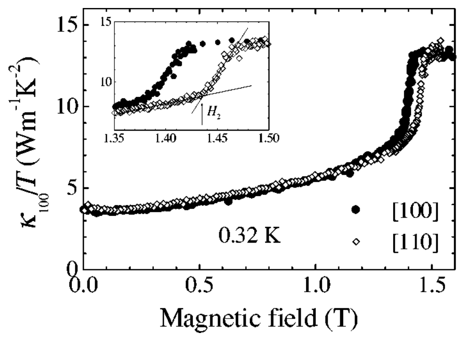

FIG. 5. Comparison of the field dependence of κ_{100}/T at 0.32 K for inequivalent crystallographic directions, [100] and [110]. Inset shows the range in the vicinity of H_{c2} on the expanded scale and definition of H_2.

trary to the ac susceptibility data which is complicated by the additional vortex pinning features, the behavior is clearly observed for both [110] and [100] directions.

IV. DISCUSSION

We start our discussion with addressing two relevant features of the dependence of the thermal conductivity on the orientation of magnetic field within the conducting plane. For a quasi-two-dimensional superconductor with line nodes running along the Fermi surface cylinder, like in a d-wave superconductor, a notable angular dependence of κ on a magnetic field orientation with respect to the crystal is expected theoretically[29,30] and observed experimentally in $YBa_2Cu_3O_x$ and $Tl_2Ba_2CuO_{6+x}$.[31–33] In these experiments, done at low temperatures (usually $T \ll T_c$) and with low values of H/H_{c2}, the anisotropy of κ was of the order of 0.5%. Another angular dependence was predicted theoretically for a superconductor with a two-component order parameter, in which an orbital part contains the term $(k_x \pm ik_y)$, where k_x and k_y are in-plane components of the electron momentum. In this case at some value of the in-plane field a phase transition from a state with an isotropic (nodeless) gap to a state with line nodes running along the Fermi surface in the direction perpendicular to the external magnetic field is predicted.[14] Therefore, in the high field state a notable anisotropy of κ on field orientation with respect to the heat flow is expected.[19]

In Fig. 5 we compare the field dependence of thermal conductivity for the [100] and [110] directions at 0.3 K. Since these are the two principal high symmetry directions within the conducting plane, they should represent the extreme cases of the maximum and the minimum of the thermal conductivity. It is clear that the difference between the curves comes almost solely from the anisotropy of the upper critical field, amounting to 3% of the H_{c2}, in good agreement with the ac susceptibility data.[15] When plotted in the dimensionless coordinates H/H_{c2}, the curves coincide within the accuracy of our experiment, of the order of 2%. Although this uncertainty is notably larger than the anisotropy observed experimentally in the cuprates, our measurements cover much broader field range extending all the way to H_{c2}, in which case much larger anisotropy is expected.[29] This

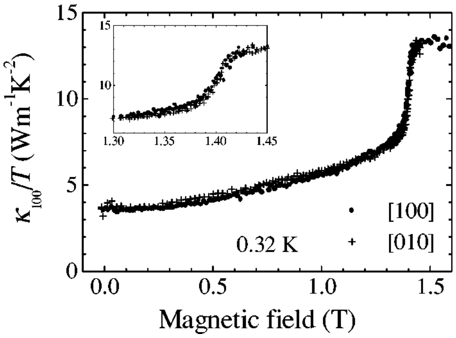

FIG. 6. Comparison of the field dependence of κ_{100}/T at 0.32 K for the longitudinal [100] and the transverse [010] magnetic field vs heat flow configurations. Inset shows the range in the vicinity of H_{c2} on the expanded scale.

observation seems to be against the superconducting state with nodes along the Fermi surface cylinder, or at least against the state having nodes *only* along the Fermi surface cylinder.

In Fig. 6 we compare the field dependence of κ_{100} for [100] and [010] directions, i.e., parallel and perpendicular to the heat flow, at 0.3 K. If we take that the rapid increase of the thermal conductivity near H_{c2} proceeds owing to a phase transition into a field-induced state with an additional line-node, located in the direction perpendicular to the field, we expect a notable thermal conductivity increase in the [010] configuration as compared to the [100] configuration. As we can see, contrary to this prediction, no anisotropy of the thermal conductivity increase is observed above H_2 in the field perpendicular to the flow. Thus our data does not support a ''polarizable'' gap, expected for a two-component order parameter above the phase transition field.

We now come to discussion of the field dependence of thermal conductivity in the in-plane fields. In Fig. 7 we compare the shapes of the dependence for the perpendicular and parallel to the plane field orientations, drawn in dimensionless field, H/H_{c2}. As can be seen, the shape of the dependence is notably different. The main difference is in a slight thermal conductivity increase for the in-plane field, and rapid restoration of the normal state value in the very vicinity (40 mT) of H_{c2}. At low temperatures, more than a half of the

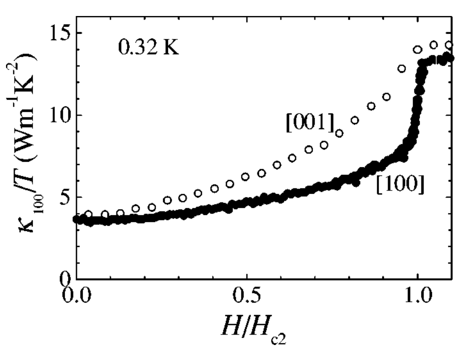

FIG. 7. Comparison of the field dependence of κ_{100}/T at 0.32 K for the in-plane and perpendicular to the plane directions. The curves are presented in the normalized field, H/H_{c2}.

thermal conductivity restoration is observed in this field interval.

It is clear that the magnetic field penetration into a sample increases the thermal conductivity. Therefore the most naive way to explain the slow increase of κ_{100}/T in the parallel field as compared to the perpendicular field is to assume that the field parallel to the plane is more difficult to penetrate into a sample, as compared to the perpendicular field case. The complications of the field dependence of magnetization in the parallel field were indeed observed in Bi-2212.[34] This situation can be realized if the critical current along the plane J_{ab} is lower than the critical current perpendicular to the plane J_{cc}, due to either pinning or surface barrier. In the low quality samples of Sr_2RuO_4 it was shown experimentally that $J_{cc} \ll J_{ab}$,[1] i.e., opposite situation is realized in Sr_2RuO_4. The same conclusion was made in the magnetization studies,[35] showing that the field is much easier to penetrate along the plane. Therefore, it is difficult to believe that the slow κ_{100} increase in the parallel field is due to the complication in field penetration. In addition, to explain the data in the parallel field in this way it is necessary to assume that the field inside a sample is several times lower than the field outside. The magnitude of the effect appears by far larger than in Bi-2212.[34] This makes this interpretation quite unlikely in the fields near H_{c2}. Besides, the data of the specific heat,[10] ac susceptibility[15] and thermal conductivity are in general agreement, although the samples of quite different shapes and orientations are used in these studies, thus making crucial role of the surface barriers unlikely.

There are two ways to interpret the flat thermal conductivity dependence in the in-plane field. One option is to assume that the scattering of quasiparticles in the in-plane field is much stronger than in the perpendicular field. Since the Volovik effect[36] (and hence the density of quasiparticles) is not expected to depend on the orientation of the field, to obtain flat dependence in the in-plane field we need to introduce additional scattering on the vortices. Alternatively, we can assume that the effect is specific to Sr_2RuO_4 and is due to a small quasiparticle density increase, which is much less in the parallel field than in the perpendicular field, as can be the case for a two-component order parameter due to a single component vortex formation.[14] From thermal conductivity experiment alone it is not possible to decide which of the contributions, from the mean free path or quasiparticle density variation, is important. However, the specific heat data, which are not sensitive to the scattering, indicates a rapid quasiparticle density increase in the parallel field near H_{c2}, favoring the situation when scattering is not of dominant importance.[15]

To our knowledge, no other superconductor shows a field dependence of the sort we observe in a parallel field. It is very different from the behavior of a conventional type II superconductors.[37] It is in contrast with the known behavior of the heavy fermion superconductors,[16,17,38] in which case gradual increase of κ is observed. It is much sharper than the dependence in organic superconductors in the perpendicular field,[39] which is in a reasonable correspondence with our data for the perpendicular field. In the cuprates the behavior at low temperatures in the perpendicular field indicates

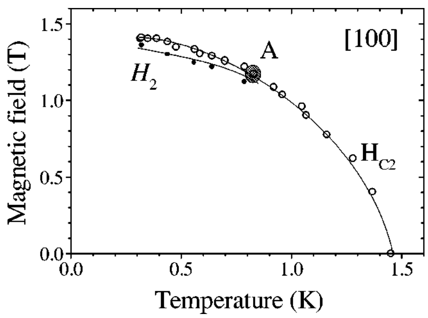

FIG. 8. Phase diagram of Sr_2RuO_4 in the in-plane magnetic field, as seen from the thermal conductivity data. The H_2 line corresponds to a point of field dependence (shown in the inset of Fig. 5) where a slope change in the vicinity of H_{c2} starts. Dashed dot A on H_{c2} line is a point of slope change in κ_{100}/T vs T dependence at H_{c2} (Fig. 4).

gradual thermal conductivity increase.[40,41] In the in-plane field, studies were performed and compared with the case of a field perpendicular to the plane in a number of materials at relatively high temperatures.[42] Naturally, these studies do not cover the field range close to H_{c2} at low temperatures, yet the opposite tendency of κ increase in the in-plane field as compared to the κ decrease in the perpendicular field was noticed, indicating that the scattering is less important in the in-plane configuration.

In view of a good coincidence of temperature-field domain of the anomalous behavior with that of the second superconducting phase in the ac susceptibility and specific heat measurements,[15] it is natural to relate the difference with the formation of a new superconducting phase. We would like to point out that both strong quasiparticle density suppression in fields below the second transition[43] and a rapid thermal conductivity increase above it are in qualitative agreement with the model of Agterberg,[14] despite the other inconsistencies discussed above.

The phase transition into high-field state at H_2 should be of the second order, since no hysteresis of $\kappa_{100}(H)$ in a magnetic field is observed. If we define the H_2 as a field of a slope change in $\kappa_{100}(H)$, as shown in Fig. 5, we can draw the line, shown in Fig. 8, summarizing the H-T phase diagram of Sr_2RuO_4 from our thermal conductivity studies. Above 0.8 K it is difficult to find firm traces of the slope change point at H_2 due to the broadening of the feature. This phase diagram is in general agreement with the one proposed in Ref. 15. The essential difference is in the observation of the second phase for magnetic fields along both the [100] and [110] directions. In conjunction with the lack of thermal conductivity anisotropy above the transition point, this makes interpretation of this feature within Agterberg model unlikely.

V. CONCLUSION

The temperature dependence of the thermal conductivity of Sr_2RuO_4 in zero magnetic field and the field dependence of thermal conductivity in perpendicular to the plane field at low temperatures show basic correspondence with those of a superconducting state with line nodes. Angular dependence of the thermal conductivity in the in-plane field does not show the anisotropy with respect to the heat flow direction, contrary to the expectation from a polarizable line-node state. The anisotropy of the field dependence of thermal conductivity with respect to the crystal lattice seems to be too small for any state with line nodes running perpendicular to the two-dimensional plane.

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