RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 58, NUMBER 16 15 OCTOBER 1998-II

High-temperature resistivity of Sr_2RuO_4: Bad metallic transport in a good metal

A. W. Tyler and A. P. Mackenzie
Interdisciplinary Research Centre in Superconductivity and Department of Physics, University of Cambridge,
Madingley Road, Cambridge CB3 OHE, United Kingdom
and School of Physics and Astronomy, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

S. NishiZaki and Y. Maeno
Department of Physics, Kyoto University, Kyoto 606-8502, Japan

(Received 30 June 1998)

We report the results of a study of the in-plane and out-of-plane resistivity of the layered perovskite superconductor Sr_2RuO_4 at temperatures between 4 and 1300 K. Although the material is a very good metal at low temperatures, with in-plane mean free paths (l) of thousands of lattice spacings, l falls smoothly to less than 1 Å at 1300 K with no sign of resistivity saturation at the Mott-Ioffe-Regel limit. Measurements of the incoherent out-of-plane resistivity over the same range of temperature are also discussed.
[S0163-1829(98)50840-5]

Understanding electrical transport in metals with very high scattering rates has long been an important goal in condensed-matter physics. The commonly used framework of semiclassical particles moving subject to the constraints of the periodic potential, and scattering in the relaxation-time approximation, is really only applicable in the limit for which the mean free path (l) is much greater than the lattice spacing (a), because particles can only be identified on this rather long length scale. The question of what happens in materials which are so disordered that l becomes comparable to a was addressed by several authors,[1,2] leading to the concept of the Mott-Ioffe-Regel limit (MIRL) for metallic conductivity, which states that the metallic state cannot exist for an arbitrarily high scattering rate. The precise numerical definition of the limit is not universally agreed upon; criteria ranging from $k_F l \sim 1$ (k_F is the Fermi wave vector) through $l \sim a$ to $k_F l \sim 2\pi$ have all been suggested. Experiments on very disordered materials also show a range of values, but there is little doubt that the concept of a minimum metallic conductivity is correct in this class of system.

The arguments for disordered materials were developed for elastic scattering at low temperatures. The issue of whether a very high scattering rate caused by strong inelastic scattering at high temperatures should have a similar effect is more controversial. Experimental observation of the appropriate limit requires the study of materials in which the scattering rate becomes high enough before high-temperature structural changes or melting occur. In practice, this means chemically stable narrow-band metals for which the Fermi velocity is sufficiently low that $l \sim a$ at temperatures of 1000 K or less. Some of the first materials in which this regime was studied extensively were the ''*A*15'' superconductors, such as Nb_3Sn, in which superconductivity at $T \sim 20$ K is thought to be due to very strong electron-phonon coupling. A number of experiments[3–5] showed the high-temperature resistivity becoming only very weakly temperature dependent, appearing to limit to values that were consistent with the MIRL as estimated from calculations of the electronic structure *and* the value of the MIRL measured at low temperatures in heavily irradiated samples of the same materials. Similar observations were made in a number of narrow-band transition-metal elements,[6] and the concept of high-temperature resistivity saturation at the MIRL was widely assumed to be a general property of metals, although it was difficult to justify theoretically.[5,7,8]

More recently, a series of metals have been discovered which show no sign of resistivity saturation at high temperatures. These include alkali-doped C_{60}, some organic compounds, and the high-temperature superconducting cuprates, which all have superconducting ground states. These observations motivated Emery and Kivelson[9] to introduce the term ''bad metals,'' and to argue that the high-temperature conduction mechanism in these materials must be unconventional. They also made the intriguing suggestion that since there was no evidence of a transition or even a crossover between the high- and low-temperature regimes, the unconventional conduction mechanism might also apply at all temperatures down to the superconducting transition, even though estimates suggest that $l \gg a$ in that temperature range. Since then, other bad metals with interesting magnetic ground states have also been studied,[10] but it has not been possible to answer the interesting question of whether bad metallic behavior at high temperatures can occur in materials which are proven to be very good metals at low temperatures.

The layered perovskite metal Sr_2RuO_4 provides an ideal opportunity to investigate this issue. At low temperatures, it is clearly a very good metal indeed, with a superconducting transition below 1.5 K.[11] The observation of quantum oscillations has provided a detailed understanding of the Fermi surface (FS) topography and the average Fermi velocity on each FS sheet, resulting in the successful calculation of a number of independently measured low-temperature transport and thermodynamic properties.[12,13] Analysis of resistivity data from a series of samples shows that the low-temperature values of $k_F l$ can be as high as 5000, dropping to approximately 6 at room temperature. Since the material has a very high melting point of approximately 2000 °C, there was a good prospect of obtaining successful high-temperature resistivity data. This provided our main motivation to study the in-plane resistivity to temperatures as high as 1300 K. We also decided to investigate the behavior of the out-of-plane resistivity, since this would give us an opportunity to study incoherent high-temperature transport in a metal with a similar crystal structure and anisotropy to the cuprates.

The high-quality single crystals used for this study were grown in an image furnace, using methods described previously.[11] Measurements of the resistivity at temperatures between 4 and 320 K were performed using a standard four-terminal ac method in a ^{4}He cryostat. Errors in the absolute value of ρ due to uncertain electrical contact geometry were reduced to a few percent by averaging results taken from many samples. For work above room temperature, we constructed a special probe (length approximately 30 cm) designed to fit into a small tube furnace. Platinum wires enclosed in fine silica tubes were used for the electrical signals (in this case the resistivity was measured using a four-terminal dc method), and a platinum 10% rhodium/platinum thermocouple was used to measure the temperature. The final contacts to the single-crystal samples were gold wires to contact pads made by applying a colloidal suspension of ultrafine gold particles (Vacuum Metallurgical Co., Ltd., Tokyo) which melt to a solid gold pad during a short initial firing at only 300 °C.[14] Once melted, these pads gave contact resistances of less than 1 Ω at room temperature, and remained strong and stable all the way to the melting temperature of bulk gold at approximately 1300 K. The reference junction for the thermocouple was in a copper block at the room-temperature end of the probe. A second thermometer was used to monitor this block as the furnace temperature was changed, and a correction was made for changes of a few degrees in its temperature. The probe fitted into a silica furnace tube with a glass-glass conical seal, so that the experiments could be carried out in flowing gases. In a typical run, data were taken by repeatedly ramping the temperature to a set point and then cooling to near room temperature, increasing the set point by approximately 100 K each time. The repeatability of the results through this cycling demonstrated that no measurable irreversible changes were taking place. By repeating this procedure in flowing oxygen and argon, we also ruled out the possibility that reversible changes of the oxygen content were having a measurable effect on the resistivity. In comparison to the cuprates, Sr_2RuO_4 is remarkably stable at these elevated temperatures.

As shown in Fig. 1, the in-plane resistivity of Sr_2RuO_4 rises smoothly to a large value in excess of 1 mΩ cm at 1300 K.[15] The size of the resistivity is particularly significant when it is converted into an estimate of the mean free path l. Standard Boltzmann transport theory for a quasi-two-dimensional multiband material gives

$$\sigma = \frac{e^2}{hd} \sum_i k_F^i (v_F \tau)^i, \tag{1}$$

where d is the interplane distance (6.37 Å at 300 K in Sr_2RuO_4), k_F is the Fermi wave vector, v_F is the Fermi velocity, τ is the relaxation time, and i denotes the band index. At low temperatures, where the mean free path becomes approximately equal for each Fermi surface sheet, this gives a simple expression for $l = v_F \tau$ in terms of ρ:

$$l = \frac{2\pi\hbar d}{\rho e^2 \sum_i k_F^i}. \tag{2}$$

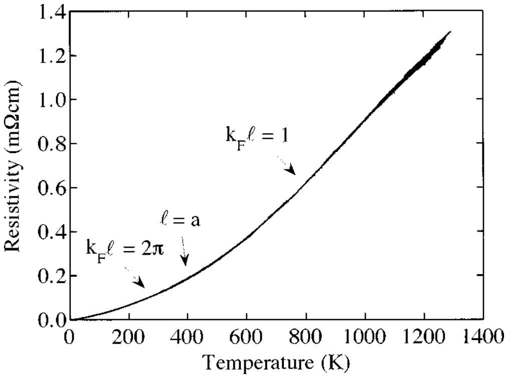

FIG. 1. The in-plane resistivity of Sr_2RuO_4 from 4 to 1300 K. Three criteria for the Mott-Ioffe-Regel limit are marked on the graph, and there is no sign of resistivity saturation, so Sr_2RuO_4 is a ''bad metal'' at high temperatures, even though it is known to be a very good metal at low temperatures.

At high temperatures, it is a better approximation to consider τ to be independent of the FS sheet, so differences in v_F for each sheet should, in principle, be taken into account. In Sr_2RuO_4, the Fermi surface parameters are known from quantum oscillation measurements,[12] and v_F is sufficiently similar for the three sheets that Eq. (2) gives an estimate for the average value of l that is accurate to within ≈20%. The main point is that any estimate based on the standard theory gives a high-temperature mean free path of less than 1 Å, or a factor of 5 less than the in-plane lattice spacing of Sr_2RuO_4 (3.87 Å at 300 K). This version of the MIRL is marked on Fig. 1, along with $k_F l = 2\pi$ and $k_F l = 1$; there is no sign of saturation near any of the criteria. Violation of the MIRL for in-plane transport in Sr_2RuO_4 is one of the central results of this study.

The out-of-plane resistivity of Sr_2RuO_4 is also of considerable interest, particularly in relation to the cuprate high-temperature superconductors. At 300 K it has a negative temperature derivative, as seen in some cuprates, but there is a maximum at around 130 K, denoting a crossover to metallic behavior at low temperatures. Below approximately 20 K, the resistivity has a quadratic temperature dependence in all directions, and Sr_2RuO_4 is a highly anisotropic Fermi liquid.[16] The temperature-independent anisotropy that is seen at low temperatures in Sr_2RuO_4 is not observed in any cuprate material.[17] The temperature at which it disappears seems to correlate with the temperature at which the in-plane scattering rate destroys coherent band formation in the c direction.[12] Although the nature of the apparently metallic out-of-plane conduction between 20 and 130 K is not yet well understood, there is little doubt that the mechanism is incoherent at room temperature. By working at much higher temperatures, we hoped to gain some insight into the mechanisms of highly incoherent conduction.

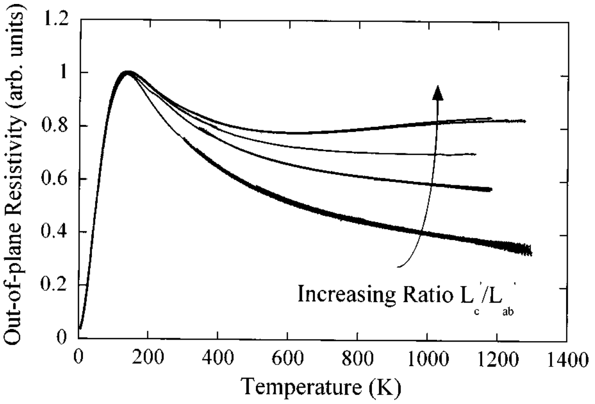

FIG. 2. The apparent out-of-plane resistivity of five samples of Sr_2RuO_4 of varying thickness, measured by a quasi-Montgomery method and normalized at 130 K. The large variation at high temperatures arises because the anisotropy falls rapidly with increasing temperature, so that crystals which are thick enough for the quasi-Montgomery method to give good results at low temperatures are no longer thick enough at high temperatures. As the thickness of the crystals is increased, however, the data converge to the correct value over the whole temperature range. The data from the two thickest crystals are almost indistinguishable in the top trace, demonstrating that this convergence has been achieved.

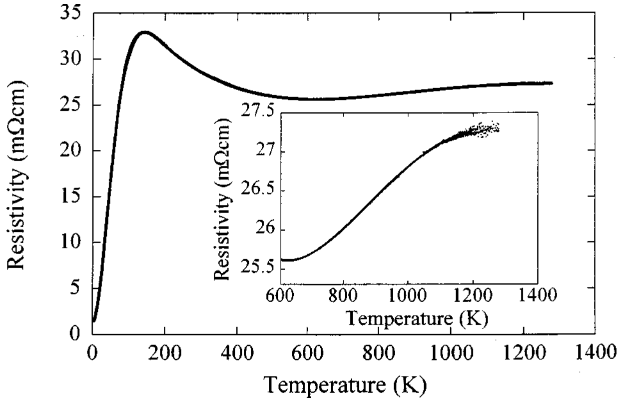

FIG. 3. The intrinsic out-of-plane resistivity of Sr_2RuO_4 from 4 to 1300 K. Although the high-temperature value is nearly 30 mΩ cm, the temperature derivative is positive between 700 and 1300 K, as shown in the inset.

The out-of-plane measurements were made using a quasi-Montgomery method in which equivalent isotropic dimensions for a sample with an anisotropic resistivity are first calculated, following van der Pauw:[18]

$$\frac{L_c^i}{L_{ab}^i}=\frac{L_c}{L_{ab}}\sqrt{\frac{\rho_c}{\rho_{ab}}}. \tag{3}$$

Here, L is an average sample dimension, subscripts ab and c refer to directions perpendicular and parallel to the c axis, the superscript i refers to the equivalent isotropic sample, and ρ is the resistivity. Instead of mounting corner contacts for a full Montgomery analysis,[19] a simple four-terminal measurement with current and voltage contacts on each face of a platelike crystal gives negligible error as long as $L_c^i \gg L_{ab}^i$. Initial studies on Sr_2RuO_4 showed, however, that the resistive anisotropy is strongly temperature dependent at high temperatures, which means that platelets which are thick enough for negligible error below room temperature are no longer adequate. Instead of trying to perform a self-consistent calculation of the high-temperature out-of-plane resistivity in these platelets, we took an empirical approach of studying a series of samples of increasing thickness until the resulting resistivity became independent of the thickness of the sample. The results are shown in Fig. 2. For the sample with the lowest apparent resistivity at 1300 K, $L_c^i/L_{ab}^i<5$ at all temperatures above 50 K, while for the thickest samples $L_c^i/L_{ab}^i>5$ even at 1300 K.

The intrinsic out-of-plane resistivity in Sr_2RuO_4 is shown in Fig. 3. By 1300 K, the resistive anisotropy is only a factor of 20, compared with approximately 1400 in these samples below 20 K.[17] A surprising feature of the results, shown in more detail in the inset to Fig. 3, is the rise in the out-of-plane resistivity between 700 and 1300 K. A point of debate in the cuprates has been the widespread observation of such metalliclike temperature dependences in materials in which estimates of the in-plane scattering rate and anisotropy strongly suggest that there is no coherent band formation perpendicular to the planes. Incoherent conductivity, however, is expected to increase with increasing temperature, because of thermally assisted hopping processes. The decreasing conductivity seen in some cuprates has led to suggestions that there must, somehow, be coherent band formation. In the present work, ''metallic'' conduction has been observed in a regime in which there seems to be no possibility of arguing for coherent band formation. It seems likely that the observations are related to effects seen in $La_{2-x}Sr_xCuO_4$.[20] In that material, the out-of-plane conductivity was shown to be strongly pressure dependent, and so very sensitive to the value of the lattice parameter. By combining the pressure measurements with knowledge of the thermal expansion of the lattice parameter, Nakamura *et al.* showed that the expansion correction to the measured temperature dependence of the out-of-plane resistivity can be large.[20] In Sr_2RuO_4, no data for the pressure dependence of the resistivity in the temperature range of interest are available, but it certainly seems plausible that our observations have a similar origin. In any case, the work presented here highlights the difficulty of relying on the observed temperature dependence of the out-of-plane resistivity when drawing conclusions about coherent band formation.

The overall picture of Sr_2RuO_4 that emerges from these measurements is that of a well-understood, highly anisotropic metal at low temperatures, making an apparently smooth crossover to a much more isotropic material with very poorly understood metallic conduction at high temperatures. Conventional energy bands exist in all directions below 20–30 K,[12] but even in-plane, these must be destroyed by 1000 K. It is also interesting to see that pronounced features in the out-of-plane resistivity, notably the maximum near 130 K, seem to have little or no effect on the in-plane resistivity at the same temperature.[21] The only point at which behavior in all directions has an obvious correlation is when the T^2 scattering rate is lost at 20–30 K. The nonquadratic scattering rates that appear in all directions are different, so the temperature-independent anisotropy that is often taken to be an important feature of an anisotropic three-dimensional metal is also lost with this characteristic temperature. This point seems worthy of further investigation.

The smooth increase of the in-plane resistivity through the Mott-Ioffe-Regel limit is particularly interesting. The fact that this kind of behavior can be observed in a material which is known to be a very good metal at low temperatures emphasizes that our current understanding of high-temperature conduction processes is very poor indeed. Other ''bad metals'' have ground states which are either superconducting with relatively high critical fields (e.g., the cuprates and alkali-doped C_{60}) or relatively poor metals with interesting magnetic behavior (e.g., manganites at some dopings or $SrRuO_3$). It has not been possible so far to confirm a conventional low-temperature metallic state by the observation of quantum oscillations in any of these materials, so the current observations on Sr_2RuO_4 clarify the problem that needs to be understood. There is now at least one example of a material in which a very smooth crossover from standard to highly nonstandard conduction processes is confirmed to exist.

While writing this manuscript we saw the results of an independent study of the resistivity of Sr_2RuO_4 to 1000 K by Berger *et al.*,[22] in which they report significantly different temperature dependences to those presented here. However, as emphasized by the authors of Ref. 22, their flux-grown crystals have high levels of disorder, and show neither a Fermi-liquid-like T^2 scattering rate nor a superconducting transition at low temperatures. We are confident that the present data accurately reflect the transport behavior of pristine stoichiometric Sr_2RuO_4.

We thank P. B. Allen, Y. Ando, D. G. Clarke, V. J. Emery, Z. Fisk, L. Forro, S. R. Julian, S. Kivelson, A. J. Millis, and H. Takagi for interesting discussions, and R. Smith for making all the silica parts for the high-temperature probe. In designing the probe, we benefited from earlier work by A. Carrington and J. R. Cooper. We gratefully acknowledge the support of the Royal Society and EPSRC (UK) and CREST-JST (Japan).

---

[1] A. F. Ioffe and A. R. Regel, Prog. Semicond. **4**, 237 (1960).

[2] N. F. Mott, Philos. Mag. **26**, 1015 (1972).

[3] Z. Fisk and G. W. Webb, Phys. Rev. Lett. **36**, 1084 (1976).

[4] M. Gurvitch, in *Superconductivity in d- and f-band Metals,* edited by H. Suhl and M. B. Maple (Academic, New York, 1980).

[5] P. B. Allen, Inst. Phys. Conf. Ser. **55**, 425 (1980), and references therein.

[6] P. B. Allen (private communication).

[7] C. C. Yu and P. W. Anderson, Phys. Rev. B **29**, 6165 (1984).

[8] P. B. Allen, in *Superconductivity in d- and f-band Metals* (Ref. 4).

[9] V. J. Emery and S. A. Kivelson, Phys. Rev. Lett. **74**, 3253 (1995), and references therein.

[10] For example, see L. Klein, J. S. Dodge, C. H. Ahn, G. J. Snyder, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, Phys. Rev. Lett. **77**, 2774 (1996).

[11] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednorz, and F. Lichtenberg, Nature (London) **372**, 532 (1994).

[12] A. P. Mackenzie, S. R. Julian, A. J. Diver, G. J. McMullan, M. P. Ray, G. G. Lonzarich, Y. Maeno, S. Nishizaki, and T. Fujita, Phys. Rev. Lett. **76**, 3786 (1996).

[13] A. P. Mackenzie, S. R. Julian, A. J. Diver, G. G. Lonzarich, N. E. Hussey, Y. Maeno, S. Nishizaki, and T. Fujita, Physica C **263**, 510 (1996).

[14] We are grateful to Y. Ando for supplying us with samples of this paint.

[15] In calculating resistivities throughout this paper, no correction was made for thermal expansion of the lattice.

[16] Y. Maeno, K. Yoshida, H. Hashimoto, S. Nishizaki, S. Ikeda, M. Nohara, T. Fujita, A. P. Mackenzie, N. E. Hussey, J. G. Bednorz, and F. Lichtenberg, J. Phys. Soc. Jpn. **66**, 1405 (1997).

[17] N. E. Hussey, A. P. Mackenzie, J. R. Cooper, S. Nishizaki, Y. Maeno, and T. Fujita, Phys. Rev. B **57**, 5505 (1998).

[18] L. J. van der Pauw, Philips Res. Rep. **13**, 1 (1958).

[19] H. C. Montgomery, J. Appl. Phys. **42**, 2971 (1971).

[20] F. Nakamura, M. Kodama, S. Sakita, Y. Maeno, T. Fujita, T. Takahashi, and N. Mori, Phys. Rev. B **54**, 10 061 (1996).

[21] The correlation between in- and out-of-plane resistivity reported in Ref. 11 has since been shown to be due to inhomogeneous current paths. The intrinsic behavior is reported in Ref. 16 and this paper, as well as Lichtenberg *et al.*, Appl. Phys. Lett. **60**, 1138 (1992).

[22] H. Berger, L. Forro, and D. Pavuna, Europhys. Lett. **41**, 531 (1998).