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Linear Resistivity from ~1 to 1050 K in $Sr_2RuO_{4-\delta}$ Single Crystals Grown by the Flux Technique

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Abstract

We present the resistivity measurements on single crystals of $Sr_2RuO_{4-\delta}$, grown by the flux technique. We discuss the striking, linear temperature dependence of resistivity that persists up to ~1050 K. The superconductivity in this system is confined below 1 K. This result suggests that the non-Fermi-liquid-alike linear temperature dependence of resistivity is not an exclusive signature of the anomalous normal state of high-T_c *cuprates but rather of layered oxides in general, especially perovskites, possibly independently of the magnitude of the superconducting temperature. In addition, such $Sr_2RuO_{4-\delta}$ samples, suitably optimized, may be used as broad range thermometers.*

Keywords: electrical resistivity, superconductivity, perovskites.

1 Introduction

It was the discovery of superconductivity <1 K in the Sr_2RuO_4 compound that has renewed the interest in ruthanates, especially as it is the first copper free layered perovskite superconductor.[1,2] It is well established that layered copper perovskites, in addition to high critical temperatures (T_cs up to 133 K; up to ≈160 K under pressure), also exhibit rather anomalous normal state properties (Refs. 3 and 4; reviews in Refs. 5–7 and references therein). One such 'anomaly' is the linear temperature dependence of resistivity: for example, the $Bi_2Sr_2CuO_6$ (T_c~8 K) and $La_{2-x}Sr_xCuO_4$ (T_c~30 K) exhibit linear resistivity up to 700 K[4] and 1100 K,[3] respectively. The understanding of electronic properties of high-T_c cuprates and of their corresponding electronic phase diagram still presents a major challenge, despite remarkable progress in both, sample preparation and advanced experimental techniques. However, there are also many superconducting oxides with rather low T_c < 15 K; for example, doped $SrTiO_{3-x}$, K_xWO_3, $LiTi_2O_4$, $Ba(PbBi)O_3$. So, given the recent enhanced interest in ruthanates it is timely to consider the electronic characteristics of all of these solids.

In this paper we discuss anomalous linear resistivity of single crystals of $Sr_2RuO_{4-\delta}$, grown by the flux technique. The resistivity persists up to ~1050 K in this material, while the superconductivity remains confined below 1 K. This suggests that (i) the linear $\rho(T)$ (non-Fermi liquid) behavior exists also in non-cuprate perovskites, and (ii) the linear temperature dependence of resistivity is not an exclusive signature of the unusual normal state of high-T_c cuprates but rather of layered oxides in general, possibly independently of the existence of superconductivity. Finally, suitably optimized $Sr_2RuO_{4-\delta}$ compound could be used as a broad range thermometer over three decades of temperatures covering low as well as high temperature regime.

2 Results and Discussion

Structurally the metallic Sr_2RuO_4 belongs to the same tetragonal K_2NiF_4 family as the aforementioned $La_{2-x}Sr_xCuO_4$ superconductor, in contrast to the ferromagnetic $SrRuO_3$ that has nearly cubic perovskite structures.[8] Both compounds have been extensively studied by a variety of techniques[9–11] and recent quantum oscillations studies in the normal state of Sr_2RuO_4 show the form of the quasiparticle spectrum that may be consistently interpreted in terms of a two-dimensional Fermi liquid.[11] The anomalous transport properties of

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cuprates have been extensively discussed in the literature.[3–5,8,12–14] Transport and electronic properties of ferromagnetic $SrRuO_3$ single crystals were discussed at length elsewhere[8] so here we concentrate on the linear resistivity of $Sr_2RuO_{4-\delta}$ over three decades of temperatures. Our Hall effect data have been reported elsewhere.[15]

The single crystals of approximate dimensions $0{\cdot}5\times0{\cdot}5\times0{\cdot}05\,mm^3$ were prepared by mixing highest purity chemical constituents, and in particular the $SrCl_2$ into the stoichiometric mixture. The crystal growth was done in Pt crucible by the flux technique[8] at $\sim$1300°C and subsequent slow cooling down to about 300°C. The X-ray analysis showed no traces of secondary phases. Detailed characterization of our samples shows that there is less than 1 ppm Pt or other impurities in our samples. However, we note that our growth technique is different from that used by Maeno *et al.*[12] and MacKenzie *et al.*[11,16] Their results were obtained on samples grown by the state-of-the-art zone melting, a technique that enables the growth of very clean samples, i.e. the degree of growth induced disorder is much smaller than in our case, with 'residual' resistivities of $\sim$1 and $\sim$50 $\mu\Omega$cm, respectively. This, as we shall see, is important for phenomenological understanding of our results.

For clarity of presentation we refer to our samples as $Sr_2RuO_{4-\delta}$, and henceforth introduce the growth induced disorder into the chemical formula. The degree of uncertainty in the exact content and distribution of oxygen is given by the '$4-\delta$', as is often done in the case of HTSC cuprates. Of course, there are other sources of disorder in our $Sr_2RuO_{4-\delta}$ samples; this is true also in the case of $La_{2-x}Sr_xCuO_4$ superconductors or any K_2NiF_4 type compound, in general. This simplification does not alter our transport results but simplifies the discussion and main conclusions.

In Fig. 1. we show results obtained on three single crystals of $Sr_2RuO_{4-\delta}$ that all show essentially linear slope of electrical resistivity up to room temperature. In particular we emphasize the lack of saturation at low temperatures in contrast to the results from Refs. 1, 11 and 16. As we already mentioned, this difference is mainly due to higher disorder in our samples, due to unavoidable differences in sample preparation technique and detailed treatment during and after the growth. Our inferred residual resistivity is $\sim$50 $\mu\Omega$cm and consequently we estimate our elastic mean free path to be rather short: only of the order of $\sim$1 nm. This is important as it probably implies that by introducing growth induced disorder (and oxygen vacancies) the consequent scattering seems to effectively 'induce' linear resistivity behavior even at low temperatures. At present it is not clear whether this

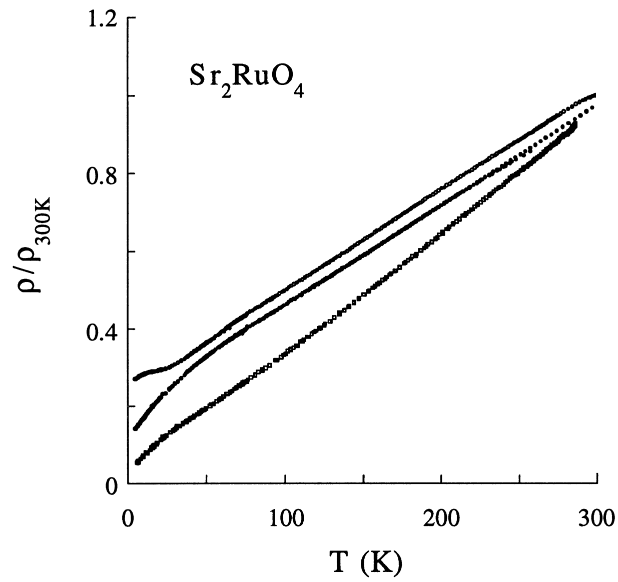

Fig. 1. Temperature dependence of resistivity for three different single crystals of $Sr_2RuO_{4-\delta}$ that show linear slope of resistivity up to room temperature. The middle sample was measured up to $\sim$1050 K (see Fig. 2).

linearity masks the Fermi-liquid T^2-term that may remain 'hidden' underneath the disorder scattering; further studies are needed to clarify this point.

Figure 2 shows the temperature dependence of resistivity $Sr_2RuO_{4-\delta}$ single crystal up to 1050 K. The slope above 750 K could be fitted with a power of 1·08; even in this range it is practically linear. We note that such linear $\rho(T)$ behavior in $Sr_2RuO_{4-\delta}$ up to $\sim10^3$ K closely resembles the linear resistivity reported[3] for single CuO_2-layer superconducting $La_{2-x}Sr_xCuO_4$ ($T_c\sim30$ K): lin-

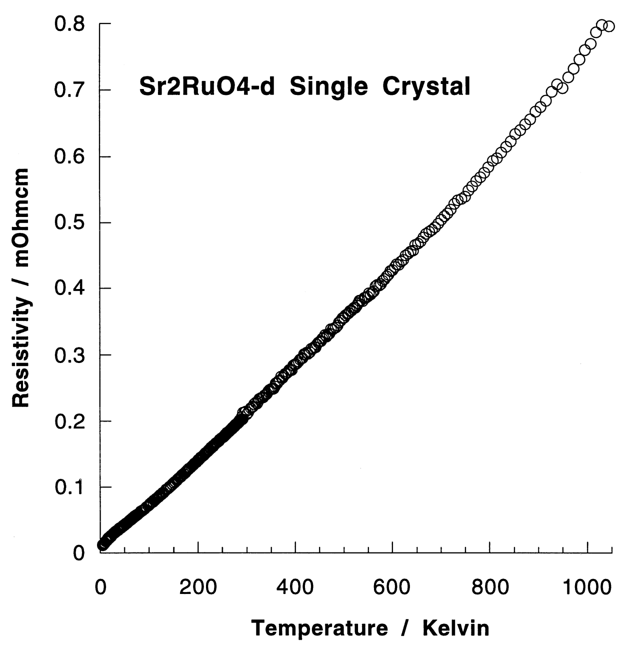

Fig. 2. Linear resistivity of $Sr_2RuO_{4-\delta}$ single crystal grown by the flux technique.

ear from ~40 to 1100 K; YBCO compound easily loses oxygen above 700 K so the resistivity suddenly increases (see Fig. 3). Our result can therefore be considered as an extension of initial reports on anomalous linear resistivities in high-T_c cuprates:[3] however, nowadays we know and therefore argue that this is not a unique feature of the high-T_c cuprates only.

It is striking to see any physical quantity exhibit linear behavior over three decades of temperature. For example, ohmic junctions show linear I–V response but only over a limited range; we were not able to find in literature examples of physical quantities that show essentially straight line response at very low and at rather high temperatures. In the simplest approximation it means that the electron scattering gives this linear resistivity over the energy range of ~1 meV up to ~0·1 eV which is again striking and requires an indepth theoretical analysis; this certainly cannot be understood in terms of the conventional transport theory in metallic solids.

Our linear resistivity down to low temperatures does not contradict the results of MacKenzie *et al.*[16] who have measured T^2 dependence of the *ab*-plane resistivity. Their transport results, and comprehensive study of magneto-oscillatory phenomena in the normal state,[11] show that their sample behaved as a rather convincing example of the Landau–Fermi liquid metal, in this case the Sr_2RuO_4 single crystal grown by zone melting technique. Our results show that even in the case of such a clean metallic oxide one can increase the 'residual' scattering and consequently observe the resistive behavior that is usually measured mainly in somewhat 'underdoped' HTSC cuprates. In the language of the well established HTSC phase diagram, the resistivity of our samples resembles behavior previously seen in cuprates close to optimum doping, or slightly 'underdoped'[3] while the

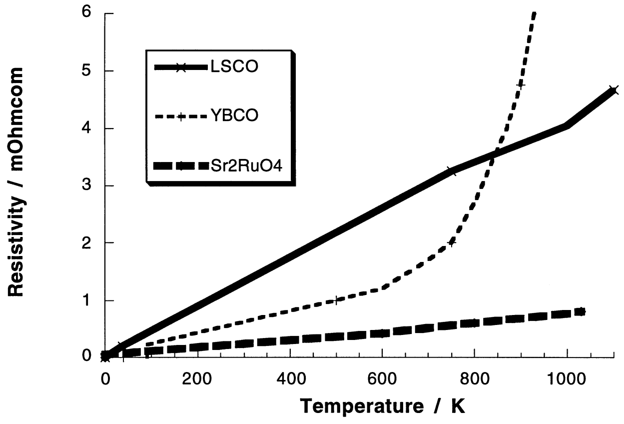

Fig. 3. Linear resistivity of $Sr_2RuO_{4-\delta}$ single crystals grown by the flux technique[5–7] compared to resistivities of ceramic sample of LSCO and YBCO high-T_c cuprate perovskites.[3]

samples of MacKenzie *et al.*[11] behave more like 'overdoped' cuprates.[16] Similar, more 'conventional' metallic behavior, with T^2 behavior at low temperatures, was also measured in electron doped superconductors, for example in $Nd_{2-x}Sr_xCO_4$ compound.[13]

At present we are unaware of any model that predicts or can convincingly account for essentially linear behavior of resistivity over three decades of temperatures and we have no coherent theoretical explanation ourselves. In our view, this result provides yet another puzzle to our present understanding of transport and normal state of layered oxides and suggests that: (i) the anomalies, like linear resistivities exist also in low-T_c layered oxides and probably even in non-superconducting phases, and (ii) the presence of CuO_2 planes is vital for truly high T_cs (of > 50 K) as high critical temperatures are still measured only in cuprates.

Finally, we note that the reflectance, ellipsometric and Raman spectra obtained by Bozovic *et al.*[17] on thin films of isotropic metallic oxides $Ca_{0.5}Sr_{0.5}RuO_3$ and $La_{0.5}Sr_{0.5}CoO_3$ closely resemble the spectra of high-T_c cuprates thus indicating that the 'anomalous' dielectric response could not be the sole root of high temperature superconductivity.[14] Most likely one needs a thorough understanding of all layered oxides in order to understand the normal and superconducting state anomalies of high-T_c cuprates.

3 Conclusions

In conclusion we have measured linear resistivity behavior that persists up to ~10^3 K in single crystals of $Sr_2RuO_{4-\delta}$ perovskite, grown by flux technique, in which the superconductivity remains confined to very low temperatures < 1 K. This suggests that the non-Fermi-liquid-alike temperature dependence of resistivity is not an exclusive signature of the unusual normal state of high-T_c cuprates but rather of layered oxides in general and especially perovskites.

Further studies, and especially in-depth theoretical analysis, are needed to account for such unusual resistivity behavior over three energy scales. On the other hand, with suitable processing optimization, such materials can be developed as very useful, broad range temperature thermometers.

Acknowledgements

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