Linear Field Dependence of the Normal-State In-Plane Magnetoresistance of Sr$_2$RuO$_4$

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The transverse and longitudinal in-plane magnetoresistances in the normal state of superconducting Sr$_2$RuO$_4$ single crystals have been measured. At low temperatures, both of them were found to be positive with a linear magnetic-field dependence above a threshold field, a result not expected from electronic band theory. We argue that such behavior is a manifestation of a novel coherent state characterized by a spin pseudo gap in the quasi-particle excitation spectrum in Sr$_2$RuO$_4$.

74.70.-b, 74.25.Fy, 74.25.Ha, 74.72.Yg

Since the discovery of superconductivity in Sr$_2$RuO$_4$, isostructural with high-$T_c$ cuprate (La,Sr)$_2$CuO$_4$, this material has emerged as a new focus of superconductivity research. There has been a growing body of experimental evidence for a spin triplet (odd-parity) pairing in Sr$_2$RuO$_4$ including results obtained in muon spin relaxation, NMR Knight shift, electrical transport, proximity effect, and specific heat measurements. In the normal state, a highly anisotropic Fermi liquid behavior, characterized by a $T^2$-dependence in the $a$- and out-of-plane resistivity ($\rho_a$ and $\rho_c$, respectively) and a cylindrical Fermi surface was found for Sr$_2$RuO$_4$ at low temperatures. The enhancements of electronic specific heat and spin susceptibility are very similar to those found in $^3$He, suggesting that Sr$_2$RuO$_4$ is a strongly correlated system. At higher temperatures, however, the situation is not very clear. While $\rho_c$ undergoes a non-metallic ($\mathrm{d}\rho_c/\mathrm{d}T<0$) to metallic ($\mathrm{d}\rho_c/\mathrm{d}T>0$) crossover around 130K a similar feature is not seen in $\rho_{ab}$, a phenomenon yet to be fully understood.

Chemically, Sr$_2$RuO$_4$ is positioned in the vicinity of both ferromagnetic and antiferromagnetic orderings. In the Ruddlesden-Popper homologous series Sr$_{n+1}$Ru$_n$O$_{3n+1}$, as the single-layer ($n=1$) member, Sr$_2$RuO$_4$ is a paramagnetic metal with no apparent local moment. The infinite-layer ($n=\infty$) member, SrRuO$_3$, is a ferromagnetic metal. The $n=2$ member of the series, Sr$_3$Ru$_2$O$_7$, which has been reported to be ferromagnetic by one group and antiferromagnetic by others, should be magnetic as well. On the other hand, if Sr$^{2+}$ ions in Sr$_2$RuO$_4$ are replaced by Ca$^{2+}$, the resultant compound Ca$_2$RuO$_4$, which is isostructural and isoelectronic to Sr$_2$RuO$_4$, is an antiferromagnetic insulator. Being in the vicinity of these very different behaviours, Sr$_2$RuO$_4$ is likely to exhibit unusual electronic and magnetic properties.

The normal-state properties of Sr$_2$RuO$_4$ can be probed by magnetoresistance (MR) measurements. The normal-state MR of Sr$_2$RuO$_4$ was first measured at 20mK in the context of determining the Fermi surface of this material through the Shubnikhov-de Haas oscillations. However, no field dependence of MR was identified in that work. More recently, Hussey et al. have measured both $c$-axis and in-plane normal-state MR of Sr$_2$RuO$_4$, focusing primarily on the $c$-axis MR ($\Delta\rho_c/\rho_{c}(H) - \rho_{c}(0)/\rho_{c}(0)$) at relatively high temperatures. The in-plane MR ($\Delta\rho_{ab}/\rho_{ab} = (\rho_{ab}(H) - \rho_{ab}(0))/\rho_{ab}(0)$) was measured only in transverse configuration for one sample, without a detailed analysis of its field dependence over the entire temperature range. Longitudinal in-plane MR results were not available in literature prior to the present study. In this paper, we report results of our systematic study of the normal-state in-plane MR for Sr$_2$RuO$_4$ in both transverse and longitudinal configurations. Our work has revealed previously unknown features in in-plane MR, strongly suggesting a novel behavior in the normal state of Sr$_2$RuO$_4$.

Single crystals used in this study were grown by a floating-zone method with details described elsewhere. Resistance measurements show a superconducting transition temperature ($T_c$) of 0.84K, which is in agreement with previous reports. The temperature was measured using a Lakeshore Cernox 1030 thermometer with relative temperature corrections, due to magnetic field, typically 0.15% at 4.2 K and -0.023% at 77.8K and 8.0T. For in-plane MR and Hall measurements, we used four rectangular shaped single crystals with dimensions around $1.2 \times 1.0 \times 0.01$, $0.9 \times 0.4 \times 0.08$, $0.8 \times 0.2 \times 0.07$, and $0.6 \times 0.3 \times 0.05$mm$^3$, respectively. For each sample, two current contacts covering the opposite ends and four voltage contacts on the two sides of the crystal were prepared.

All RuO$_2$ layers were electrically shorted along the $c$-axis to ensure a homogeneous current distribution. For transverse and longitudinal MR measurements, the magnetic field $H$ was applied perpendicular and parallel to the injected current $I$, respectively. In order to exclude the Hall contribution to MR, only the symmetric part of $\Delta\rho_{ab}(H) = \rho_{ab}(H) - \rho_{ab}(0)$ under field reversal was included. For in-plane Hall measurements, the magnetic field was applied parallel to $c$-axis with a current bias applied along the $ab$-plane. The Hall voltage $V_H$, which contains only the asymmetric contributions under...
the field reversal, was found to vary linearly with \( H \) up to 5T. By fitting \( V(H) \) data in the linear regime using \( V(H) = R_H \cdot H \cdot I/d \) (d is the thickness of the sample), the in-plane Hall coefficient \( R_H \) was obtained.

The transverse in-plane MR, \( \Delta \rho_{ab}/\rho_{ab} \) (\( H \perp I \)), between 0 and 7.3T at various temperatures is shown in Fig. [1]. Similar results have been obtained in separate samples. \( \Delta \rho_{ab}/\rho_{ab} \) is seen to be positive, growing rapidly in magnitude with decreasing \( T \). At low fields, \( \Delta \rho_{ab}/\rho_{ab} \) at a fixed temperature is very small, which can be described by the sum of a linear and a quadratic term. When \( H \) exceeds a threshold value \( H_0 \), \( \Delta \rho_{ab}/\rho_{ab} = K_S (H - H_0) \), where \( K_S \) and \( H_0 \) are functions of temperature. The solid lines in Fig. [1] represent fits to the data above \( H_0 \) using this form. The temperature dependence of \( H_0 \) is shown in the inset of Fig. [1]. We note that in Ref. 20, results on \( \Delta \rho_{ab}/\rho_{ab} \), plotted against \( H^2 \), were presented. While \( \Delta \rho_{ab}/\rho_{ab} \) might be quadratic in \( H \) at the two highest temperatures shown (56K and 82K), a deviation from this behavior can be seen at all other temperatures. (We have re-plotted the low temperature data against \( H \) and found both the magnitude and the field dependence of \( \Delta \rho_{ab}/\rho_{ab} \) given in Ref. 20 in agreement with those obtained in the present study at overlapping temperatures.)

Within the band theory for solids, MR is proportional to \((H/\rho(0))^2\) in the low-field limit. Linear MR can be expected only in some special circumstances. For example, a square Fermi surface can lead to linear MR due to the presence of the sharp corners. Local-density approximation calculations have shown that the Fermi surface of \( \text{Sr}_2\text{RuO}_4 \) consists of three roughly cylindrical-like sheets with no such sharp corners, thus excluding this from being the origin of the observed linear MR. For systems with multi-band electronic structure involving two types of charge carriers, the field dependence of MR can be written as \( \Delta \rho/\rho(0) = aH^2/(b + cH^2) \) where \( a, b, \) and \( c \) are positive, field-independent quantities determined by the relaxation rates of each type of carriers. It is clear that this expression will not lead to \( \Delta \rho/\rho(0) \sim H \) as observed experimentally. In some elemental metals (such as K, In, and Al), linear MR has been found and attributed to the boundary and disorder effects. Since superconductivity in \( \text{Sr}_2\text{RuO}_4 \) is extremely sensitive to the amount of disorder, the linear \( \Delta \rho_{ab}/\rho_{ab} \), found reproducibly in superconducting \( \text{Sr}_2\text{RuO}_4 \) with \( \rho(0) \approx 1 \mu\Omega \cdot \text{cm} \), is unlikely due to effects of the impurities and/or structural defects.

Linear MR was observed previously in heavy fermion \( \text{CeCu}_6 \) at very low temperatures. In another heavy fermion compound, \( \text{UPt}_3 \), the MR was found to be positive and proportional to \( H^2 \). The observed linear MR was interpreted as being a consequence of the opening of a pseudo gap in a coherent state formed at low temperatures. In this picture, the pseudo gap is suppressed by the magnetic field, resulting in enhanced scattering rates and therefore positive MR. NMR studies of \( \text{Sr}_2\text{RuO}_4 \) provided the first hint that a coherent normal state may also be present in this material at low temperatures. The emergence of such a state is signaled by a sharp change of slope in \( 1/T_1 \) vs. temperature curve for both \( ^{101}\text{Ru} \) and planar \( ^{17}\text{O} \) sites around 80K. Our in-plane MR results also indicate that this temperature is special for \( \text{Sr}_2\text{RuO}_4 \). As shown in Fig. [2], a sign change from positive to negative in \( \Delta \rho_{ab}/\rho_{ab} \) is seen between 7-80 K, similar to what has been observed in c-axis MR. It should be noted that the range of the data scattering in \( \Delta \rho_{ab}/\rho_{ab} \) at 7.3 T is around 0.004%. The corrections in temperature due to the MR of the thermometer is around -0.016% at 87 K and 8.0 T, which cannot lead to the observed sign change in \( \Delta \rho_{ab}/\rho_{ab} \). While the physical origin of the sign reversal in MR has not been understood at the present time, it is not inconceivable that this is associated with the emergence of a coherent normal state in \( \text{Sr}_2\text{RuO}_4 \) at low temperatures. As will be argued later, such a state may be accompanied by the opening of a pseudo gap in the quasi-particle excitation spectrum. Similar to \( \text{CeCu}_6 \), an applied magnetic field suppresses the pseudo gap, leading to linear in-plane MR for \( \text{Sr}_2\text{RuO}_4 \).

The in-plane Hall coefficient \( R_H \) of \( \text{Sr}_2\text{RuO}_4 \), should be subject to the same scattering processes as that for the in-plane MR. An interesting question is whether \( R_H \) will reflect the presence of a pseudo gap in the quasi-particle excitation spectrum. The in-plane \( R_H \) was measured for one of our samples in the same magnetic field and current configuration as that for the transverse in-plane MR. The results are shown in Fig. [3]. The Hall coefficient is negative at high temperatures, changing its sign around 130K, which is roughly where \( \rho_c \) undergoes a nonmetallic-metallic crossover. It reaches a maximum between 70-80K before changing its sign again around 30K. Prior to the present work, the Hall coefficient \( R_H \) of \( \text{Sr}_2\text{RuO}_4 \) has been measured on crystals prepared by different groups. Our results agree well with those published earlier, indicating that features shown in \( R_H(T) \) are intrinsic. In Ref. 19, an isotropic-\( \ell \) approximation within the multi-band picture of \( \text{Sr}_2\text{RuO}_4 \) was used to calculate the low temperature limit (\( T < 1 \)K) of \( R_H \) where the isotropic-\( \ell \) approximation did not seem to work. It is interesting to note that the characteristic temperature where \( R_H \) shows a maximum coincides with that below which we believe the pseudo gap emerges. Whether the maximum in \( R_H \) is another signature for the opening of a pseudo gap in \( \text{Sr}_2\text{RuO}_4 \), and how strongly the presence of such a gap will affect \( R_H \), are issues to be resolved.

In order to infer the relative contribution of the orbital and spin motions to MR and the physical origin of this pseudo gap, we have measured the longitudinal in-plane
MR of Sr$_2$RuO$_4$. As shown in Fig. 4, the longitudinal in-plane MR $\Delta \rho^\parallel_{\rho ab}$ also revealed a linear behavior above $H_0$, which increases with increasing temperature (see the inset of Fig. 4). More striking is that the magnitude of $\Delta \rho^\parallel_{\rho ab}$ is greater than that of $\Delta \rho^\perp_{\rho ab}$ below approximately 10K, indicating that the spin contribution to the in-plane MR is important. Therefore, the pseudo gap may be of spin origin. Results obtained in NMR $^{17}$O Knight shift measurements in Sr$_2$RuO$_4$ point to the same direction. When the $^{17}$O Knight shift results were decomposed into spin susceptibilities of three Ru d-orbitals, $d_{xy}$, $d_{xz}$, and $d_{yz}$, a broad maximum around 40K was found. This is consistent with uniform susceptibility results where a broad peak was seen around the same temperature. The decrease of spin susceptibility at low temperatures may then be taken as an indication of the opening of a pseudo gap in the spin excitation spectrum.

It has been suggested theoretically that the spin fluctuations in Sr$_2$RuO$_4$ are predominantly ferromagnetic. Experimentally, $1/T_1$ obtained from the NMR measurements for both O(1) and Ru sites shows an identical temperature dependence. In general, $1/T_1$ can be expressed by the sum of $q$-dependent imaginary part of the dynamical electron susceptibility with appropriate form factors, which should have different $q$-dependence for $^{195}$Ru and planar $^{17}$O sites, leading to different temperature dependence in their respective $1/T_1$. However, if the dynamical electron susceptibility has a sharp peak around $q = (0, 0)$, corresponding to ferromagnetic fluctuations, then the above mentioned behavior in $1/T_1$ can be explained. On the other hand, this scenario will lead to negative MR, which contradicts the experimental observation of positive MR.

Results obtained in inelastic neutron scattering (INS) measurements carried out recently on Sr$_2$RuO$_4$ have resolved this apparent contradiction between NMR and magnetotransport results. In this experiment, incommensurate magnetic spin fluctuations located at $q_0 = (\pm 0.6a, \pm 0.6/a, 0)$, originally predicted by band calculations were found at low temperatures. The intrinsic $q$-width for these incommensurate fluctuations was found to be narrow ($\Delta q = 0.13 \pm 0.02\text{Å}$ at 10K). The temperature dependence of $1/T_1$ obtained in NMR can be reconstructed from the INS results, confirming that the dominating spin fluctuations are incommensurate rather than ferromagnetic as suggested in Ref. 26. The energy dependence of the imaginary part of the dynamic susceptibility at $q_0$, which measures the dissipation of the spin fluctuations, shows a gradual drop below 7meV, indicating the presence of a gap in the spin excitation spectrum. We note here that this energy scale (7 meV) agrees well with the value of the pseudo gap inferred from the temperature dependence of other physical quantities such as $1/T_1$ and MR.

Physical insight into the nature of the normal state of Sr$_2$RuO$_4$ can be obtained from the high-pressure experiments. It was shown that the superconducting transition temperature $T_c$ was suppressed by an applied pressure $p$. By extrapolating results obtained up to 1.2 GPa, $T_c$ is expected to become zero around $p_c \approx 3$ GPa. In normal state, $\rho_{ab}$ was found to vary non-monotonically with $p$ at low $T$. With increasing $p$, $\rho_{ab}$ first increases, and then decreases as $p$ is increased further above a threshold value, close to 3 GPa. At 8 GPa, $\rho_{ab}$ was found to show a $T^4$-$\rho$ dependence, characteristic of a two-dimensional electronic system close to a ferromagnetic quantum critical point. Based on this, we propose the following phase diagram for Sr$_2$RuO$_4$, shown schematically in Fig. 4. At $T = 0 K$, a direct superconductor (SC)-to-ferromagnetic metal (FM) transition, tuned by pressure, occurs at $p = p_c \approx 3$ GPa. At low $T$ and low $p$, Sr$_2$RuO$_4$ is superconducting. A pseudo gap regime emerges above $T_c$, similar to the quantum spin disordered regime discussed in quantum critical phenomena. As $p$ is further increased, the pseudo gap is suppressed, leading to an increase in $\rho_{ab}$, as observed experimentally. When the system enters the fluctuating regime at a finite $T$, $\rho_{ab}$ decreases with the increasing $T$ since the system is driven towards a ferromagnetic state. We suggest that the magnetic field similarly suppresses the pseudo gap, resulting in a positive MR. In this picture, as $T$ increases, a larger threshold field ($H_0$) is needed to overcome the thermal smearing so that linear MR can be observed, consistent with our experimental observations.

A pseudo gap has been observed in high-$T_c$ cuprate superconductors. However, the origin and physical consequences of pseudo gap in high-$T_c$ materials may be different from that in Sr$_2$RuO$_4$. The in-plane MR observed in (La,Sr)$_2$RuO$_4$ is positive, but not linear in field dependence. When the pseudo gap opens, the Hall coefficient was found to become strongly temperature-dependent. A suppression in resistivity was found in underdoped (La,Sr)$_2$RuO$_4$ as the pseudo gap opened. A similar feature has not been observed in Sr$_2$RuO$_4$. As mentioned above, the pseudo gap in Sr$_2$RuO$_4$ is present only in a small portion of the Fermi surface. The magnitude of the gap is also small as compared with that of the high-$T_c$ cuprates, resulting in unobservable effects in $\rho_{ab}$ and $\rho_c$.

Finally we briefly mention our $\Delta \rho_c/\rho_c$ results which will be discussed in detail in a future publication. As mentioned above, previously published work on $\Delta \rho_c/\rho_c$ of Sr$_2$RuO$_4$ was focused on relatively high-temperature (>22 K) behavior although data obtained at 3.6 K were also shown. The magnitude of our longitudinal $c$-axis MR, $\Delta \rho^\parallel_{\rho c}/\rho_c$, is comparable with that of the transverse MR, $\Delta \rho^\perp_{\rho c}/\rho_c$. The difference, $\Delta \rho^\perp_{\rho c}/\rho_c - \Delta \rho^\parallel_{\rho c}/\rho_c$, the "pure" orbital MR, has very good $T^2$-dependence. A sign reversal from positive at low $T$ to negative at high $T$ was found in both $\Delta \rho^\perp_{\rho c}/\rho_c$ and $\Delta \rho^\parallel_{\rho c}/\rho_c$ around 75 K, consistent with previous observations. However, different from the previous study, we found that $\Delta \rho_c(H)/\rho_c = \alpha H^2 - \beta H^4$ (where $\alpha$ and $\beta$ are constants), actually de-
scribes well our $\Delta \rho_{ab}^\perp/\rho_c$ and $\Delta \rho_{ab}^\parallel/\rho_c$ data.\footnote{1}

In summary, we have studied the magnetic field dependence of in-plane MR of Sr$_2$RuO$_4$ up to 7.3T. The linear positive transverse and longitudinal in-plane MR, not expected from band theory, has been found above a threshold field at low temperatures. We argue that such behavior is a manifestation of a coherent pseudo gap state in Sr$_2$RuO$_4$ formed at low temperatures. Theoretical input is needed to fully understand the experimental results.

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FIG. 1. In-plane transverse MR, $\Delta \rho_{ab}^\perp/\rho_c$ (H $\perp$ ab, I $\parallel$ ab) for Sr$_2$RuO$_4$. The solid lines are linear fits to the data (see text). $H_0(T)$ is shown in the inset.

FIG. 2. In-plane transverse MR for Sr$_2$RuO$_4$ at temperatures close to 75K.

FIG. 3. In-plane Hall coefficient $R_{BH}(T)$ with a broad peak around 70-80K.

FIG. 4. In-plane longitudinal MR $\Delta \rho_{ab}^\parallel/\rho_c$ (H $\parallel$ ab, I $\parallel$ ab) for Sr$_2$RuO$_4$. The solid lines are linear fits to the data. $H_0(T)$ is shown in the inset. Note that $\Delta \rho_{ab}^\parallel/\rho_c$ is larger than $\Delta \rho_{ab}^\perp/\rho_c$ below 10K.

FIG. 5. Schematic phase diagram for Sr$_2$RuO$_4$, where SC denotes the superconducting while FM the ferromagnetic metal phase. A zero-temperature SC-FM transition at $p = p_c \approx 3$ GPa is indicated (see text).
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