Electronic crossover in the highly anisotropic normal state of Sr$_2$RuO$_4$
from pressure effects on electrical resistivity

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We investigated pressure dependence of the electrical resistivity of the only copper-free layered-perovskite superconductor Sr$_2$RuO$_4$. The characteristic temperature $T_M$, where the out-of-plane resistivity $\rho_c$ changes from metallic behavior to nonmetallic behavior with increasing temperature, increases with hydrostatic pressure. In contrast, the absolute value of $\rho_c$ at $T_M$ is almost independent of pressure. Such a change over in $\rho_c$ is ascribable to the crossover from the diffusion process of quasiparticles between the adjacent layers to the thermally assisted hopping process. The pressure effects on $\rho_c$ are possibly characterized by the enhanced transfer integral along the $c$ axis by pressure. On the other hand, in the temperature dependence of the in-plane resistivity, we observed an intriguing hump that appears at low $T$ less than $T_M$ in the pressure range from 1.5 to 5 GPa, and vanishes on applying higher pressure. This behavior may be comprehensible from the view of the two-dimensional ferromagnetic spin fluctuations enhanced by pressure. [S0163-1829(98)04246-5]

I. INTRODUCTION

Since the discovery of superconductivity in Sr$_2$RuO$_4$, this ruthenate has been attracting a lot of interest, in spite of its low critical temperature $T_c \sim 1$ K. Part of the interest is owing to the fact that this ruthenate is the first noncooper superconductor with a layered perovskite structure similar to high-$T_c$ cuprates. Reflecting its nearly cylindrical Fermi surfaces (FS) the superconducting state of Sr$_2$RuO$_4$ is highly anisotropic. The ratio of the coherence lengths $\xi_{ab}/\xi_c \sim 26$ (Ref. 5) is larger than that of its isostructural La$_{1.85}$Sr$_{0.15}$CuO$_4$.

Recently, both NMR (Ref. 6) and specific heat measurements showed that in Sr$_2$RuO$_4$ a finite density of states remains even at $T \rightarrow 0$ K. Mackenzie et al. reported a strong suppression of $T_c$ by nonmagnetic impurities. These results suggest unconventional superconductivity in this ruthenate. Some theorists have discussed the possibility of an exotic p-wave pairing, including a nonunitary state.

The normal-state properties of Sr$_2$RuO$_4$ are as fascinating as its superconductivity. A highly anisotropic Fermi-liquid model has been successfully adopted to describe the normal state. The quasi-two-dimensional electronic state is characterized by the peculiar anisotropic resistivity. The resistivity along the $c$ axis $\rho_c$ is nonmetallic ($d\rho_c/dT < 0$) at $T > T_M = 130$ K, but becomes metallic ($d\rho_c/dT > 0$) at $T < T_M$. In contrast, the resistivity in the $ab$ plane $\rho_{ab}$ exhibits the metallic superlinear dependence on $T$ over the measured temperatures and does not show any change across $T_M$. Therefore, there is a crossover at $T_M$ from a two-dimensional (2D) metal to a 3D metal with decreasing temperature.

In contrast with Sr$_2$RuO$_4$, $\rho_c$ of the lightly doped high-$T_c$ cuprates exhibits a steep semiconductorlike upturn at low temperatures, whereas $\rho_{ab}$ remains metallic. From the high-pressure measurements on optimally doped La$_{2-x}$Sr$_x$CuO$_4$, even the apparent metallic temperature dependence of $\rho_c$ at high temperatures is describable by such a nonmetallic process as a hopping between adjacent CuO$_2$ planes. This is the so-called confinement of the carriers within the $ab$ planes. As is widely accepted, such peculiar quasi-2D electronic states which is clearly reflected in $\rho_c$ is closely related with the appearance of high $T_c$.

In order to understand the anisotropic electronic states of Sr$_2$RuO$_4$, we have measured the normal-state resistivities $\rho_{ab}$ and $\rho_c$ under hydrostatic pressure up to 8 GPa. In addition to controlling the dimensionality or anisotropy, applying pressures serves to control the electronic state through adjusting the effective correlation energy $U/W$, where $U$ is the on-site Coulomb repulsion energy and $W$ is the one-particle band width.

Shirakawa et al. reported anisotropy in the pressure dependence of resistivity at room temperature up to 1.2 GPa, namely, the increase (+2%/GPa) of $\rho_c$ and the decrease ($-6%$/GPa) of $\rho_{ab}$. $T_c$ decreases with pressure and is expected to vanish at about 3 GPa. Presently, it is far from being clearly understood how such physical properties correlate with $U/W$, and hence we require higher pressure measurements.

II. EXPERIMENT

Single crystals Sr$_2$RuO$_4$ were grown by a floating-zone method using a commercial infrared furnace (NEC Machinery, SC-E15HD). The starting materials were 99.99% pure SrCO$_3$ and 99.9% pure Ru$_2$O$_3$. They were weighed in a nonstoichiometric molar ratio of Sr:Ru = 2:1.2. The excess Ru is added because of the high vapor pressure of Ru at high temperatures on growing crystals. The powders were ground in a dry-nitrogen atmosphere and the mixture was prereacted in air at 900 and 1150 °C for a total of 48 h with intermediate...
regrinding. After being pressed into a rod with a diameter of 6 mm, it was sintered in air at 1350 °C for 4 h. The growth of the crystals was performed in air with a feed speed of 20 mm/h. The crystals exhibit a tetragonal crystal structure and a $T_M$ of $~0.7 \, \text{K}$. The resistivity was measured using a standard four probe method, except for a ring contact geometry for $\rho_c$. The typical size of samples was $0.2 \times 0.6 \times 0.01 \, \text{mm}^3$ for $\rho_{ab}$ and $0.3 \times 0.3 \times 0.01 \, \text{mm}^3$ for $\rho_c$. The shortest dimension was along the $c$ axis. Silver paste (Dupont, 6838) was used for attaching electrodes and was cured in air at $500 \, \text{°C}$ for 5 min. We attained a contact resistance of $0.5 \, \Omega$.

The pressures up to 8 GPa was generated by using a cubic-anvil device with a pressure transmitting medium of a mixture of Fluorinert FC70 and FC77 (3M Co.). This device achieves nearly isotropic compression of the sample space, namely, quasi-hydrostatic pressure. First, we measured the pressures up to 8 GPa without being interrupted by any scattering process, 2,14 that is, the mean-free path along the $c$ axis $l_c$ is longer than $d$.19 The coherent metallic conduction along the $c$ axis is possible with the help of slight dispersions of the cylindrical Fermi surfaces.

III. RESULTS

A. Pressure dependence of out-of-plane resistivity $\rho_c$

Figure 1 shows the temperature dependence of $\rho_c$ at hydrostatic pressures of 0.1 MPa (ambient pressure), 1.5, 3.0, 5.0, and 8.0 GPa. Application of pressure was found to induce a pronounced change in the magnitude of $\rho_c$ and a shift in $T_M$. At low temperatures, we observed the quadratic temperature dependence of resistivity for any pressure: $\rho_c = \rho_{c0} + AT^2$, where $\rho_{c0}$ and $A$ are residual resistivity and a numerical coefficient, respectively. This behavior is ascribed by the fact that the quasiparticles with nearly 2D character around Fermi surfaces can travel the interlayer distance $d$ without being interrupted by any scattering process, 2,14 that is, the mean-free path along the $c$ axis $l_c$ is longer than $d$.19 The coherent metallic conduction along the $c$ axis is possible with the help of slight dispersions of the cylindrical Fermi surfaces.

We should note that, in contrast with the previous results up to 1.2 GPa, 18 in which changes in $\rho_{c0}$ and $A$ were not resolved, we found that both parameters systematically decrease at higher pressures. The characteristic temperature $T^*$, where $\rho_c$ deviates from the $T^2$ dependence, increases from 16 K (0.1 MPa) to 47 K (8.0 GPa), as shown in Fig. 2. Above $T^*$, $\rho_c$ is still metallic with $d\rho_c/dT > 0$, although the coherent conduction along the $c$ axis based on the band picture starts to break down owing to $l_c < d$. With further increasing temperature, $d\rho_c/dT$ decreases and finally changes the sign to negative at $T_M$. With pressure, the peak temperature $T_M$ remarkably increases from 134 K (0.1 MPa) to 210 K (8.0 GPa). It should be noted here that $\rho_c$ is $\approx 32 \, \mu\Omega \cdot \text{cm}$ at $T_M$ and the value is almost independent of applied pressures. This is reminiscent of the Mott-Ioffe-Regal maximum metallic resistivity $\rho_{max}$. 20 In fact, it is very interesting that the value is almost equal to $\rho_{max} = h/e^2k_F$, where $h$ is the Plank constant, if $k_F$ is replaced by the dispersion along the $c$ axis for a cylindrical Fermi surface of Sr$_2$RuO$_4$. $\Delta k_F(\beta - \text{FS}) = 0.008 \, \text{Å}^{-1}$ at ambient pressure. 2 Here, the $\beta$-Fermi surface has the largest $\Delta k_F$ among the three cylindrical FS sheets. As far as we know, such a pressure-independent value for the metal-nonmetal crossover has never been reported. This is intimately connected with the unusual increase (+13% at 8 GPa) of $\rho_c$ at 295.5 K with applying pressure as shown in the inset of Fig. 1, which is in agreement with results by Shirakawa et al. 18

B. Pressure dependence of in-plane resistivity $\rho_{ab}$

Pressure effects on $\rho_{ab}$ are qualitatively different from those on $\rho_c$, as shown in Fig. 3. The in-plane $\rho_{ab}$ shows metallic behavior at any pressure over the whole temperature range. No anomaly is observed in $\rho_{ab}$ at $T_M$. An interesting finding is that a hump appears in $\rho_{ab}$ at 30–50 K below $T_M$ in the pressure range from 1.5 to 5 GPa, where no indication of the corresponding change was observed in $\rho_c$. Below the temperature where the hump appears, $\rho_{ab}$ follows $\rho_{ab} = \rho_{ab0} + AT^n$ with $n = 1.3–1.5$ ($n = 2$ at ambient pressure). The hump in $\rho_{ab}$ is most pronounced at 3 GPa. After suppressing the hump by applying higher pressure, we can fit $\rho_{ab}$ at 8.0 GPa well to $\rho_{ab} = \rho_{ab0} + AT^n$ with $n \sim 1.3$ over the whole temperature. In contrast, $\rho_c$ at low temperatures

FIG. 1. The temperature dependence of the out-of-plane resistivity $\rho_c$ at several pressures. The inset shows the pressure dependence of $\rho_c$ at 295.5 K.

FIG. 2. The variation of $\rho_c$ with $T^2$ at several pressures. Broken lines show fitting results by using the relation $\rho = \rho_{c0} + AT^2$. The arrows show the characteristic temperature $T^*$, where $\rho_c$ deviates from the $T^2$ dependence.
exactly keeps the $T^2$ dependence even at 8.0 GPa. This pressure effect could not be detected in the previous results at low pressures.\(^{18}\)

The inset of Fig. 3 shows the pressure dependence of $\rho_{ab}$ at 295.5 K. We can see a large decrease of about $-30\%$ from the ambient pressure to 8.0 GPa. Neutron powder diffraction\(^{21}\) gave compressibilities of $2.24 \times 10^{-1}$ and $2.56 \times 10^{-1}/$GPa along the $a$ and $c$ axis at 300 K, respectively. The effect of lattice contraction on $\rho_{ab}$ is estimated as $-2\%$ at 8.0 GPa. This value is too small to explain the observed reduction of $-30\%$ in $\rho_{ab}$, suggesting a significant change of the electronic state of Sr$_2$RuO$_4$ due to pressure.

### IV. DISCUSSIONS

Let us examine the temperature and pressure dependence of $\rho_c$. In spite of $l_c < d$, $\rho_c$ retains the metallic behavior between $T^*$ and $T_M$. This implies the gradual crossover from the coherent metallic conduction to the incoherent conduction.\(^{14}\) More phenomenologically, Hussey et al.\(^{22}\) analyzed it by using a two-component model; namely, the combined effects of independent coherent and incoherent channels along the $c$ axis over all the temperature range. Similar situations have been discussed in the typical anisotropic conductors, such as organic conductors\(^{24\sim25}\) and high-$T_c$ cuprates.\(^{24\sim25}\)

Above $T^*$, we consider a diffusion (intermediate temperature region: $T^* \approx T \approx T_M$) and thermally hopping (high-temperature region: $T \approx T_M$) process for $\rho_c$, in which the in-plane quasiparticles hop between the adjacent planes with a transfer frequency $\tau_c^{-1}$. For this process, we expect

$$\sigma_c = e^2 N(E_F) d^2 \tau_c^{-1}.$$  

Here, $\sigma$ is the conductivity, $e$ is the elementary charge, $N(E_F)$ is the electronic density of states, and $d^2\tau_c^{-1} = D_c$ is the diffusion coefficient as $d$ is the jumping distance (= interlayer distance). It should be noted that this is continuously connected to the Drude expression at $l_c = d$.\(^{26}\)

Because of the quasicylindrical topology of the Fermi surfaces, their modulations from cylinders control the conduction perpendicular to the conductive planes through the transfer integral along the $c$ axis, $t_c(\Delta k)$. When $t_c$ is very small, a large number of in-plane scattering events can take place before a quasiparticle travels the interlayer distance. In this case, the transfer rate is given by\(^{23\sim25}\)

$$\tau_c^{-1} = \frac{1}{\hbar^2} \tau_{ab}^{-1},$$  

where $\hbar = h/2\pi$, and $\tau_{ab}^{-1}$ is the in-plane scattering rate. In addition, we assume the parallel resistivity and the common $T_M$ for the three Fermi-surface conductance. These assumptions describe the resistive behavior of Sr$_2$RuO$_4$ at the ambient pressure well.\(^{14}\) From Eqs. (1) and (2), we obtain the following equation for $T^* \approx T \approx T_M$:

$$\rho_c = \frac{1}{\sigma_c} = \frac{\hbar^2}{e^2 N(E_F) d^2} \tau_{ab}^{-1} \frac{\Sigma_{i}^2}{\Sigma_{i}^2} \approx \rho_{ab}(\Sigma_{i}^2)^{-1},$$  

where $i$ is labeled to each of the three FS sheets. This equation implies that $\rho_c$ is governed by the in-plane scattering rate and $t_c$. The former explains why the metallic temperature dependence is observed at the intermediate temperature region in spite of $l_c < d$. We expect that the crossover between such the diffusive metallic conduction and the Drude behavior takes place at $T^*$.

At high temperatures, the transfer rate is dominated by the thermal activations. When the thermal energy $k_B T$ is much larger than the effective band width $W_c = 4t_c$, which is based on the velocity distribution along the $c$ axis, we expect the conduction due to the thermally assisted hopping with the rate

$$\tau_c^{-1} = \tau_{0c}^{-1} \exp[-\alpha W_c(P)/k_B T]^n,$$  

where $\alpha$ is a numerical factor and $n$ depends on the dimensionality and the hopping range. Using $t_c = 17$ K for the $\beta$-FS in Ref. 2 which gives the largest $t_c$, $T_M$ at ambient pressure is comparable to $-8t_c$ or $2W_c$.

The scaling of $\rho_c$ against the normalized temperature of $T/T_M$ at each pressure shown in Fig. 4 serves to clarify the metal to nonmetal crossover. This nearly universal behavior implies the same scaling factor of the pressure dependence of $T_M$, $A$ of the $T^2$ term, and $\rho_c$ in the high-$T$ region, namely, the transfer integral $t_c$.\(^{27}\) From analyzing pressure dependence of $T_M$ and $A$ with an assumption that the variation of $t_c$ with pressure is independent of the three FS branches, we
estimate that \( t_c \) at 8.0 GPa is a factor of 1.6–1.9 larger than that at ambient pressure, as shown in Fig. 5.

Because both \( T_M \) and \( W_c \) are expected to be proportional to \( t_c \), the exponential term of Eq. (4) gives a same value at the crossover temperature \( T_M \) at each pressure, namely, a unique \( \tau_c^{-1} \) as a critical value at \( T_M \). With assumptions of weak pressure dependence of \( N(E_F) \) and lattice contraction of \(-2\%\) at 8.0 GPa,\(^{21}\) Eq. (1) exactly reproduces our observation that \( \rho_s(T_M) \) is insensitive to pressure. The unusual large increase of \( \rho_s \) at room temperature shown in the inset of Fig. 1 is understandable if one considers the behavior of \( \rho_s(T) \).

The above analyses let us confirm that the crossover from diffusive metallic transport to thermal hopping transport occurs at \( T_M \). This may be further supported by the analysis of optical spectra of \( \text{Sr}_2\text{RuO}_4 \) (Ref. 28) which shows that the Drude-like term appears below \( T_M \) in \( \sigma_s(\omega) \).

On the other hand, although \( \rho_c \) in the semiconducting regions is scaled by \( T/T_M \), we cannot fit it well by using any simple curve, such as Eqs. (1) and (4). Measurements of \( \rho_c \) at higher temperature above 300 K is needed to clear this problem.

Now we concentrate on the contrasting behavior of the in-plane and out-of-plane conductions under high pressures even in the coherent low-T region. Equation (3) requires the same temperature dependence of \( \rho_{ab} \) and \( \rho_c \) also for \( T^* \approx T \leq T_M \). Although \( \rho_{ab} \) becomes proportional to \( T^{-1.3} \) at 8.0 GPa after having a hump in the intermediate pressures, \( \rho_c \) still follows the \( T^2 \) law under the pressure and does not have any humplike structure. This suggests a peculiar twodimensional electronic state of \( \text{Sr}_2\text{RuO}_4 \).

Let us discuss this intriguing pressure dependence of \( \rho_{ab} \) from the viewpoint of spin fluctuations. \( \text{Sr}_2\text{RuO}_4 \) is conjectured to be near a ferromagnetic instability because the homologous \( \text{SrRuO}_3 \) is an itinerant ferromagnet (\( T_C \approx 160 \text{ K} \)). Such a conjecture is one of the grounds for the \( p \)-wave superconductivity in \( \text{Sr}_2\text{RuO}_4 \) because the parallel-spin correlations disfavor both \( s \) and \( d \) superconductivity. Experimentally, there are many observations\(^{6–8}\) to support the Cooper pairing with an exotic spin-triplet symmetry, but no decisive one to determine the pairing mechanism. It is obviously important for the pairing mechanism whether the ferromagnetic correlations exist or not. For example, Mazin et al.\(^ {12} \) suggest that the electron-paramagnon coupling is responsible for the superconductivity of \( \text{Sr}_2\text{RuO}_4 \). Therefore, it is interesting to investigate how ferromagnetic spin fluctuations work in the low-\( T \) normal-state properties. In fact, the NMR results\(^ {5} \) seem to show that \( \text{Sr}_2\text{RuO}_4 \) is a Pauli paramagnet with an exchange enhancement by ferromagnetic spin correlation. In addition, very recently, Ikeda et al.\(^ {29} \) have shown that the homologous \( \text{Sr}_2\text{CaRu}_2\text{O}_7 \) with the ferromagnetic Curie temperature at 3 K is well described by the self-consistent renormalization (SCR) theory with the 3D ferromagnetic spin fluctuations.

The SCR theory with ferromagnetic spin fluctuations\(^ {30} \) predicts that, in the vicinity of magnetic instability, the temperature dependence of resistivity at low temperatures changes from \( T^2 \) for the paramagnetic ground state to \( T^4 \) for the ferromagnetic one. Here, \( n \) depends on the magnetic dimensionality: \( n = 4/3 \) (2D), 5/3 (3D). Moreover, when the ground state is ferromagnetic, the humplike crossover is visible as the spin-fluctuation term governs resistivity at low temperatures. Figure 6 reveals the variation of \( \rho_{ab} \) divided \( \rho_0 \) with \( T^{4/3} \) at several pressures, where \( \rho_0 \) is the fitting coefficient of \( \rho = \rho_0 + B(P)T^{4/3} \). This figure seems to support, at least in a qualitative sense, that pressure enhances 2D ferromagnetic spin correlations, leading to ferromagnetic ground state of \( \text{Sr}_2\text{RuO}_4 \) under high pressure. If this is the case, \( \text{Sr}_2\text{RuO}_4 \) will be the first example to study the 2D itinerant ferromagnetic system, helping us to understand the qualitative difference between \( \rho_{ab} \) and \( \rho_c \) under pressure even in the coherent region. Moreover, the decrease of \( T_c \) with applying pressure\(^ {18} \) might imply the competition between superconductivity and enhanced ferromagnetic correlations.

A crucial question arising from the above discussion is whether the two dimensionality of the spin correlations is actually enhanced by pressure in the vicinity of the quantum boundary region, leading to the magnetic ground state. Judging from the lowest-temperature behavior of \( \rho_c \) in Fig. 1 and \( \rho_{ab} \) in Fig. 3, applied pressure enhances the three-dimensional character of the electronic state of \( \text{Sr}_2\text{RuO}_4 \). Moreover, from the dc magnetic susceptibility,\(^ {13} \) the spin state at ambient pressure is rather isotropic. Therefore, the alternative scenario for the appearance of the hump may be based on the 3D ferromagnetic fluctuation in the quasi-two-dimensional electronic state. For this scenario, it is a key point to explain why the hump is observed only in \( \rho_{ab} \).
In any case, we presume the magnetic origin for the appearance of the hump. For determination of the mechanism, it is very important to confirm how the SCR theory quantitatively explains, the other observed physical properties of Sr$_2$RuO$_4$, and to perform the further measurements under pressure, such as magnetic susceptibility, resistivity in magnetic fields, and NMR to observe the spin susceptibility directly.


Using the Fermi surface parameters in Ref. 2, the mean-free path along the c axis $l_c$ becomes equal to the interlayer distance $d$ for $\rho_{c} \sim 4 \\text{m}\Omega \text{cm}$.


Within a Drude description, conductivity is given by $\sigma = e^2 N(E_F) v^2 \tau$ as $v = l/\tau$.

According to Ref. 2, $A$ is proportional to $l_c^{-2}$.

