Electronic crossover in the highly anisotropic normal state of Sr_2RuO_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_2RuO_4. The characteristic temperature T_M, where the out-of-plane resistivity ρ_c changes from metallic behavior to nonmetallic behavior with increasing temperature, increases with hydrostatic pressure. In contrast, the absolute value of ρ_c at T_M is almost independent of pressure. Such a change over in ρ_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 ρ_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_2RuO_4,[1] 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 noncopper superconductor with a layered perovskite structure similar to high-T_c cuprates. Reflecting its nearly cylindrical Fermi surfaces (FS),[2–4] the superconducting state of Sr_2RuO_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[7] measurements showed that in Sr_2RuO_4 a finite density of states remains even at $T \rightarrow 0$ K. Mackenzie *et al.*[8] reported a strong suppression of T_c by nonmagnetic impurities. These results suggest unconventional superconductivity in this ruthenate. Some theorists[9–12] have discussed the possibility of an exotic *p*-wave paring, including a nonunitary state.

The normal-state properties of Sr_2RuO_4 are as fascinating as its superconductivity. A highly anisotropic Fermi-liquid model has been successfully adopted to describe the normal state.[2,13] The quasi-two-dimensional electronic state is characterized by the peculiar anisotropic resistivity.[1,14] The resistivity along the *c* axis ρ_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 ρ_{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_2RuO_4, ρ_c of the lightly doped high-T_c cuprates exhibits a steep semiconductorlike upturn at low temperatures,[15,16] whereas ρ_{ab} remains metallic. From the high-pressure measurements on optimally doped $La_{2-x}Sr_xCuO_4$,[17] even the apparent metallic temperature dependence of ρ_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 ρ_c is closely related with the appearance of high T_c.

In order to understand the anisotropic electronic states of Sr_2RuO_4, we have measured the normal-state resistivities ρ_{ab} and ρ_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.*[18] reported anisotropy in the pressure dependence of resistivity at room temperature up to 1.2 GPa, namely, the increase (+2%/GPa) of ρ_c and the decrease (−6%/GPa) of ρ_{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_2RuO_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 RuO_2. 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

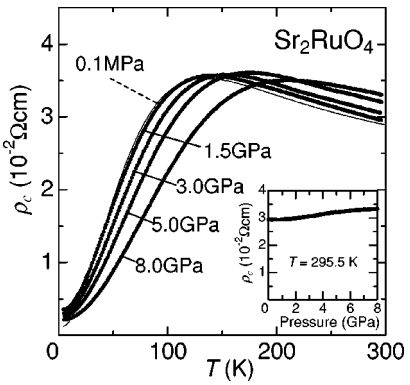

FIG. 1. The temperature dependence of the out-of-plane resistivity ρ_c at several pressures. The inset shows the pressure dependence of ρ_c at 295.5 K.

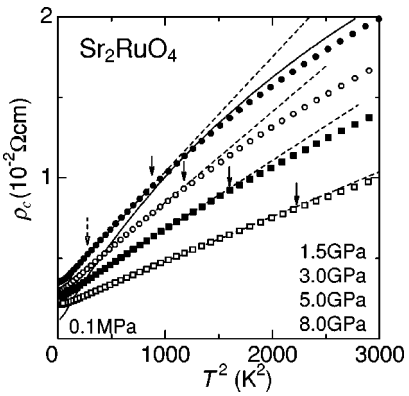

FIG. 2. The variation of ρ_c with T^2 at several pressures. Broken lines show fitting results by using the relation $\rho=\rho_0+AT^2$. The arrows show the characteristic temperature T^*, where ρ_c deviates from the T^2 dependence.

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_c of $\sim$0.7 K.

The resistivity was measured using a standard four probe method, except for a ring contact geometry for ρ_c.[1] The typical size of samples was $0.2\times0.6\times0.01$ mm^3 for ρ_{ab} and $0.3\times0.3\times0.01$ mm^3 for ρ_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 °C for 5 min. We attained a contact resistance of 0.5Ω.

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, quasihydrostatic pressure. First, we measured ρ_{ab} and ρ_c between 4.2 and 300 K at ambient pressure. Next, the crystals were mounted in the high-pressure cell. The resistivity at each pressure was measured from 4.2 to 300 K, after the applied pressure was set at room temperature. The pressure applied to the anvil unit was actively controlled to be constant within 3% during the temperature excursion.

III. RESULTS

A. Pressure dependence of out-of-plane resistivity ρ_c

Figure 1 shows the temperature dependence of ρ_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 ρ_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 ρ_{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 ρ_{c0} and A were not resolved, we found that both parameters systematically decrease at higher pressures. The characteristic temperature T^*, where ρ_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^*, ρ_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 ρ_c is $\approx$32 mΩ 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_2RuO_4, $\Delta k_F(\beta-\mathrm{FS})=0.008$ Å^{-1} at ambient pressure.[2] Here, the β-Fermi surface has the largest Δk_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 ρ_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 ρ_{ab}

Pressure effects on ρ_{ab} are qualitatively different from those on ρ_c, as shown in Fig. 3. The in-plane ρ_{ab} shows metallic behavior at any pressure over the whole temperature range. No anomaly is observed in ρ_{ab} at T_M. An interesting finding is that a hump appears in ρ_{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 ρ_c. Below the temperature where the hump appears, ρ_{ab} follows $\rho_{ab}=\rho_{ab0}+AT^n$ with $n=1.3$–1.5 ($n=2$ at ambient pressure). The hump in ρ_{ab} is most pronounced at 3 GPa. After suppressing the hump by applying higher pressure, we can fit ρ_{ab} at 8.0 GPa well to $\rho_{ab}=\rho_{ab0}+AT^n$ with $n\sim1.3$ over the whole temperature. In contrast, ρ_c at low temperatures

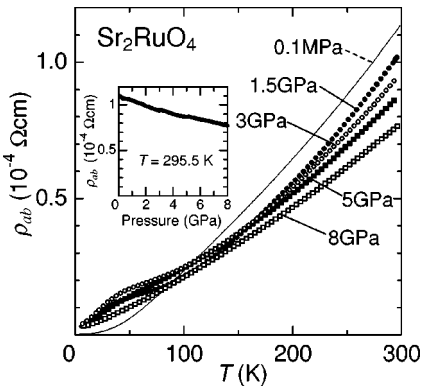

FIG. 3. The temperature dependence of the in-plane resistivity ρ_{ab} at several pressures. The inset shows the pressure dependence of ρ_{ab} at 295.5 K.

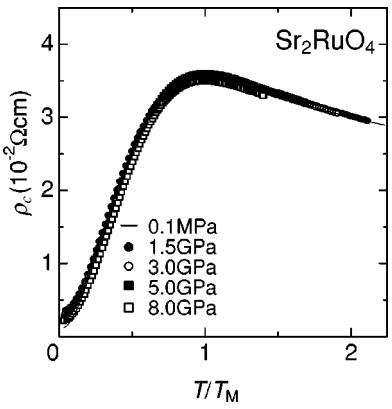

FIG. 4. The variation of ρ_c with the normalized temperature T/T_M at several pressures.

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 ρ_{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 2.24×10^{-1} and 2.56×10^{-1}%/GPa along the *a* and *c* axis at 300 K, respectively. The effect of lattice contraction on ρ_{ab} is estimated as -2% at 8.0 GPa. This value is too small to explain the observed reduction of -30% in ρ_{ab}, suggesting a significant change of the electronic state of Sr_2RuO_4 due to pressure.

IV. DISCUSSIONS

Let us examine the temperature and pressure dependence of ρ_c. In spite of $l_c<d$, ρ_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[23] and high-T_c cuprates.[24,25]

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

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

Here, σ 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, $t_c(\propto k_F \Delta k_F)$. 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[23–25]

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

where $\hbar = h/2\pi$, and τ_{ab}^{-1} is the in-plane scattering rate. In addition, we assume the parallel resistivity and the common τ_{ab} for the three Fermi-surface conductance. These assumptions describe the resistive behavior of Sr_2RuO_4 at the ambient pressure well.[14] From Eqs. (1) and (2), we obtain the following equation for $T^*\leqslant T\leqslant T_M$:

$$\rho_c = \frac{1}{\sigma_c} = \frac{\hbar^2}{e^2 N(E_F) d^2} \tau_{ab}^{-1} (\Sigma t_{c,i}^2)^{-1} \propto \rho_{ab} (\Sigma t_{c,i}^2)^{-1}, \tag{3}$$

where *i* is labeled to each of the three FS sheets. This equation implies that ρ_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_BT 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

$$\tau_c^{-1} = \tau_{c0}^{-1} \exp[-\alpha W_c(P)/k_B T]^n, \tag{4}$$

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

The scaling of ρ_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 ρ_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

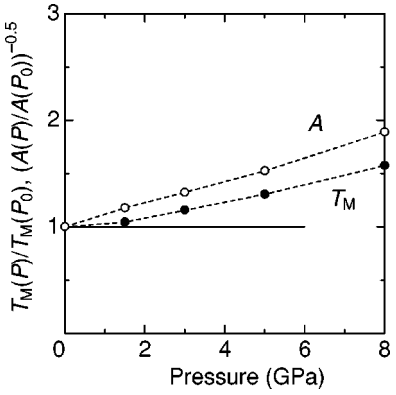

FIG. 5. The pressure dependences of the normalized T_M and A by using the values at ambient pressure, 134 K and 10 $\mu\Omega$ cm/K^2, respectively. Here, both T_M and $\sqrt{A^{-1}}$ are expected to be proportional to t_c.

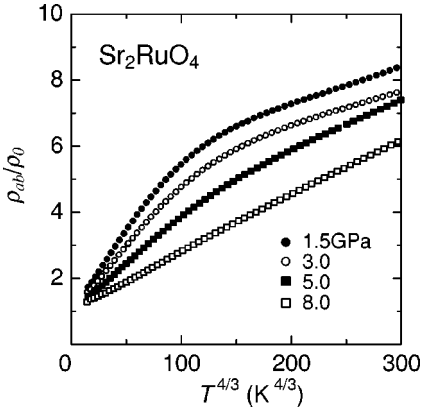

FIG. 6. The variation of ρ_{ab} divided ρ_0 with $T^{4/3}$ at several pressures, where ρ_0 is the fitting coefficient of $\rho=\rho_0+B(P)T^{4/3}$.

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 τ_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 $\rho_c(T_M)$ is insensitive to pressure. The unusual large increase of ρ_c at room temperature shown in the inset of Fig. 1 is understandable if one considers the behavior of $\rho_c(T)$.

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 Sr_2RuO_4 (Ref. 28) which shows that the Drude-like term appears below T_M in $\sigma_c(\omega)$.

On the other hand, although ρ_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 ρ_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 ρ_{ab} and ρ_c also for $T^* \leqslant T \leqslant T_M$. Although ρ_{ab} becomes proportional to $T^{\sim 1.3}$ at 8.0 GPa after having a hump in the intermediate pressures, ρ_c still follows the T^2 law under the pressure and does not have any humplike structure. This suggests a peculiar two-dimensional electronic state of Sr_2RuO_4.

Let us discuss this intriguing pressure dependence of ρ_{ab} from the view point of spin fluctuations. Sr_2RuO_4 is conjectured to be near a ferromagnetic instability because the homologous $SrRuO_3$ is an itinerant ferromagnet ($T_C \sim 160$ K). Such a conjecture is one of the grounds for the p-wave superconductivity in Sr_2RuO_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 Sr_2RuO_4. Therefore, it is interesting to investigate how ferromagnetic spin fluctuations work in the low-T normal-state properties. In fact, the NMR results[6] seem to show that Sr_2RuO_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 $Sr_2CaRu_2O_7$ 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^n 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 ρ_{ab} divided ρ_0 with $T^{4/3}$ at several pressures, where ρ_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 Sr_2RuO_4 under high pressure. If this is the case, Sr_2RuO_4 will be the first example to study the 2D itinerant ferromagnetic system, helping us to understand the qualitative difference between ρ_{ab} and ρ_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 ρ_c in Fig. 1 and ρ_{ab} in Fig. 3, applied pressure enhances the three-dimensional character of the electronic state of Sr_2RuO_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 ρ_{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_2RuO_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.

Moreover, in our discussions we assumed that the pressure effect operates equally for the three degenerated FSs:[3,4] one is a nearly circular cylindrical Fermi surface dominated by Ru $4d_{xy}$ orbital character, and the other two are rather tetragonal prismatic FS dominated by Ru $4d_{yz}$ and $4d_{zx}$ orbital character, which originate from reconnecting four crossing planes (quasi-1D FS) by the weak *yz-zx* hybridization. It is very interesting to clarify how the pressure effects work for an individual FS.

V. CONCLUSION

We have observed several fascinating characteristics of the in-plane and out-of-plane conductivity of Sr_2RuO_4 induced by applied pressure. We confirm that the change over from the metallic to nonmetallic conduction of ρ_c originates from the mechanism of the crossover from the diffusion process of quasiparticles between the adjacent layers to the thermally assisted hopping. We have explained why this crossover mechanism gives a pressure-insensitive critical value of ρ_c at the crossover temperature of T_M, which plays a key role for the unconventional increase of ρ_c at room temperature with pressure. The systematic change with increasing pressure, such as in T_M, is probably understood in terms of the enhanced transfer integral along the *c* axis by pressure.

As for ρ_{ab}, we found an intriguing hump at 30–50 K below T_M in the pressure range from 1.5 to 5 GPa, where we do not observe any corresponding changes of ρ_c. This may give evidence that Sr_2RuO_4 is in the vicinity of the magnetic instability with the two-dimensional ferromagnetic spin fluctuations. Therefore, Sr_2RuO_4 may provide the first opportunity for the extensive studies of the itinerant 2D ferromagnet, which could play a key role for its *p*-wave superconductivity.

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