Non-Fermi-Liquid Behavior in Sr_2RuO_4 with Nonmagnetic Impurities

Naoki Kikugawa[1,2] and Yoshiteru Maeno[1,3]

[1]*Department of Physics, Kyoto University, Kyoto 606-8502, Japan*
[2]*Venture Business Laboratory, Kyoto University, Kyoto 606-8501, Japan*
[3]*International Innovation Center, Kyoto University, Kyoto 606-8501, Japan*

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We report that the quasi-two-dimensional Fermi-liquid behavior of the spin-triplet superconductor Sr_2RuO_4 breaks down in the vicinity of the critical impurity concentration for the onset of magnetic order induced by *nonmagnetic* Ti^{4+} impurities. The non-Fermi-liquid behavior is interpreted in terms of the two-dimensional antiferromagnetic fluctuations, which arise mainly from the nesting within one of the Fermi-surface sheets. We argue against the main role of such magnetic fluctuations in the pairing mechanism.

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It has been widely recognized that physical properties in highly correlated electron systems such as f-electron systems and high-T_c cuprates show pronounced deviation from the conventional Fermi-liquid behavior [1]. The non-Fermi-liquid behavior is often observed in the vicinity of a quantum critical point at which magnetic ordering temperature is driven to zero by varying parameters such as pressure and chemical substitution, or even in the vicinity of a metamagnetic transition point by varying a magnetic field [2]. In addition, unconventional superconductivity often emerges near such a magnetic instability point [3]. Therefore, it has become increasingly important to study a variety of highly correlated electron systems, especially the ones for which the Fermi-liquid behavior as well as the origin of its breakdown can be well understood, in order to establish generic implication of such non-Fermi-liquid behavior.

For this purpose, layered perovskite Sr_2RuO_4 may be regarded as an ideal system, because this system will provide a detailed set of data to deepen our knowledge of non-Fermi-liquid behavior. First, its superconductivity [4] with the transition temperature $T_c = 1.5$ K is of unconventional pairing symmetry, most probably spin triplet [5]. Second, the details of the entire Fermi surfaces have been quantitatively characterized by the quantum oscillation measurements [6]. The Fermi surface is composed of three nearly cylindrical sheets: α and β bands reflecting quasi-one-dimensional character associated with d_{yz} and d_{zx} orbits, and an γ band with quasi-two-dimensional character associated with the d_{xy} orbit. On the basis of the Fermi-surface topography, the strongly anisotropic normal state properties are described quantitatively by the framework of a quasi-two-dimensional Fermi liquid [7]. In that study, it was revealed that the strong correlations among the electrons play an essential role in the physical properties of Sr_2RuO_4.

At first, theoretical insight suggested that ferromagnetic spin fluctuations may be the main mechanism of the spin-triplet pairing in Sr_2RuO_4, on the basis of the similarity to the Fermi-liquid properties of superfluid ^{3}He and to the ferromagnetic relative compound $SrRuO_3$ [8]. Inelastic neutron scattering measurement, however, revealed a strong incommensurate (antiferromagnetic) fluctuation with a large Stoner factor close to 1, but no discernible response around $\boldsymbol{q} = 0$ [9]. The incommensurate wave vector $\boldsymbol{Q}_{ic} \sim (2\pi/3, 2\pi/3, 0)$ is in accord with the nesting vector within the β Fermi surface, predicted by the band calculation [10]. Thus, the pairing mechanism in Sr_2RuO_4 seems more involved than initially expected because of the multiband effects. On the basis of the magnetic properties in the normal state, a number of scenarios for the mechanism and gap structure of the spin-triplet superconductivity in Sr_2RuO_4 have been proposed [11–13]; the validity of these models is now under active debate.

Very recently, we reported that the substitution of *nonmagnetic* impurity Ti^{4+} (the electron configuration $3d^0$) for Ru^{4+} ($4d^4$ in the low spin configuration) in Sr_2RuO_4 induces local moment which has Ising anisotropy with an easy axis along the c axis [14]. The induced moment is attributable to Ru^{4+} spins surrounding Ti, because interruption of the d-p hybridization paths by Ti^{4+} tends to localize the $4d$ electrons in the quasi-one-dimensional orbits. Furthermore, magnetic ordering with glassy behavior appears for $x \geq 2.5\%$ in $Sr_2Ru_{1-x}Ti_xO_4$. Thus, $Sr_2Ru_{1-x}Ti_xO_4$ shows the rich ground states varying from the spin-triplet superconductivity with the quasi-two-dimensional Fermi-liquid state to the magnetic ordering with glassy behavior. However, the relation between the superconductivity and the magnetism still remains unclear for $Sr_2Ru_{1-x}Ti_xO_4$.

In this Letter, we report the emergence of non-Fermi-liquid behavior in the vicinity of the critical concentration $x_c \sim 2.5\%$ for the magnetic ordering in $Sr_2Ru_{1-x}Ti_xO_4$. Our results for resistivity and specific heat strongly suggest that the non-Fermi-liquid behavior is driven by the

two-dimensional antiferromagnetic fluctuations. We also present the role of the antiferromagnetic fluctuations in the pairing mechanism of Sr_2RuO_4.

A series of single-crystalline $Sr_2Ru_{1-x}Ti_xO_4$ with x up to 9% were grown by a floating-zone method with an infrared image furnace (NEC Machinery, model SC-E15HD) [14,15]. The analyzed Ti concentrations by electron-probe microanalysis (EPMA) are in good agreement with the nominal concentrations within the detection uncertainty. The in-plane resistivity ρ_{ab} measurements were performed by a standard four-probe dc method between 4.2 and 300 K and by a low frequency ac method between 0.3 and 5 K. The specific heat C_P was measured by a thermal relaxation method from 0.5 to 30 K (Quantum Design, model PPMS).

Figure 1 shows the temperature dependence of ρ_{ab} with a small amount of x up to 0.50%. Two features in the figure should be noted. First, T_c is rapidly suppressed with the initial rate $dT_c/dx \approx -8$ K/Ti%, reflecting the high sensitivity to translational symmetry breaking, characteristic of unconventional superconductivity [16]. No superconducting transition is detected beyond $x \approx 0.15\%$, much below the critical concentration for the occurrence of the magnetic ordering at $x_c \sim 2.5\%$. Second, a rapid enhancement of the residual resistivity ρ_{ab0} with x, defined by the extrapolation of the low-temperature resistivity to $T = 0$ K, is observed. The upturn of ρ_{ab} below 1 K observed for $x = 0.25\%$ crystals suggests a connection with fluctuations near the critical disorder for disappearance of superconductivity, although theoretical examination predicts specific temperature dependence leading to reduction in ρ_{ab} [17]. The T_c as a function of ρ_{ab0} is shown in Fig. 2. The open symbols are taken from previous studies [16], in which the T_c is suppressed by the crystal defects and the aluminum contamination. The present results indicate a universal trend quantitatively consistent with the previous ones: The superconductivity of Sr_2RuO_4 is completely suppressed at the critical resistivity of $\rho_{ab0} \sim 1.1\ \mu\Omega$ cm. The critical value is in good agreement with the mean-free path l_{ab} falling below the superconducting coherence length $\xi_{ab} \sim 900$ Å [16].

Figure 3 shows the temperature dependence of the inelastic part of the in-plane resistivity $\Delta\rho_{ab} \equiv \rho_{ab}(T) - \rho_{ab0}$ with x up to 3% plotted as $\log(\Delta\rho_{ab})$ vs $\log T$. The T-squared dependence $\Delta\rho_{ab} = AT^n$ with $n = 2$ satisfied below about 30 K for $x = 0$ starts to break down with Ti substitution. The exponent sharply deviates from 2 and becomes nearly unity at $x_c = 2.5\%$ between 0.3 and 2 K, in almost one decade of temperature range. This result strongly suggests a breakdown of the simple Fermi-liquid behavior near the critical concentration x_c. The origin and analysis of the low-temperature behavior for $x = 3\%$ will be described below. In the inset of Fig. 3, the x dependence of ρ_{ab0} is displayed. The broken line represents $d\rho_{ab0}/dx = 4.25\ \mu\Omega$ cm/Ti% expected for the unitarity scattering [18]. This expectation is in good agreement with the experimental value of $d\rho_{ab0}/dx \sim 5\ \mu\Omega$ cm/Ti%. Thus, nonmagnetic impurity acts as a strong potential scatterer in Sr_2RuO_4, similar to the substitution effect of nonmagnetic (Zn^{2+}) impurity in high-T_c cuprates [19].

Figure 4 shows the specific heat divided by temperature C_P/T for $x = 0$, 2.5, and 9% plotted as functions of T^2. The data for $x = 0$ was obtained by applying the magnetic field of 0.2 T along the c axis in order to suppress the superconductivity ($T_c = 1.44$ K). There is no evidence of magnetic field dependence of the C_P/T at $T \geq T_c$ within the experimental resolution, consistent with the previous report [20]. We can clearly see the enhancement of the C_P/T at $x_c = 2.5\%$. The enhancement cannot be simply explained by the impurity band induced by the

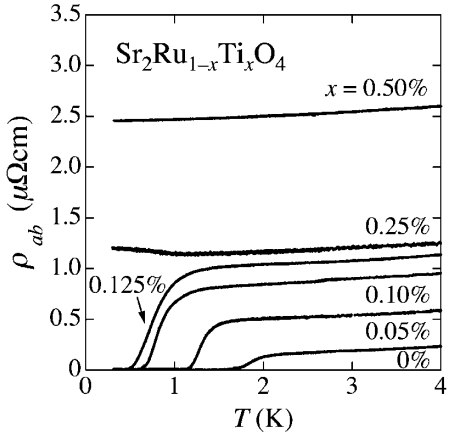

FIG. 1. Temperature dependence of the in-plane resistivities in $Sr_2Ru_{1-x}Ti_xO_4$ with x up to 0.50%.

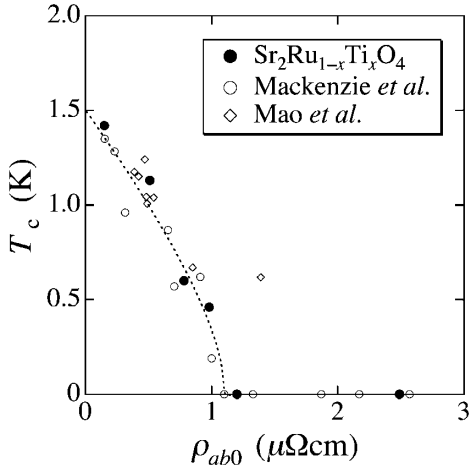

FIG. 2. Superconducting transition temperature T_c as a function of the in-plane residual resistivity ρ_{ab0} for $Sr_2Ru_{1-x}Ti_xO_4$. Previous results for different means of disorder (Ref. [16]) are added. The broken line shows the Abrikosov-Gor'kov pair-breaking function.

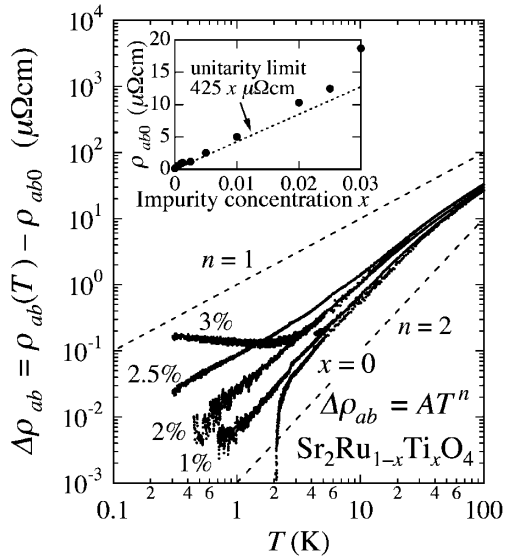

FIG. 3. Temperature dependence of the inelastic part of the in-plane resistivity $\Delta\rho_{ab} \equiv \rho_{ab}(T) - \rho_{ab0}$ in $Sr_2Ru_{1-x}Ti_xO_4$ plotted as $\log(\Delta\rho_{ab})$ vs $\log T$. The broken lines represent $\Delta\rho_{ab} = AT^n$ with $n = 1$ and 2. Inset: Residual resistivity as a function of nonmagnetic Ti concentration. The broken line represents the unitarity limit [18].

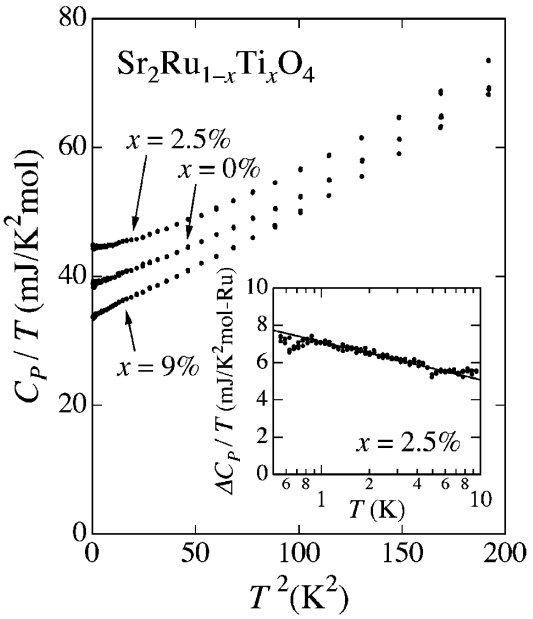

FIG. 4. Specific heat divided by temperature C_P/T plotted against T^2. The inset shows temperature dependence of $\Delta C_P/T = C_P/T\ (x = 2.5\%) - C_P/T\ (x = 0)$ plotted as $\Delta C_P/T$ vs $\log T$. The line is a guide for the eye.

substitution effect, because such an effect is not observed for $x = 9\%$. In addition, for $x = 2.5\%$, a logarithmic upturn behavior, which deviates from the simple Fermi-liquid behavior in pure Sr_2RuO_4, is observed as demonstrated in the inset of Fig. 4, where $\Delta C_P/T = C_P/T\ (x = 2.5\%) - C_P/T\ (x = 0)$. This logarithmic behavior is similar to that seen in the heavy fermion systems in the vicinity of the magnetic instability [21].

The T-x phase diagram of $Sr_2Ru_{1-x}Ti_xO_4$ is presented in Fig. 5(a). Further substitution of Ti beyond the suppression of the superconductivity at $x \sim 0.15\%$ leads to the magnetic ordering beyond x_c. Figures 5(b) and 5(c) show the value of C_P/T at 0.5 K and the power-law exponent n of the resistivity. Estimation of n for $x \geq 3\%$ is not straightforward, since the localization behavior with a logarithmic term is observed [14]. We obtained n by fitting $\rho_{ab} = \rho_{ab0} + AT^n - B\ln(T/T_0)$, where T_0 corresponds to the characteristic temperature of the localization behavior, shown in Fig. 5(a). We note that the choice of T_0 barely affects the determination of n. We can clearly see the anomaly in both n and C_P/T centered around x_c.

Next, we discuss the origin of the non-Fermi-liquid behavior in the vicinity of x_c. In $Sr_2Ru_{1-x}Ti_xO_4$ with $x = 9\%$, incommensurate magnetic ordering with the wave vector $\boldsymbol{Q}_{ic}$, which is close to the position of the *inelastic* neutron scattering peak in pure Sr_2RuO_4 [9], is also observed below about 25 K by the *elastic* neutron scattering measurement, where the Stoner factor reaches 1 at x_c [22]. This result indicates that the two-dimensional antiferromagnetic spin fluctuations at $\boldsymbol{Q}_{ic}$ become static by *nonmagnetic* Ti substitution. Moreover, the resistivity and specific heat at x_c show $\Delta\rho_{ab} \propto T^n$ with $n = 1$ and $\Delta C_P/T \propto -\ln T$, respectively. These results are in good agreement with the expectation of the self-consistent-renormalization theory [23] for two-dimensional antiferromagnetic spin fluctuations.

Finally, we discuss the implication of the current result for the mechanism of the spin-triplet superconductivity in Sr_2RuO_4. Recently, it has been proposed that the nesting property of α and β Fermi surfaces leads to the spin-triplet pairing in Sr_2RuO_4, as long as the magnetic susceptibility is strongly anisotropic [11]. Recent ^{87}Sr NMR measurement for a series of $Sr_2Ru_{1-x}Ti_xO_4$ revealed that anisotropic antiferromagnetic fluctuations are enhanced continuously toward x_c [24]. In the case that the antiferromagnetic fluctuations play a constructive role in the superconductivity, the rate of the suppression in T_c by Ti substitution, which enhances the antiferromagnetic fluctuations, would be slower [25] than that observed for crystals with other kinds of impurities [16], with which the enhanced antiferromagnetic fluctuations are absent. However, the T_c in $Sr_2Ru_{1-x}Ti_xO_4$ is as rapidly suppressed by the strong scattering as with other native defects shown in Fig. 2. Thus, our result does not appear to be consistent with the theories based on the antiferromagnetic fluctuation mechanism of superconductivity. Nevertheless, it is not easy to give reliable estimation of T_c based on a simple RPA approximation. In order to examine the mechanism, it is thus important to clarify the roles of spin fluctuations [26] and correlations [12] of the γ band as well.

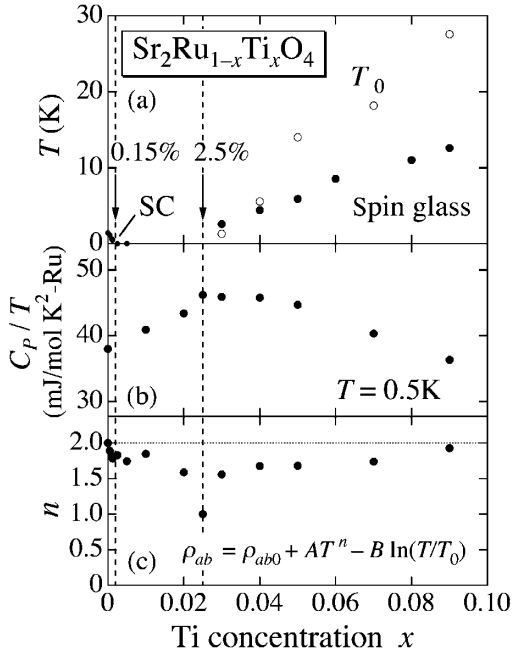

FIG. 5. (a) Phase diagram of $Sr_2Ru_{1-x}Ti_xO_4$, (b) the value of C_P/T at 0.5 K, and (c) the power-law exponent n of the resistivity as a function of Ti concentration x.

In summary, we reported the non-Fermi-liquid behavior emerging near the spin-triplet superconductivity. The substitution of the nonmagnetic impurity changes the ground state of $Sr_2Ru_{1-x}Ti_xO_4$ from the spin-triplet superconductivity ($0 \leq x \leq 0.15\%$) to the magnetic ordering ($x \geq 2.5\%$) with glassy behavior through band-selective modification of magnetic fluctuations. The non-Fermi-liquid behavior, attributable to the enhanced two-dimensional antiferromagnetic fluctuations, are observed in the vicinity of the magnetic instability. The fluctuations, however, do not seem to play a constructive role in the occurrence of the spin-triplet superconductivity in Sr_2RuO_4.

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[18] ρ_{ab0} in a two-dimensional system is given as $\rho_{ab0} = \frac{4\pi}{e^2}(x/\sum_i^{\alpha,\beta,\gamma} n_i)\sin^2\delta_0$. Here $\hbar$ is the Dirac constant, n_i the carrier concentration for each Fermi-surface (α, β, γ), and δ_0 the phase shift. We obtain $\rho_{ab0} = 425x\ \mu\Omega$ cm in $Sr_2Ru_{1-x}Ti_xO_4$ if we assume the unitarity limit, namely $\delta_0 = \pi/2$.
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