Double metamagnetic transition in the bilayer ruthenate $Sr_3Ru_2O_7$

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We report a double metamagnetic transition in $Sr_3Ru_2O_7$ detected by magnetization and magnetic torque measurements. In addition to the reported metamagnetic transition [R. S. Perry, L. M. Galvin, S. A. Grigera, L. Capogna, A. J. Schofield, A. P. Mackenzie, M. Chiao, S. R. Julian, S. I. Ikeda, S. Nakatsuji, Y. Maeno, and C. Pfleiderer, Phys. Rev. Lett. **86**, 2661 (2001)], an unexpected second transition started to appear in the higher-field region below 2 K, exhibiting stronger anisotropy between inter- and intraplane field directions. We shall discuss metamagnetism of $Sr_3Ru_2O_7$ based upon spin-fluctuation theory as well as possible origins of the second metamagnetic transition.

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Recently, Ruddlesden-Popper (RP) -type ruthenates $Sr_{n+1}Ru_nO_{3n+1}$ have attracted considerable attention due to a rich variety of ground states. The $n=1$ compound Sr_2RuO_4,[1] which is of a layered structure similar to the high-T_c superconductor $La_{2-x}Sr_xCuO_4$, is an unconventional superconductor with spin triplet symmetry, confirmed by the nuclear-magnetic resonance measurement.[2] Although the detailed symmetry of the order parameter is still under debate, the superconductivity is believed to be mediated by magnetic excitations. On the other hand, the $n=\infty$ compound $SrRuO_3$,[3,4] with a pseudocubic structure, is known to exhibit a ferromagnetic transition at 160 K with a magnetic moment of 1 μ_B/Ru. This compound provides a rare example of itinerant $4d$ ferromagnets.

The $n=2$ compound $Sr_3Ru_2O_7$ (Refs. 5 and 6) is of significant importance because of its intermediate dimensionality between paramagnetic Sr_2RuO_4 and ferromagnetic $SrRuO_3$, and thus is expected to play a role in connecting both compounds. In fact, the bilayer $Sr_3Ru_2O_7$ is an exchange-enhanced paramagnet characterized by a large Wilson ratio greater than 10. In addition, a pressure-induced ferromagnetic state was found below 70 K.[6] These findings clearly demonstrate that $Sr_3Ru_2O_7$ is on the verge of ferromagnetic instability.

It should be also noted that a metamagnetic transition was found in a moderate magnetic field of 5–7 T at low temperatures.[7] Contrary to the well-known $3d$ or $5f$ itinerant metamagnets,[8] metamagnetic transition in $4d$ itinerant systems is hardly reported. Since $4d$ electrons in ruthenates are characterized by both strong itinerancy and orbital degrees of freedom in a t_{2g} manifold, $Sr_3Ru_2O_7$ provides a unique system in comparison with other systems. It is of particular interest to see how these properties are relevant to the system's metamagnetism. Furthermore, it is reported that both the T^2 coefficient of the resistivity and the specific heat tend to diverge toward the metamagnetic transition field, suggestive of a magnetic instability point. Grigera *et al.*[9] and Millis *et al.*[10] discussed the results from the viewpoint of quantum criticality tuned by magnetic field.

These facts motivated us to investigate the metamagnetism of $Sr_3Ru_2O_7$ in more detail. Here we present magnetization data of $Sr_3Ru_2O_7$ as well as magnetic torque data in magnetic fields of up to 40 T with the use of pulse magnets at low temperatures down to 0.5 K. In contrast to resistivity data dominated by scattering events, those measurements give more direct information on magnetic properties. The main result of this paper is the observation of a second metamagnetic transition in slightly higher field than the first one observed before. In this paper, we shall discuss the metamagnetism of $Sr_3Ru_2O_7$ based upon spin-fluctuation theory, and address possible origins of the double metamagnetic transition.

High quality single crystals of $Sr_3Ru_2O_7$ were grown by a floating-zone method[11] in Tsukuba. Powder x-ray-diffraction measurements revealed that those single crystals do not contain any impurity phases such as ferromagnetic $SrRuO_3$ ($T_c=160$ K) or superconducting Sr_2RuO_4 ($T_c=1.5$ K). This is also supported by magnetization measurements with use of a commercial superconducting quantum interference device magnetometer. Those samples exhibited the residual in-plane resistivity of $\rho_{res}\simeq 0.8$–2.0 $\mu\Omega$ cm, which is lower than that reported by Perry *et al.*[7] ($\rho_{res}\simeq 2-15$ $\mu\Omega$ cm). In this paper, we present experimental results of the samples taken from different two batches. The difference between them is only the residual resistivity value, and we found that all the samples exhibited the same behavior.

Measurements were carried out with the use of pulse magnets at the Institute for Solid State Physics, University of Tokyo. Pulsed magnetic fields of up to 40 T were generated by capacitor discharge. Temperature was cooled to 0.5 K by directly pumping liquid ^{3}He. The samples were immersed in liquid to ensure thermal contact.

Magnetization measurements were performed by a conventional induction method where a sample is put in pickup coils and the induced voltage proportional to dM/dt is recorded during rapid field sweeps. In addition, we measured the magnetic torque of $Sr_3Ru_2O_7$ up to 40 T with the use of a recently developed microcantilever.[12] With this device, it becomes possible to measure the magnetic torque of samples of less than 1 μg even in pulsed magnetic fields. The sample mass was evaluated by the eigenfrequency shift. The micro-

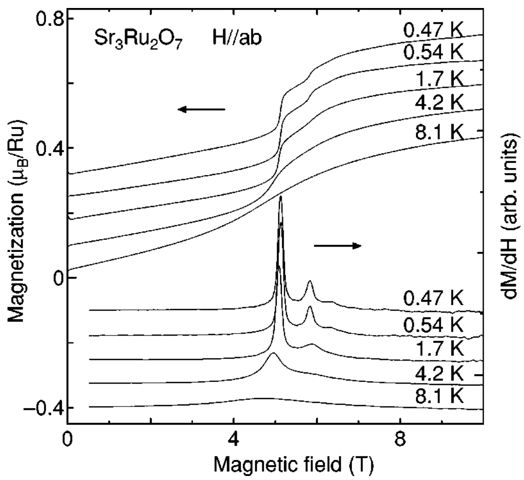

FIG. 1. Magnetization data of $Sr_3Ru_2O_7$ at various temperatures, together with dM/dH data. The magnetic fields were applied in $H\|ab$. The data are offset for clarity.

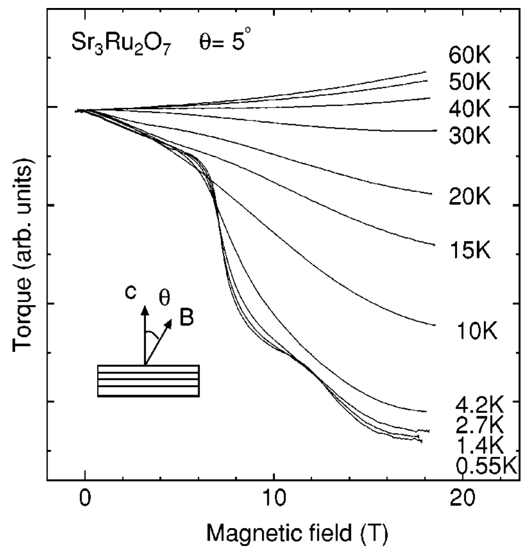

FIG. 2. Magnetic torque data of $Sr_3Ru_2O_7$ at different temperatures between 0.55 and 60 K. The magnetic field was applied in a direction tilted by 5° from the c axis. The data during a down sweep are shown.

cantilever was mounted on a rotating stage, and thus the field orientation was changed by a step of 1°. The exact field orientation within the ab plane was not determined, but the anisotropy of the susceptibility within the plane was shown to be less than 1% in the low-field region.

Shown in Fig. 1 are magnetization data of $Sr_3Ru_2O_7$ in magnetic fields along $H\|ab$, together with dM/dH data. The data during down sweeps are shown. No hysteresis was observed down to the lowest temperature of 0.47 K. Upon lowering temperature, a sudden magnetization increase at $\mu_0H_1 \sim 5.1$ T, characteristic for metamagnetic transition, was observed below ~10 K. In Ref. 7, Perry *et al.* observed a metamagnetic transition at 5.5 and 7.7 T in magnetic fields along $H\|ab$ and $H\|c$, respectively, at low temperature. The present result is consistent with their results.

Interestingly, an additional transition was found at $\mu_0H_2 \sim 5.8$ T at temperatures below 1.7 K, which were more clearly seen as a peak in dM/dH data. The transition becomes sharper with decreasing temperature, and exhibited a steplike structure attributable to the second metamagnetic transition. This result was not recognized in the previous magnetization measurements[7] probably due to the rather high temperature of 2.8 K, but was clearly seen in the low-temperature resistivity data. The magnetization jump associated with the second transition was evaluated to be about one-third of the first magnetization jump.

We also measured magnetization in pulsed fields along $H\|c$, but it exhibited a hysteresis loop caused by the eddy current effect, though a metamagnetic transition consistent with the previous report was observed.[13] In addition, the eddy current brings about the Joule heating effect, which becomes intolerable at low temperature. Since the effect of Joule heating is proportional to r^2 (r: the sample radius), it is important to reduce the sample dimensions to avoid these effects. A cantilever magnetometer is useful because the sample mass of 1 μg, which is four orders-of-magnitude smaller than that necessary for magnetization measurements, is enough for accurate measurements.

Figure 2 shows the magnetic torque data of $Sr_3Ru_2O_7$ at different temperatures. Magnetic fields were applied in a direction tilted by 5° from the c axis. Magnetic torque τ per unit volume is represented as $\tau = M \times H$, where M is the magnetization and H is the external field. In addition to a metamagnetic transition at ~7 T, an anomaly at ~12 T, attributable to the second metamagnetic transition, started to appear upon cooling. Similar to the magnetization measurements in $H\|ab$, no hysteresis was observed in the magnetic torque data in $H\|c$. It is of interest to point out that the sign of the low-field magnetic torque data changed between 20 and 30 K, where anomaly in the lattice constants[14] and sign change of the magnetoresistance[15] were reported.

In order to study the second transition in detail, we measured the magnetic torque of a sample at various tilt angles θ as shown in Fig. 3(a). The definition of θ is shown in the inset of Fig. 3(b). Arrows indicate the kinklike structure in the magnetic torque. In the field direction near $H\|ab$, both transition fields are located at similar positions, but they started to deviate when the magnetic field was tilted toward the c axis. The present results are summarized in Fig. 3(b). The first transition field $H_1(\theta)$ showed relatively isotropic behavior with respect to field orientation, while the second transition field $H_2(\theta)$ exhibited rather strong angle dependence but was not scaled with an in-plane field component.

Although metamagnetic transitions have been widely observed in a vast category of materials, the associated mechanisms are different between localized spin systems and itinerant electron systems. The former is explained by optimal spin alignment of localized moments subject to magnetic field, while itinerant metamagnetism as splitting of the up-spin band and the down-spin band triggered by magnetic field. The most typical systems exhibiting itinerant metamagnetism are $3d$ and $5f$ systems such as YCo_2,[16] $Co(S_{1-x}Se_x)_2$,[17] or UCoAl.[18] Common features to those compounds are a broad peak in temperature-dependent susceptibility and strongly exchange-enhanced paramagnetism.

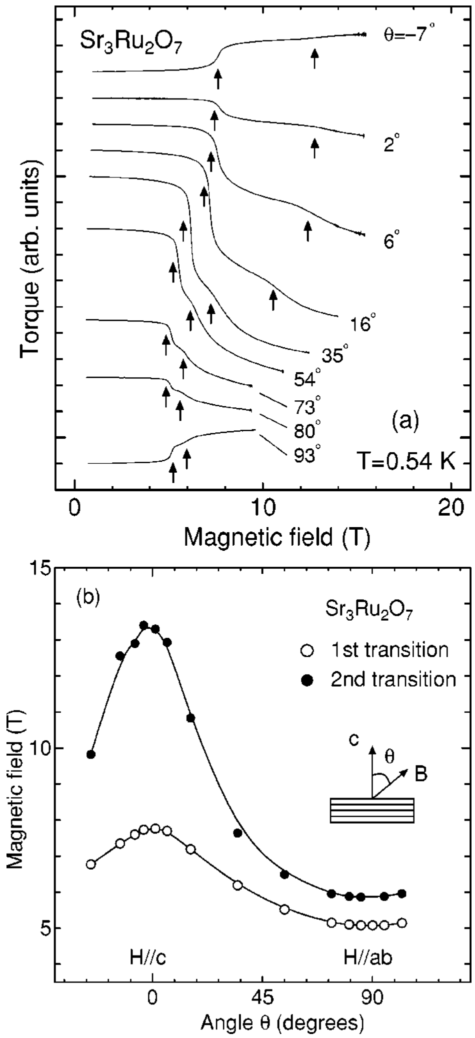

FIG. 3. (a) Magnetic torque data of $Sr_3Ru_2O_7$ at 0.54 K at different field angles. (b) Angle dependence of the transition fields obtained by magnetic torque measurements. The inset shows the definition of the angle θ.

In fact, both features are recognized in $Sr_3Ru_2O_7$ as reported by Ikeda *et al.*,[6,19] thus ensuring the analysis based upon spin-fluctuation theories of itinerant metamagnetism.

Itinerant metamagnetic transition is often explained based upon the phenomenological Landau theory with the Stoner model taking account of thermal spin fluctuation.[20] According to this, the free energy F is expanded with magnetization M up to the sixth order at $T=0$ as

$$F=\frac{1}{2}aM^2+\frac{1}{4}bM^4+\frac{1}{6}cM^6, \tag{1}$$

where a, b, and c are the Landau expansion coefficients determined by the band structure. By differentiating the equation with M, we obtain $H=aM+bM^3+cM^5$, from which the condition for first-order metamagnetic transition is derived, i.e., $a>0$, $b<0$, $c>0$, and $\frac{3}{16}<ac/b^2<\frac{9}{20}$. At finite temperature, the theory is expanded, taking account of thermal spin fluctuations, and successfully reproduces the maximum in susceptibility.

Here we compare the experimental results with the above-mentioned standard theory.[20] We note here that main features are dominated by the first metamagnetic transition H_1. First, ac/b^2 is related to the magnetic susceptibility χ by the equation $ac/b^2=(5/28)[1-\chi(0)/\chi(T_{\max})]^{-1}$. According to Ref. 6, $\chi(0)_{H\|ab}/\chi(T_{\max})_{H\|ab}$ and $\chi(0)_{H\|c}/\chi(T_{\max})_{H\|c}$ are 0.6 and 0.75, respectively, which in turn give $ac/b^2=0.44$ and 0.71, respectively. Therefore, the system is in fact on the verge of or slightly outside of the first-order metamagnetic transition line. The absence of hysteresis in the present data down to 0.5 K is consistent with this. Second, the transition field is theoretically obtained by the equation $H_c(0)=(21/40)\Delta M(0)\{[1/\chi(T_{\max})]-[1/21\chi(0)]\}$, where ΔM is the induced magnetization. The calculated transition field of $\sim$4.8 T is found to be in agreement with the observed field of $\mu_0H_1^{\|ab}\sim$ 5.1 T and $\mu_0H_1^{\|c}\sim$7.7 T [see Fig. 3(b)]. Although these estimations are rather crude, metamagnetic transition of $Sr_3Ru_2O_7$ is likely to be explained by the Stoner theory taking into account thermal fluctuations.

However, it is not straightforward to explain the following experimental results based upon the standard theory. One is that the temperature dependencies of the transition fields are opposite each other as clearly seen in the dM/dH data in Fig. 1; $dH_1/dT<0$ and $dH_2/dT>0$. According to the standard theory,[20] it is expected that the transition field increases with temperature, obeying T^2 dependence. In fact, this tendency is widely observed in $3d$ itinerant systems.[8] The reason for the opposite temperature dependence of H_1 may be relevant to the anomalous temperature dependence of the rotation angle of the RuO_6 octahedron:[14] the rotation angle increases with decreasing temperature, but starts to decrease below 40 K. Therefore, the bandwidth, sensitively affected by the rotation angle, becomes broader upon lowering temperatures below 40 K, and as a result higher magnetic field is required to induce the metamagnetic transition.

The other concerns the high-field magnetization of $Sr_3Ru_2O_7$ shown in Fig. 4. The magnetic field was applied in the ab plane. The magnetization continued to increase up to the highest magnetic field without saturation. The inset shows the Arrott plot (H/M vs M^2) of the data. It should be stressed that a linear relation after the transition was observed in a wide field range reaching 40 T, which is far above the transition field of $\sim$5 T. This result shows stark contradiction if we assume Eq. (1) to hold in the entire field region; due to the existence of the sixth-order term ($\frac{1}{6}cM^6$) in the free-energy expansion, the curve will have a substantial curvature in the high field region instead of a straight line. Concerning this, Takahashi and Sakai[21] proposed a different spin-fluctuation theory of itinerant metamagnetism based upon free-energy expansion up to the fourth-order term. This may give a satisfactory explanation, consistent with the observed Arrott plot.

Finally, we discuss possible origins of the second metamagnetic transition. As is well known, successive metamag-

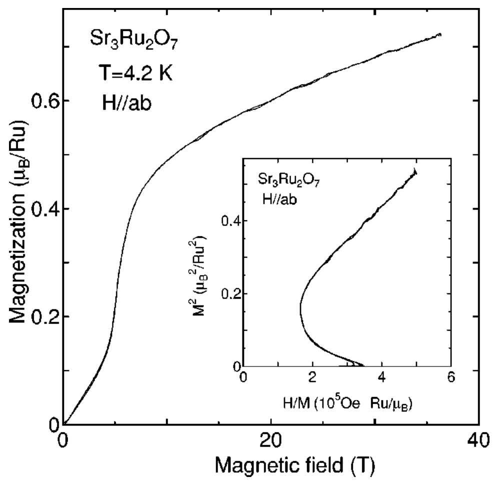

FIG. 4. High-field magnetization of $Sr_3Ru_2O_7$ in magnetic field along $H\|ab$ at 4.2 K. The inset is the Arrott plot of the data shown in the main frame.

netic transitions are observed in localized spin systems, but hardly observed in itinerant electron systems. A few examples are reported in $3d$ oxide compounds such as YCo_3 (Ref. 22) and $ThCo_5$.[23] In these systems, successive metamagnetic transitions have been explained based upon the fact that there are two or three different crystallographic Co sites. Then, the local density of states differs depending on the site, and as a result the different condition for the Stoner criteria gives different transition fields. In this argument, it is assumed that the Fermi-surface (FS) branches are well characterized by the orbital at respective sites due to weak hybridization.

In $Sr_3Ru_2O_7$, neutron-diffraction experiments have been carried out, and different results are obtained: Huang *et al.* reported a crystal structure of $Pban$,[24] in which two different Ru sites exist, but recent experimental results by Shaked *et al.*[25] claimed with better refinement that Ru sites are equivalent. According to the above-mentioned argument, the present results imply the existence of two crystallographic Ru sites supporting the former crystal structure. Based upon this model, however, it might be difficult to explain remarkable differences between the observed transitions such as the induced magnetization jump.

So far, we neglect the orbital degrees of freedom in the t_{2g} manifold, but this may also play a role in the metamagnetism. $Sr_3Ru_2O_7$ accommodates four electrons in the triply degenerate $4d$ orbitals (d_{xy}, d_{yz}, and d_{zx}), forming a low-spin state ($S=1$). In general, $4d$ electrons are more itinerant than $3d$ electrons, and the orbital origin of each FS branch is less clear due to strong hybridization. As discussed in Sr_2RuO_4,[26,27] however, the weak interaction between d_{xy} and d_{yz}/d_{zx} orbitals[28] allows us to assign the dominant orbital character to each FS branch. Then, respective branches could lead to different metamagnetic transition fields in analogy to orbital-dependent superconductivity in Sr_2RuO_4,[29] despite the rather simplified argument neglecting its strong itinerancy.

In summary, we reported high-field magnetization and magnetic torque data of $Sr_3Ru_2O_7$, and found a second metamagnetic transition in the higher-field region than the previously reported one. At present the origin of the second transition is not clear, but more detailed neutron-scattering experiments will give a clue to understanding this problem. We also discussed the metamagnetism of $Sr_3Ru_2O_7$ based upon spin-fluctuation theory and found that it displays typical behavior expected for itinerant electron metamagnetism except for a few observations such as the opposite temperature dependence of the first transition.

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