Evidence for the $\text{Sr}_2\text{RuO}_4$ intercalations in the $\text{Sr}_3\text{Ru}_2\text{O}_7$ region of the $\text{Sr}_3\text{Ru}_2\text{O}_7$ - $\text{Sr}_2\text{RuO}_4$ eutectic system

S. Kittaka\textsuperscript{a}, S. Yonezawa\textsuperscript{a}, H. Yaguchi\textsuperscript{a,b}, Y. Maeno\textsuperscript{a}, R. Fittipaldi\textsuperscript{a,c}, A. Vecchione\textsuperscript{a,c}, J.-F. Mercure\textsuperscript{d}, A. Gibbs\textsuperscript{d}, R. S. Perry\textsuperscript{d,e}, and A. P. Mackenzie\textsuperscript{d}

\textsuperscript{a}Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan
\textsuperscript{b}Department of Physics, Faculty of Science and Technology, Tokyo University of Science, Noda 278-8510, Japan
\textsuperscript{c}CNR-INFM Regional Laboratory “SuperMat” and Department of Physics, University of Salerno, I-84081 Baronissi (Sa), Italy
\textsuperscript{d}School of Physics and Astronomy, University of St. Andrews, St. Andrews KY16 9SS, UK
\textsuperscript{e}School of Physics, University of Edinburgh, Edinburgh EH9 3JZ, UK

E-mail: kittaka@scphys.kyoto-u.ac.jp

Abstract. Although $\text{Sr}_3\text{Ru}_2\text{O}_7$ has not been reported to exhibit superconductivity so far, ac susceptibility measurements revealed multiple superconducting transitions occurring in the $\text{Sr}_3\text{Ru}_2\text{O}_7$ region cut from $\text{Sr}_3\text{Ru}_2\text{O}_7$ - $\text{Sr}_2\text{RuO}_4$ eutectic crystals. Based on various experimental results, some of us proposed the scenario in which $\text{Sr}_2\text{RuO}_4$ thin slabs with a few layers of the $\text{RuO}_2$ plane are embedded in the $\text{Sr}_3\text{Ru}_2\text{O}_7$ region as stacking faults and multiple superconducting transitions arise from the distribution of the slab thickness. To examine this scenario, we measured the resistivity along the $ab$ plane ($\rho_{ab}$) using a $\text{Sr}_3\text{Ru}_2\text{O}_7$-region sample cut from the eutectic crystal, as well as along the $c$ axis ($\rho_c$) using the same crystal. As a result, we detected resistance drops associated with superconductivity only in $\rho_{ab}$, but not in $\rho_c$. These results support the $\text{Sr}_2\text{RuO}_4$ thin-slab scenario. In addition, we measured the resistivity of a single crystal of pure $\text{Sr}_3\text{Ru}_2\text{O}_7$ with very high quality and found that pure $\text{Sr}_3\text{Ru}_2\text{O}_7$ does not exhibit superconductivity down to 15 mK.

1. Introduction

The Ruddlesden-Popper series of layered perovskites $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$ is a fascinating research subject because the $n=1$ member $\text{Sr}_2\text{RuO}_4$ is now believed to be a spin-triplet superconductor \[1\] \[2\]. The $n=2$ member of this series $\text{Sr}_3\text{Ru}_2\text{O}_7$ is known to exhibit an enhanced Pauli paramagnetism with a metamagnetic transition \[3\] \[4\] \[5\]. Although crystals of $\text{Sr}_3\text{Ru}_2\text{O}_7$ are highly refined, the superconductivity in $\text{Sr}_3\text{Ru}_2\text{O}_7$ has not been discovered so far. Recently, $\text{Sr}_3\text{Ru}_2\text{O}_7$-$\text{Sr}_2\text{RuO}_4$ eutectic crystals were successfully grown \[6\] and surprisingly, multiple superconducting transitions were observed in the ac susceptibility measurements using a $\text{Sr}_3\text{Ru}_2\text{O}_7$-region sample cut from eutectic crystals \[7\], as presented in Fig. 1(a). However, it has been revealed that this superconductivity is not a bulk property of $\text{Sr}_3\text{Ru}_2\text{O}_7$ because these superconducting transitions are easily suppressed by small ac magnetic fields and no anomaly was observed in the specific heat \[7\]. The most plausible scenario of this superconductivity is that $\text{Sr}_2\text{RuO}_4$ thin slabs with a few layers of $\text{RuO}_2$ planes are embedded in the $\text{Sr}_3\text{Ru}_2\text{O}_7$ region and the multiple superconducting transitions arise from the distribution of the slab thickness.
In order to obtain additional evidence for the origin of the superconductivity observed in the Sr$_3$Ru$_2$O$_7$ region cut from eutectic crystals, we measured the resistivity along the $ab$ plane ($\rho_{ab}$) of a Sr$_3$Ru$_2$O$_7$-region sample cut from the eutectic crystal, which we designate below as a eutectic Sr$_3$Ru$_2$O$_7$ sample, as well as along the $c$ axis ($\rho_c$) of the same sample. In addition, we measured $\rho_c$ of a pure (i.e. non-eutectic) Sr$_3$Ru$_2$O$_7$ sample with very high quality in order to clarify whether or not pure Sr$_3$Ru$_2$O$_7$ exhibits superconductivity at low temperatures.

2. Experimental

Resistivity was measured using a conventional four-probe method with an ac current. Single crystals of the Sr$_3$Ru$_2$O$_7$-Sr$_2$RuO$_4$ eutectic system and those of single-phase Sr$_3$Ru$_2$O$_7$ were grown by a floating-zone method. The size of the eutectic Sr$_3$Ru$_2$O$_7$ sample was approximately 1.5 $\times$ 0.7 mm$^2$ in the $ab$ plane and 0.3 mm along the $c$ axis. The results of ac susceptibility and specific heat measurements using this eutectic Sr$_3$Ru$_2$O$_7$ sample were reported in Ref. [7]. The dimensions of the pure Sr$_3$Ru$_2$O$_7$ sample are 0.44 $\times$ 0.28 mm$^2$ in the $ab$ plane and 1.14 mm along the $c$ axis. The $\rho_{ab}$ measurements on the eutectic Sr$_3$Ru$_2$O$_7$ sample were performed down to 0.3 K with a $^3$He cryostat (Oxford Instruments, model Heliox VL). After the $\rho_{ab}$ measurements, we removed the electrical leads and attached another set of wires again on the same sample and performed $\rho_c$ measurements down to 0.1 K with an adiabatic demagnetization refrigerator (Cambridge Magnetic Refrigerator, mFridge50). The $\rho_c$ measurements using the pure Sr$_3$Ru$_2$O$_7$ sample were performed with a $^3$He-$^4$He dilution refrigerator (Cryoconcept, model DR-JT-S-100-10) down to 15 mK. In this study, we used a cylinder of permalloy (Hamamatsu Photonics K.K., E989-28) in order to reduce remanent fields such as the earth field.

3. Results and Discussion

3.1. Resistivity of the eutectic Sr$_3$Ru$_2$O$_7$

Figure 1(b) shows temperature dependence of $\rho_{ab}$ and $\rho_c$ of the eutectic Sr$_3$Ru$_2$O$_7$ sample. The values of $\rho_{ab}$ and $\rho_c$ are approximately 1 $\mu\Omega$cm and 300 $\mu\Omega$cm at 1.5 K, respectively. The in-plane resistivity is nearly the same as those of pure Sr$_3$Ru$_2$O$_7$ crystals with very high quality [5]. This low value of the in-plane resistivity indicates that Sr$_3$Ru$_2$O$_7$ in the eutectic system crystallized with high quality. Also, macroscopic Sr$_2$RuO$_4$ domains in the eutectic system are high quality because its $T_c$ is nearly 1.5 K [7], which is one of the best $T_c$ of Sr$_2$RuO$_4$ reported. It is interesting that both Sr$_2$RuO$_4$ and Sr$_3$Ru$_2$O$_7$ spontaneously crystallize with high quality in this eutectic system.

In $\rho_{ab}$ measurements, two clear resistance drops were observed at 1.05 and 1.32 K. These transition temperatures well coincide with those observed in the ac susceptibility [Fig. 1(a)]. However, in $\rho_c$ measurements, no obvious transition was observed. These results are consistent with the Sr$_2$RuO$_4$ thin-slab scenario because they imply that superconducting inclusions embedded in the eutectic Sr$_3$Ru$_2$O$_7$ are too thin along the $c$ axis to shortcircuit the current path along the interlayer direction. This behavior is in sharp contrast with $\rho_c$ for the Sr$_2$RuO$_4$-Ru eutectic system, in which the emergence of the “3-K” superconductivity in the interface of Ru lamellae results in a large drop in $\rho_c$. [8]. Although we observed two clear transitions in the $\rho_{ab}$ measurement using the eutectic Sr$_3$Ru$_2$O$_7$ sample, $\rho_{ab}$ does not become zero down to low temperatures. This non-zero resistivity implies that Sr$_2$RuO$_4$ inclusions in this eutectic Sr$_3$Ru$_2$O$_7$ sample do not completely form a path between the voltage contacts. The Sr$_2$RuO$_4$ inclusions are probably well separated in this sample. In some cases, eutectic Sr$_3$Ru$_2$O$_7$ samples exhibit zero resistivity (e. g., Ref. [9]), probably because Sr$_2$RuO$_4$ inclusions in such samples link a path between the voltage contacts. Now, on the basis of various experiments, we believe that the origin of the superconductivity observed in the eutectic Sr$_3$Ru$_2$O$_7$ sample is the presence of several monolayers of RuO$_2$ planes intercalated in Sr$_3$Ru$_2$O$_7$ as stacking faults. For example, two monolayers of RuO$_2$ planes intercalated in Sr$_3$Ru$_2$O$_7$ is schematically drawn in Fig. 1(c). In fact, such stacked monolayers of RuO$_2$ planes have been observed with a transmission electron microscope [9].
Figure 1. (a) Temperature dependence of the real part of the ac susceptibility for a eutectic Sr$_3$Ru$_2$O$_7$ sample with $\mu_0H_{ac}=0.58$ $\mu$T and $f=3011$ Hz (sample 1 in Ref. [7]). (b) Temperature dependences of the resistivity along the $c$ axis and that along the $ab$ plane for the eutectic Sr$_3$Ru$_2$O$_7$ sample measured at $f=89.1$ Hz ($I=0.5$ mA-rms for $\rho_{ab}$ and $I=0.1$ mA-rms for $\rho_c$). (c) A schematic image of two monolayers of RuO$_2$ planes (Sr$_2$RuO$_4$) intercalated in bilayers (Sr$_3$Ru$_2$O$_7$). Oxygens are located at the corner of the octahedra.

3.2. Resistivity of pure Sr$_3$Ru$_2$O$_7$ with high quality

We are also interested in the possibility of superconductivity in pure Sr$_3$Ru$_2$O$_7$. In order to examine the superconductivity in pure Sr$_3$Ru$_2$O$_7$, we consider it important (i) to use single crystals of single-phase Sr$_3$Ru$_2$O$_7$ with very high quality, (ii) to cool down the sample to sufficiently low temperature, and (iii) to perform measurements in zero field by excluding the geomagnetic field. Therefore, we measured $\rho_c$ down to 15 mK using a single crystal of pure Sr$_3$Ru$_2$O$_7$ with the in-plane residual resistivity of 0.4 $\mu$Ωcm, which is one of the highest-quality Sr$_3$Ru$_2$O$_7$ grown so far. In addition, by placing the sample in a cylinder of permalloy, we reduced the residual field to be lower than 0.1 $\mu$T.

Figure 2 shows temperature dependence of $\rho_c$ of the pure Sr$_3$Ru$_2$O$_7$ sample. The $\rho_c$ monotonically decreases with decreasing temperature and no anomaly indicating a superconducting transition was observed down to 15 mK. From this measurement, we conclude that pure Sr$_3$Ru$_2$O$_7$ does not become superconducting down to 15 mK.

4. Conclusion

We measured the resistivity along the $c$ axis as well as along the $ab$ plane of a eutectic Sr$_3$Ru$_2$O$_7$ sample. The resistance drops due to the multiple superconducting transitions were observed only for $\rho_{ab}$, but not for $\rho_c$. This result indicates that superconductors with thin thickness along the $c$ axis are embedded in the eutectic Sr$_3$Ru$_2$O$_7$. Now, we are convinced that the origin of the superconductivity observed in the eutectic Sr$_3$Ru$_2$O$_7$ sample is the presence of Sr$_2$RuO$_4$ inclusions embedded in Sr$_3$Ru$_2$O$_7$ as stacking faults. In order to search for superconductivity in pure Sr$_3$Ru$_2$O$_7$, we also measured the resistivity along the $c$ axis using a single crystal of best-quality pure Sr$_3$Ru$_2$O$_7$. However, no resistance anomaly associated with the superconducting transition was observed down to 15 mK. Therefore, we conclude that pure Sr$_3$Ru$_2$O$_7$ does not become superconducting down to 15 mK.
Figure 2. Temperature dependence of the resistivity along the $c$ axis of the pure $\text{Sr}_3\text{Ru}_2\text{O}_7$ sample with very high quality measured at $I=0.01$ mA-rms with $f=7$ Hz without the geomagnetic field. The inset shows the low-temperature region below 0.3 K.

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