Hydrostatic Pressure Study on 3-K Phase Superconductivity in Sr$_2$RuO$_4$-Ru Eutectic Crystals by AC Magnetic Susceptibility Measurements

Hiroshi Yaguchi, Hiromichi Watanabe and Akira Sakaue
Department of Physics, Faculty of Science and Technology, Tokyo University of Science, Noda 278-8510, Japan

Abstract.
We have investigated the effect of hydrostatic pressure on 3-K phase superconductivity in Sr$_2$RuO$_4$-Ru eutectic crystals by means of AC magnetic susceptibility measurements. We have found that the application of hydrostatic pressure suppresses the superconducting transition temperature $T_c$ of the 3-K phase with a pressure coefficient of $dT_c/dP \approx -0.2$ K/GPa, similar to the case of the 1.5-K phase. We have also observed that the effect of hydrostatic pressure on the 3-K phase seems to be elastic whilst that of uniaxial pressure is plastic.

1. Introduction
Sr$_2$RuO$_4$ is the first layered perovskite superconductor without copper [1], isostructural to the cuprate high-temperature superconductor La$_{2-x}$Ba$_x$CuO$_4$. More importantly, Sr$_2$RuO$_4$ is known to be a spin-triplet superconductor [2]. The original superconducting phase in Sr$_2$RuO$_4$ occurs with a sharp transition at a $T_c$ of 1.5K, called the 1.5-K phase. Besides this, the eutectic system, a two-phase composite structure of a single-crystalline Sr$_2$RuO$_4$ matrix and lamellar microdomains of ruthenium metal embedded in it, shows a broad superconducting transition with an enhanced onset of approximately 3 K, called the 3-K phase [3]. The 3-K phase transition is, on further cooling, followed by the superconducting transition to the original phase (the 1.5-K phase) in Sr$_2$RuO$_4$. (Figure 1 displays a SEM photograph of a polished surface of a Sr$_2$RuO$_4$-Ru eutectic crystal. Figure 2 shows typical AC magnetic susceptibility data for the 1.5-K phase in Sr$_2$RuO$_4$ and for the 3-K phase in Sr$_2$RuO$_4$-Ru.)

Whilst the origin of the 3-K phase still remains uncertain, 3-K phase superconductivity is non-bulk, and is believed to occur near Sr$_2$RuO$_4$ interfaces with Ru inclusions [4, 5, 6]. Sigrist and Monien’s phenomenological theory [7] within the framework of Ginzburg Landau formalism, which assumes spin-triplet pairing similar to Sr$_2$RuO$_4$, successfully describes important aspects of the 3-K phase. Examples of experiments in support of the 3-K phase being unconventional superconductivity include tunnelling measurements on S/N junctions obtaining zero bias conductance peaks [8, 9, 10], and transport measurements on micro-fabricated samples observing hysteresis in the $I$-$V$ characteristics attributable due to the chiral $p$-wave state [11].

Recent uniaxial-pressure studies on the 3-K phase have revealed that both in-plane pressure (parallel to the $ab$-plane) and out-of-plane pressure (parallel to the $c$-axis) enhance 3-K phase superconductivity in Sr$_2$RuO$_4$-Ru eutectic crystals, particularly in volume fraction [12, 13]. The
enhancement by in-plane pressure is even greater than that by out-of-plane pressure. This fact probably supports the scenario that the rotation of the RuO$_6$ octahedra plays an important role in the occurrence of the 3-K phase[7] because in-plane pressure will affect the Ru-Ru lattice constant in the $ab$-plane directly, and the out-of-plane pressure will affect it indirectly.

In our recent hydrostatic pressure study, we used SQUID magnetometer (MPMS, Quantum Design) to measure the DC magnetisation at temperatures down to 1.8 K, and found that the shielding fraction or the apparent superconducting volume fraction associated with the 3-K phase is suppressed by the application of hydrostatic pressure [14]. In the present work, we have investigated the AC magnetic susceptibility of Sr$_2$RuO$_4$-Ru eutectic crystals under hydrostatic pressure at temperatures down to 1.2 K to obtain insight into the mechanism of 3-K phase superconductivity.

![SEM image](image.png)

**Figure 1.** SEM image of a polished surface of a Sr$_2$RuO$_4$-Ru eutectic crystal. The darker part corresponds to Sr$_2$RuO$_4$ and the brighter part corresponds to Ru inclusions.

![AC susceptibility data](image.png)

**Figure 2.** Example of AC susceptibility data for the 1.5 K phase in single-phase Sr$_2$RuO$_4$ and for the 3-K phase in the eutectic system Sr$_2$RuO$_4$-Ru.

2. Experimental

The samples used in the present study were Sr$_2$RuO$_4$-Ru eutectic crystals grown by a floating zone method [15], which were cut and polished such that approximate dimensions of the samples were $1 \times 1 \times 2$ mm$^3$ with the longest dimension parallel or perpendicular to the $c$-axis. We used a mutual inductance method to measure AC susceptibility at temperatures down to 1.2 K with an AC measurement field of 2 $\mu$T-rms at a frequency of 777 Hz. Hydrostatic pressure was applied using a beryllium-copper clamp cell with Daphne 7373 oil (Idemitsu Co. Ltd.) as the pressure transmitting medium; pressures were determined from the pressure dependence of the superconducting critical temperature of tin [16].

3. Results and Discussion

Figure 3 shows the temperature dependence of the real and imaginary parts of the AC magnetic susceptibility of a Sr$_2$RuO$_4$-Ru eutectic crystal under hydrostatic pressure, with the AC field along the $c$-axis. These measurements were performed in the order of increasing applied pressure, and a subsequent measurement was done after the applied pressure was released. We have made a similar set of measurements on four eutectic crystals. All of the data in Fig. 3 are,
for convenience, multiplied by a common factor such that the real part of the 0-GPa (before) trace tends to −1 at temperatures well below $T_c$. Similar to Fig. 2, rather broad changes associated with 3-K phase superconductivity are seen in both of the real and imaginary parts of the AC susceptibility, and those changes shift towards lower temperatures with increasing hydrostatic pressure; 3-K phase superconductivity is suppressed by the application of hydrostatic pressure. However, the shielding fraction, corresponding to the drop in the real part, hardly changes with pressure. These results enable the pressure coefficient to be determined as $\frac{dT_c}{dP} \approx -0.2$ K/GPa; the dissipation peak in the imaginary part associated with the 3-K phase has been used to define $T_c$ of the 3-K phase. Measurements on the other samples also yield close values for the pressure coefficient. (We here note that hydrostatic pressure studies on Sr$_2$RuO$_4$ obtained the pressure coefficient of the superconducting transition temperature for the 1.5-K phase to be $-0.2$ K/GPa [17, 18, 19], which is very close to the value for the 3-K phase.)

Therefore, the basic relation in tetragonal symmetry

$$\frac{dT_c}{dP} = \frac{dT_c}{dP_{||a}} + \frac{dT_c}{dP_{||b}} + \frac{dT_c}{dP_{||c}} = 2\times\frac{dT_c}{dP_{||a}} + \frac{dT_c}{dP_{||c}},$$

(1)

constrains at least either $dT_c/dP_{||a}$ or $dT_c/dP_{||c}$ of the 3-K phase to be negative. Nevertheless, it has been reported that uni-axial pressure both parallel and perpendicular to the c-axis greatly enhances 3-K phase superconductivity in eutectic crystals [12, 13], showing an apparent disagreement with the present hydrostatic-pressure study.

**Figure 3.** The real (left) and imaginary (right) parts of the AC susceptibility of a Sr$_2$RuO$_4$-Ru eutectic crystal at various hydrostatic pressures. The bottom trace of each panel was taken after the applied pressure was released.
The above apparent discrepancy is very likely to be due to the difference between the natures of hydrostatic and uni-axial pressure effects on this eutectic system. The effect of hydrostatic pressure is modest and appears to be elastic since the 0-GPa traces of “before” and “after” in Fig. 3 well coincide with each other. By contrast, the effect of uniaxial pressure has been reported to be dramatic and plastic; the effect remains even after the applied pressure is released [13]. These facts presumably support the idea that uniaxial pressure induces and/or promotes the rotation of the RuO$_6$ octahedra in Sr$_2$RuO$_4$-Ru eutectic crystals [12, 13, 20], leading to large enhancement of 3-K phase superconductivity, whilst hydrostatic pressure is unlikely to affect such rotation considerably.

4. Summary
In summary, we have found that hydrostatic pressure suppresses the superconducting transition temperature $T_c$ of the 3-K phase in Sr$_2$RuO$_4$-Ru crystals, and have evaluated its pressure coefficient $dT_c/dP$ to be approximately $-0.2$ K/GPa, very close to that of the 1.5-K phase. It has turned out that hydrostatic pressure effect, which suppresses the 3-K phase in a rather modest manner, presents a striking contrast with the uniaxial pressure effect largely enhancing the 3-K phase. Also the effect of hydrostatic pressure on the 3-K phase in Sr$_2$RuO$_4$-Ru eutectic crystals appears to be elastic, in contrast to uni-axial pressure causing plastic deformation to occur in eutectic crystals. Such elastic deformation will be related to the rotation of the RuO$_6$ octahedra in Sr$_2$RuO$_4$-Ru eutectic crystals.

Acknowledgements
We thank S. Kawada, S. Suzuki, S. Kittaka and Y. Maeno for their supports and discussions.

References
[1] Maeno Y, Hashimoto H, Yoshida K, Nishizaki S, Fujita T, Bednorz J G and Lichtenberg F 1994 *Nature* **372** 532
[2] Mackenzie A P and Maeno Y 2003 *Rev. Mod. Phys.* **75** 657
[3] Maeno Y, Ando T, Mori Y, Ohnishi E, Ikeda S, Nishizaki S and Nakatsuji S 1998 *Phys. Rev. Lett.* **81** 3765
[4] Ando T, Akima T, Mori Y and Maeno Y 1999 *J. Phys. Soc. Jpn.* **68** 1651
[5] Yaguchi H, Wada M, Akima T, Maeno Y and Ishiguro T 2003 *Phys. Rev. B* **67** 214519
[6] Ying Y A, Xin Y, Clouser B W, Hao E, Staley N E, Myers R J, Allard L F, Fobes D, Liu T, Mao Z Q and Liu Y 2009 *Phys. Rev. Lett.* **103** 247004
[7] Sigrist M and Monien H 2001 *J. Phys. Soc. Jpn.* **70** 2409
[8] Mao Z Q, Nelson K D, Jin R and Liu Y 2001 *Phys. Rev. Lett.* **87** 037003
[9] Kawamura M, Yaguchi H, Kikugawa N, Maeno Y and Takayanagi H 2005 *J. Phys. Soc. Jpn.* **74** 531
[10] Yaguchi H, Takizawa K, Kawamura M, Kikugawa N, Maeno Y, Meno T, Akazaki T, Sembak K and Takayanagi H 2006 *J. Phys. Soc. Jpn* **75** 125001
[11] Kambara H, Kashiwaya S, Yaguchi H, Asano Y, Tanaka Y and Maeno Y 2008 *Phys. Rev. Lett.* **101** 267003
[12] Yaguchi H, Kittaka S and Maeno Y 2000 *J. Phys.: Conf. Seri.* **150** 052285
[13] Kittaka S, Yaguchi H and Maeno Y 2009 *J. Phys. Soc. Jpn.* **78** 103705
[14] Yaguchi H, Kawada S, Watanabe H and Suzuki S 2010 *J. Phys. Soc. Jpn.* **79** 125004
[15] Mao Z Q, Maeno Y and Fukazawa H 2000 *Mat. Res. Bull.* **35** 1813
[16] Smith T F and Chu C W 1967 *Phys. Rev.* **159** 353
[17] Shirakawa N, Murakat K, Nishizaki S, Maeno Y and Fujita T 1997 *Phys. Rev. B* **56** 7890
[18] Forsythe D, Julian S R, Bergemann C, Pugh E, Steiner M J, Alireza P L, McMullan G J, Nakamura F, Haselwimmer R K W, Walker I R, Saxena S S, Lonzarich G G, Mackenzie A P, Mao Z Q and Maeno Y 2002 *Phys. Rev. Lett.* **89** 166402
[19] Svitalskiy O, Headley S, Tozer S W, Palm E C, Murphy T P, Shul' yazev D and Suslov A V 2008 *Phys. Rev. B* **77** 052502
[20] Kittaka S, Taniguchi H, Yonezawa S, Yaguchi H and Maeno Y 2010 *Phys. Rev. B* **81** 180510(R)