Field-temperature phase diagram of superconductivity in Sr$_2$RuO$_4$-Ru under out-of-plane uniaxial pressure

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Abstract. One of the interesting issues on the spin-triplet superconductor Sr$_2$RuO$_4$ ($T_c = 1.5$ K) is the origin of the $T_c$ enhancement up to about 3 K in the eutectic system Sr$_2$RuO$_4$-Ru. We have recently revealed a striking fact that $T_c$ of pure Sr$_2$RuO$_4$ is also enhanced up to 3.2 K by uniaxial pressure along the $c$ axis ($P_{\parallel c}$). When $P_{\parallel c}$ is applied to Sr$_2$RuO$_4$-Ru, there is a crossover at $P_{\parallel c}^* \sim 0.4$ GPa from behavior attributable to interfacial superconductivity (SC) to behavior similar to that in pure Sr$_2$RuO$_4$ under $P_{\parallel c}$. We focus on the field-temperature phase diagrams of Sr$_2$RuO$_4$-Ru above and below $P_{\parallel c}^*$. We revealed that the $H$-$T$ curves of Sr$_2$RuO$_4$-Ru both above and below $P_{\parallel c}^*$ are concave-up. This fact suggests that the $P_{\parallel c}$-originated SC above $P_{\parallel c}^*$ as well as the interfacial SC below $P_{\parallel c}^*$ are granular-like. We also found that the $H$-$T$ curves of these SCs do not scale with $T_c$’s, which is possibly related to a difference in the spatial distribution of superconducting regions.

1. Introduction

Sr$_2$RuO$_4$ has been attracting much interest as a leading candidate of a spin-triplet chiral $p$-wave superconductor [1, 2]. One of the interesting issues for this compound is the enhancement of the superconducting transition temperature $T_c$ in the eutectic system Sr$_2$RuO$_4$-Ru; the onset $T_c$ of Sr$_2$RuO$_4$-Ru is $\sim 3$ K, while $T_c$ of pure Sr$_2$RuO$_4$ and Ru are 1.5 K and 0.49 K, respectively [3]. In the eutectic system, lamellae of Ru metal, whose typical dimension is $10 \times 10 \times 1 \mu$m$^3$, are embedded in a Sr$_2$RuO$_4$ single crystal with stripe patterns. The nonbulk nature of this SC was revealed by the absence of specific-heat anomaly around 3 K [4]. The zero-bias conductance peak observed above 1.5 K indicates that the interface between Sr$_2$RuO$_4$ and Ru is superconducting and that this SC is non $s$-wave [5, 6]. The anisotropic upper critical field similar to that of the 1.5-K SC in Sr$_2$RuO$_4$ indicates that it is the Sr$_2$RuO$_4$ region around the interfaces that becomes superconducting [7].

We have investigated uniaxial pressure effects on Sr$_2$RuO$_4$ and Sr$_2$RuO$_4$-Ru in order to clarify the mechanism of the $T_c$ enhancement, because a possible origin of the $T_c$ enhancement is anisotropic lattice distortion around the interfaces. We reported that $T_c$ of “pure” Sr$_2$RuO$_4$...
is enhanced up to 3.2 K under pressure along the $c$ axis ($P_{\parallel c}$) \[8\]. Here, “pure” means that the sample contains almost no Ru inclusions. The result strongly supports our scenario that the anisotropic lattice distortion plays an important role in the $T_c$ enhancement. We also clarified that $P_{\parallel c}$ substantially enhances the shielding fraction of the 3-K SC in the eutectic system, based on DC magnetization measurements \[9, 10\]. Moreover, in Sr$_2$RuO$_4$-Ru under $P_{\parallel c}$, we found a crossover at $P_{\parallel c}^* \sim 0.4$ GPa from behavior attributable to interfacial SC to behavior similar to that in pure Sr$_2$RuO$_4$ under $P_{\parallel c}$ \[8\].

In this article, we report results of AC susceptibility measurements of Sr$_2$RuO$_4$-Ru under $P_{\parallel c}$ for several DC magnetic fields. We then compare the field-temperature ($H$-$T$) phase diagram of the $P_{\parallel c}$-originated SC above $P_{\parallel c}^*$ with that of the interfacial SC below $P_{\parallel c}^*$.

2. Experimental

Uniaxial pressure was applied along the $c$ axis at room temperature using a piston-cylinder-type pressure cell (Fig. 1(a)). The room-temperature pressure value was confirmed to be in a reasonable agreement with low-temperature pressure value determined by the superconducting transitions of tin and lead \[11\]. When the screws at both ends are tightened, the dish-shaped springs hold the pressure. The pistons press a sample directly; no pressure medium is used. The inner parts and the outer body of the cell are made of Be-Cu and polybenzimidazole (hard plastic), respectively.

The single crystals used in this study were grown in the floating zone method with Ru self-flux \[12\]. The dimensions of the sample reported here are $1.70 \times 1.46$ mm$^2$ in the $ab$ plane and 0.40 mm along the $c$ axis. The top and bottom surfaces were polished to be parallel to each other. The side surfaces were covered with epoxy (Stycast 1266, Emerson-Cuming) to prevent a breakdown of the sample (Fig. 1(b)) \[10\]. A space between the epoxy and the inner wall of the cell allows the epoxy to spread freely under pressure.

\[\chi''(T) = \frac{1}{i\chi''(T)} - i\chi''(T)\]

Figure 1. Schematic diagram of (a) the pressure cell, (b) the top and side views of the sample surrounded by Stycast 1266, and (c) the set-up for the AC susceptibility measurement.
than 5 degrees. $\chi_{AC}$ is related to the read-out voltage of the LIA ($V_{LIA} = V_x + iV_y$) as

$$\chi_{AC} = -iC_1V_{LIA}/fH_{AC}^0 + C_2.$$  

Here, $H_{AC}^0$ and $f$ are the amplitude and frequency of the applied AC magnetic field, and $C_1$ and $C_2$ are complex coefficients. The values of $C_1$ and $C_2$ were chosen so that $\chi'(4 K) = 0$ and $\chi'(0.3 K) = -1$ under $P_{\parallel c} = 0$ and $H_{DC} = 0$ for each sample; thus $|\chi'|$ corresponds to the AC shielding fraction. For the $\chi_{AC}$ curves under $P_{\parallel c} > 0$ or $H_{DC} > 0$, $C_1$ and $C_2$ determined under $0$ GPa and $0$ T were used.

3. Results and Discussion

Figure 2 represents the temperature dependence of $\chi_{AC}$ of Sr$_2$RuO$_4$-Ru under $P_{\parallel c} = 0$ and $P_{\parallel c} = 0.5$ GPa > $P^*$. An enlarged view around the superconducting onset is shown in Fig. 2(b). The shielding fraction between 1.7 K and 3.0 K is enhanced, consistent with previous reports [9, 10]. The decrease of the volume fraction at lower temperatures is attributable to partial destruction of SC due to excess or inhomogeneity of distortion as well as to cracks in the sample [8]. Next, we measured $\chi_{AC}$ of the same sample at 0.5 GPa under several DC magnetic fields (Fig. 3). In the $P_{\parallel c}$-enhanced shielding region ($0 < |\chi'| < 0.05$), $\chi'(T)$ curves exhibit parallel shift to lower temperature with increasing field (Fig. 3(b)). This behavior is different from that of the interfacial SC below $P^*_{\parallel c}$: the interfacial SC does not exhibit parallel shift, as exemplified in the region of $0 < |\chi'| < 0.6$ in Fig. 5 of Ref. [13].

![Figure 2](image)

**Figure 2.** Zero-field temperature dependence of the real part of the AC susceptibility of Sr$_2$RuO$_4$-Ru under $P_{\parallel c} = 0$ and $P_{\parallel c} = 0.5$ GPa. (a) the entire behavior and (b) an enlarged view around the superconducting onset. These data were obtained with $\mu_0H_{AC} = 2 \mu$T-rms and $f = 293$ Hz.

In order to illustrate the difference between the 3-K SC in Sr$_2$RuO$_4$-Ru above $P^*_{\parallel c}$ ($P_{\parallel c}$-originated SC) and that below $P^*_{\parallel c}$ (interfacial SC), we present in Fig. 4 the $H$-$T$ phase diagram of these SCs. Figure 4 also contains the $H$-$T$ phase diagram of the bulk SC of Sr$_2$RuO$_4$ at $P_{\parallel c} = 0$ [15]. All the $H$-$T$ phase diagrams are determined by $\chi_{AC}$. We define $T_c$ in the following two ways: $T_c^{\text{cross}}$ is defined as the temperature of the intersection of the extrapolation of the most rapidly changing part of $\chi'$ and that of the normal state $\chi'$ as shown in Fig. 3(b), and $T_c^{\text{onset}}$ is defined as the temperature at which $\chi'$ reaches $-0.005$. We used the steepest slope in $0 < |\chi'| < 0.05$ to determine $T_c^{\text{cross}}$ of $P_{\parallel c}$-originated SC, so that $T_c^{\text{cross}}$ captures the characteristics of the $P_{\parallel c}$-originated SC. For the same reason, the steepest slope in $0 < |\chi'| < 0.6$ is used for $T_c^{\text{cross}}$ of the interfacial SC.

We have revealed that (1) the $H$-$T$ curves of the $P_{\parallel c}$-originated SC and the interfacial SC are concave up with decreasing temperature, while that of the bulk SC of Sr$_2$RuO$_4$ is convex up,
Figure 3. Temperature dependence of the real part of the AC susceptibility of Sr$_2$RuO$_4$-Ru at $P_{||c} = 0.5$ GPa under different $H_{\text{DC}}$'s, measured with $\mu_0H_{\text{AC}} = 2 \mu\text{T}$-rms and $f = 293$ Hz: (a) the entire behavior and (b) an enlarged view of the $P_{||c}$-enhanced shielding region.

Figure 4. Comparison of $H$-$T$ phase diagrams for $H \parallel c$ based on two different $T_c$ definitions: $T^\text{cross}_c$ and $T^\text{onset}_c$ (see text). Solid and dashed lines indicate fitting curves with Eq. (1) for a model of an assembly of intergranular Josephson junctions [14]. These curves reproduce the concave-up behavior of the $P_{||c}$-originated SC above $P^*_c$, as well as that of the interfacial SC below $P^*_c$.

and that (2) the slopes of the $H$-$T$ curve of the $P_{||c}$-originated SC are smaller than those of the interfacial SC resulting in a crossing at around 0.04 T. Note that these features are not affected by the definitions of $T_c$.

It is known that $H$-$T$ curves of granular superconductors exhibit concave-up behavior with decreasing temperature. A calculation of the effective critical field $H_j$ of an assembly of intergranular Josephson junctions revealed that $H_j$ exhibits $1/T$ dependence in the vicinity of $T_c(H=0)$ [14, 16]. Therefore, in order to clarify the origin of the concave-up behavior for the $P_{||c}$-originated SC, we fitted the equation

$$\mu_0H_j(T) = a \left( \frac{1}{T} - \frac{1}{T_c(H=0)} \right)$$

(1)

to the data in the range $0.5T_c(H=0) < T < T_c(H=0)$. The well-fitted results shown in Fig. 4 imply that both the $P_{||c}$-originated SC and the interfacial SC have granular-like features. The difference in the slope of the $H$-$T$ curve is possibly related to certain changes in the spatial distribution of superconducting regions caused by pressure, such as an increase of intergranular...
distance or a change in the shape of superconducting grains themselves. We note, however, that possibilities of extrinsic origins such as cracks have not been excluded.

4. Conclusion
We measured the AC susceptibility of $\text{Sr}_2\text{RuO}_4$-Ru under uniaxial pressure along the $c$ axis ($P_{\|c}$) in several DC magnetic fields. We revealed the field-temperature ($H$-$T$) phase diagrams of $\text{Sr}_2\text{RuO}_4$-Ru above and below $P_{\|c}^*$. We clarified that the $H$-$T$ phase diagrams are concave up with decreasing temperature both in the $P_{\|c}$-originated SC above $P_{\|c}^*$ and in the interfacial SC below $P_{\|c}^*$, while the phase diagram is convex up in the bulk SC of $\text{Sr}_2\text{RuO}_4$. This result suggests that the $P_{\|c}$-originated SC is somewhat inhomogeneous as well as the interfacial SC. We also revealed that the slope of the $H$-$T$ curve is smaller in the $P_{\|c}$-originated SC than in the interfacial SC. This is probably related to a spatial-distribution change of superconducting grains.

In the future, measurements in higher fields would be interesting, because a crossover from granular-like behavior to a bulk-like behavior of the $H_j(T)$ curve is proposed in Ref. [8] when the field reaches the bulk $H_{c2}$. Another future direction is to try to reduce cracks in the sample caused by the pressure. Such cracks prevent us from more quantitative analyses. A reduction of cracks is probably achieved by controlling the amount of epoxy surrounding the sample.

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