Superconducting behaviour via percolation in Sr$_2$RuO$_4$-Sr$_3$Ru$_2$O$_7$ eutectic crystals

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Abstract. We report on superconducting behavior of Sr$_3$Ru$_2$O$_7$ macrodomain taken out from Sr$_2$RuO$_4$-Sr$_3$Ru$_2$O$_7$ eutectic crystals. Transport measurements performed down to 300 mK provide evidence of a supercurrent flowing through the whole sample. Structural and compositional analyses are used to estimate the percentage of possible Sr$_2$RuO$_4$ inclusions. On the base of a model that describes the system as a proximity network made of superconducting Sr$_2$RuO$_4$ grains dispersed in the normal Sr$_3$Ru$_2$O$_7$ matrix we determine the conditions for having a percolating superflow through the system.

1. Introduction

The Ruddlesden-Popper ruthenate series, Sr$_{n+1}$Ru$_n$O$_{3n+1}$, has become a prototype for exploring unconventional phenomena as due to a subtle interplay between anisotropy, strong correlations, and multiband effects. The $n = 1$ member, Sr$_2$RuO$_4$, is a spin-triplet superconductor[1], the infinite-layer SrRuO$_3$ is an itinerant ferromagnetism, while the bilayered Sr$_3$Ru$_2$O$_7$ manifests a complex metamagnetic behaviour[2]. Recently, a route for tailoring new materials has been found by studying Sr-based layered ruthenates eutectic systems. One of the main motivations for carrying out eutectic growth is the possibility to develop in-situ composite materials where the properties of the distinct constituents merge to a variable extent into those of a single material. The Sr$_2$RuO$_4$-Sr$_3$Ru$_2$O$_7$ eutectic turns out to be a suitable system for studying the interaction between an unconventional superconductor and a metal on the verge of a magnetic instability, whose matching occurs at the nanoscale with high quality interfaces[3, 4, 5].

Here, we investigate the superconducting behavior of Sr$_3$Ru$_2$O$_7$ region taken out from a Sr$_2$RuO$_4$-Sr$_3$Ru$_2$O$_7$ eutectic crystal. We present evidence of a supercurrent flowing through a macroscopic Sr$_3$Ru$_2$O$_7$ domain as cut from the eutectic. The compositional analysis is then used to get indications about defect distributions of Sr$_2$RuO$_4$ grains within the Sr$_3$Ru$_2$O$_7$ normal host. Finally, a theoretical simulation, based on a model where the Sr$_3$Ru$_2$O$_7$ domain is made of a proximity network with variable size and percentage of Sr$_2$RuO$_4$ defects, is considered to yield definite limits on the application of the percolation scenario.
2. Sample preparation and measurements set up
The eutectic crystals involved in these experiments were grown using the floating zone technique. Details of the growth of these crystals and the performed compositional analysis are reported elsewhere[3, 4]. To probe the transport properties of Sr$_3$Ru$_2$O$_7$ domains taken out from the eutectic crystals, it was necessary to select crystals where the two phases were separated and no intermixing between them was detectable, at least using polarized light optical microscopy (PLOM) and scanning electron microscopy. Moreover, a careful inspection by PLOM on well polished surfaces along both a-b and a-c planes showed the presence of clusters of tens of µm$^2$ size randomly dispersed throughout the surface over distances ranging from 10 to 50 µm (Fig. 1). The transport measurements were made using a four probe technique. We measured down to 0.3 K using a Heliox $^3$He refrigerator and the measurement environment was shielded with superconducting and cryoperm shields.

3. Results and discussion
We firstly assess by X-ray and EDS analysis the good quality of the samples checking for the amount of Sr$_2$RuO$_4$ present in the selected crystals. Fig. 2 shows weak (002) peak of Sr$_2$RuO$_4$ visible in the X-ray diffraction pattern for the a-b plane of sample in Fig. 1. Then, we have searched for Sr$_2$RuO$_4$ grains through elemental composition analysis by an Energy Dispersive X-ray Spectrometer (EDS). To statistically explore the chemical composition of the samples, about 500 EDS spots analysis have been performed over the whole sample surface. The EDS data set is represented in the histogram in Fig. 3, where the number of events versus the corresponding value 2 N(Ru)/N(Sr) is reported. The 2 N(Ru)/N(Sr) values distribution follows a Gaussian distribution, whose mean value and standard deviation are 2 N(Ru)/N(Sr) = 1.30 ± 0.02. Taking into account that pure Sr$_3$Ru$_2$O$_7$ and Sr$_2$RuO$_4$ phases are characterized by 2 N(Ru)/N(Sr)
where the grains of Sr$_2$RuO$_4$ embedded within the Sr$_3$Ru$_2$O$_7$ matrix are responsible for the superconducting behavior rather than the Sr$_3$Ru$_2$O$_7$ itself. We have performed a statistical simulation to individuate which size of the Sr$_2$RuO$_4$ grains can be consistent with the transport analysis. The working hypotheses are the following: 1) the simulated system has a size of 1$\mu$m$^3$, 2) different percentages $x$ of Sr$_2$RuO$_4$ grains are considered, 3) the size of the Sr$_2$RuO$_4$ grains is parameterized in terms of the linear dimensions $d_a$, $d_b$, and $d_c$ such as the volume is given (for symmetry one can assume $d_a=d_b$), 3) two Sr$_2$RuO$_4$ grains are proximity connected if their distance is not larger than the coherence length within the Sr$_3$Ru$_2$O$_7$ host along the respective symmetry axis ($\xi^a_N$; $\xi^b_N$; $\xi^c_N$ where $\xi^a_N \approx 40$ T$^{-1}$ K nm and $\xi^c_N \approx 2.5$ T$^{-1}$ K mm; $\xi^a_N = h\nu_F^2 / 2\pi k_B T$, $i = a, c$ being the axis directions, respectively), 4) the grains are randomly distributed without any spatial correlation between them, 5) the critical size extracted from the simulation is associated with the grain’s distribution within the given volume such as the percolation threshold is obtained. We begin with three different percentages $x = 0.03, 0.10, 0.15$ of Sr$_2$RuO$_4$ dispersed in the eutectic Sr$_3$Ru$_2$O$_7$ assuming a given value $d_c = 1$nm for the grain size along the c-axis and $\xi^c_N = 40$nm for the normal in plane coherence length as provided by the

**Figure 3.** SEM image of a polished $a-b$ plane of a Sr$_3$Ru$_2$O$_7$ domain taken out from the eutectic crystals and its related statistical EDS compositional analysis. The histogram represents the number of EDS analyses versus different values of $k = 2 N$(Ru)/N(Sr). The black line is the fitting gaussian curve with a mean value of 1.30 ± 0.02

**Figure 4.** I-V characteristics of Sr$_3$Ru$_2$O$_7$ region acquired at $T=1.30$ K varying the magnetic field applied parallel to the c-axis.
percolating condition is not favoured by the growing of the Sr

4. Conclusion

In conclusion we have considered the superconducting behaviour of Sr

![Image](image_url)

**Figure 5.** Evolution of the c-axis coherence length for the normal part of the superconducting network at $\xi_{N,c} = 40$nm as a function of $d_a$ (a) and $d_c$ (b) for $d_c = 1$nm (a) and $d_a = 20$nm (b). $P$ and $D$ stand for percolating and disconnected distribution.

theoretical estimation at $T = 1$ K. Hence, the critical amplitude $\xi_{N,p}$ for the coherence length in the normal host can be extracted as a function of the linear size $d_a$ of the Sr$_2$RuO$_4$ grain. In Fig. 5 it is reported the behaviour of $\xi_{N,c}$ versus $d_a$. As one expects, the general trend indicates a growth of the percolating coherence length as a function of the in plane grain dimensions at any value of the Sr$_2$RuO$_4$ percentage. The change in the quantity of dispersed Sr$_2$RuO$_4$ grains does not modify the lower limit for the percolating c-axis coherence length. Otherwise, for values of $x$ that are greater than $\sim 0.03$ there is an increase of a-b plane grain size below which the percolation threshold of the c-axis coherence length keeps a value of the order of $\sim 10$nm. In this context, if the percentage of Sr$_2$RuO$_4$ grains is about 10% and the grain size is such that $d_a \leq 80$nm and $d_c \sim 1$ nm, then below a percolating temperature of $T_p \sim 0.25$ K one would observe a non zero supercurrent flowing through the sample at macroscopic scale.

By varying the size along of the Sr$_2$RuO$_4$ along the c-axis it is possible to obtain the evolution of the correspondent percolating coherence length. In Fig. 5b, we have reported the behaviour of $\xi_{N,p}$ as a function of $d_a$ at different values of $x$ assuming a given grain size in the a-b plane (i.e. $d_a = d_b = 20$ nm). As one can observe, the critical threshold is modified in a way that the percolating condition is not favoured by the growing of the Sr$_2$RuO$_4$ inclusions along the c-axis direction.

References

[1] Mackenzie A P and Maeno Y 2003 Rev. Mod. Phys. 75 657
[2] Perry R S et al. 2001 Phys. Rev. Lett. 86 2661
[3] Fittipaldi R et al. 2004 J. Cryst. Growth 282 152
[4] Fittipaldi R et al. 2008 Europhysics Letters 83 27007
[5] Kittaka S et al. 2008 Phys. Rev. B 77 214511