Layered strontium ruthenates are currently a subject of intense investigation due to many fascinating properties exhibited by Sr$_2$RuO$_4$ in the context of spin-triplet superconductivity and by Sr$_3$Ru$_2$O$_7$ in relation with metamagnetic quantum criticality [1–5]. The recent progress [6] in preparing samples with eutectic solidification of superconducting (SC) Sr$_2$RuO$_4$ and metamagnetic Sr$_3$Ru$_2$O$_7$ have opened a novel route to explore systems where unconventional superconductivity and collective magnetic behavior can be matched at the nanoscale with high interface quality. A preliminary characterization of this type of compound, via current-voltage (I–V) measurements, has shown an unusual temperature and field behavior of the critical current [7]. It emerged a picture of a strongly coupled Josephson weak-link network where the Sr$_2$RuO$_4$ grains should be responsible for the SC behavior mediated by an unusual proximity effect through the Sr$_3$Ru$_2$O$_7$ domains. The screening fraction has been estimated via susceptibility measurements to be nearly 100% of the volume of Sr$_3$Ru$_2$O$_7$ region [8]. The peculiar sensitivity of the screening to small applied fields has been considered as an evidence of an intrinsic granularity. So far, the nature of the SC state in the eutectic turns out to be controversial, mainly for the lack of detailed structural characterization about the concentration and the size of Sr$_2$RuO$_4$ inclusions within Sr$_3$Ru$_2$O$_7$ eutectic.

In this letter, we perform a detailed transport and structural analysis of the Sr$_2$RuO$_4$–Sr$_3$Ru$_2$O$_7$ compound profile of a special sample selection within the eutectic itself. We have investigated: i) one nominal Sr$_2$RuO$_4$–Sr$_3$Ru$_2$O$_7$ interface and ii) a macrodomain of Sr$_3$Ru$_2$O$_7$. The I–V characteristics measured in i) shows a supercurrent $I_s$ even at distances of $\sim$2000 $\mu$m far from the interface between Sr$_2$RuO$_4$ and Sr$_3$Ru$_2$O$_7$. $I_s$ does not decay exponentially as in a conventional proximity effect. The same measurements performed for the case ii) give evidence of a supercurrent flowing through the Sr$_3$Ru$_2$O$_7$ domain as well. Systematic compositional and microstructural studies yield limits on the distribution and size of Sr$_2$RuO$_4$ inclusions within the Sr$_3$Ru$_2$O$_7$ matrix. Their presence cannot be more than a few percent of the total volume and they consist of one, two or three individual layers of Sr$_2$RuO$_4$ in a Sr$_3$Ru$_2$O$_7$ matrix interrupting the stack of the bilayered structure along the c-axis. We speculate that the small amount of Sr$_2$RuO$_4$ within the Sr$_3$Ru$_2$O$_7$ cannot

**Superconductivity in Sr$_2$RuO$_4$–Sr$_3$Ru$_2$O$_7$ eutectic crystals**

R. Fittipaldi$^{1,2}$, A. Vecchione$^{1,2}$, R. Ciancio$^{1,2,5}$, S. Pace$^{1,2}$, M. Cuoco$^{1,2}$, D. Stornaiuolo$^{3}$, D. Born$^{3}$, F. Tafuri$^{3,4}$, E. Olsson$^{5}$, S. Kittaka$^{6}$, H. Yaguchi$^{6}$ and Y. Maeno$^{6}$

1 CNR-INFM, Laboratorio Regionale SuperMat - Baronissi (SA), Italy, EU
2 Dipartimento di Fisica “E. R. Caianiello”, Università di Salerno - Baronissi (SA), Italy, EU
3 Dipartimento Ingegneria dell’Informazione, Seconda Università di Napoli - Avesssa (CE), Italy, EU
4 Dipartimento di Fisica “E. R. Caianiello”, Università di Salerno - Baronissi (SA), Italy, EU
5 Department of Applied Physics, Chalmers University of Technology, University of Chalmers - Goteborg, Sweden, EU
6 Department of Physics, Kyoto University - Kyoto 606-8502, Japan

received 17 March 2008; accepted in final form 4 June 2008
published online 3 July 2008

**PACS**

61.05.C- – Ruthenates
61.74.Sv – Critical currents
61.74.Pq – Ruthenates

**Abstract** – Superconducting behavior has been observed in the Sr$_2$RuO$_4$–Sr$_3$Ru$_2$O$_7$ eutectic system as grown by the flux-feeding floating-zone technique. A supercurrent flows across a single interface between Sr$_2$RuO$_4$ and Sr$_3$Ru$_2$O$_7$ areas at distances that are far beyond those expected in a conventional proximity effect. The current-voltage characteristics within the Sr$_3$Ru$_2$O$_7$ macrodomain, as extracted from the eutectic, exhibit signatures of superconductivity in the bilayered Sr$_3$Ru$_2$O$_7$ region. Detailed microstructural, morphological and compositional analyses address issues on the concentration and the size of Sr$_2$RuO$_4$ inclusions within the Sr$_3$Ru$_2$O$_7$ matrix. We speculate on the possibility of inhomogeneous superconductivity in the eutectic Sr$_3$Ru$_2$O$_7$ and exotic pairing induced by the Sr$_2$RuO$_4$ inclusions.

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account for the observed superflow at the interface and within the Sr₃Ru₂O₇ even as a percolative phenomena.

The synthesis of Sr₃RuO₄-Sr₃Ru₂O₇ eutectic crystals by a flux-feeding floating-zone technique has been previously reported [6]. Inspection with the polarized light optical microscope (PLOM) within the a-c planes confirmed the expected lamellar solidification microstructure of the eutectic Sr₂RuO₄-Sr₃Ru₂O₇ [6]. The morphology of the crystals has been investigated by a scanning electron microscope (SEM) equipped with electron back scattered (EBS) detector, yielding no detectable precipitates. Moreover, like other single-phase ruthenates, the eutectic crystals can be cleaved in the a-b planes. The crystal selected for this work (hereafter called sample 1) contains both lamellar pattern and large domains of the two phases. From this sample, we have cut a sample (sample 2) consisting of two macrodomains of Sr₂RuO₄ and Sr₃Ru₂O₇ in contact with each other. This is a crucial aspect for the present investigation: the requirement is to obtain a controlled arrangement of the two phases inside the sample. Indeed, to make sure that just one interface is involved in the transport measurements, both the top and bottom sides of the a-c surface have been polished until the same pattern of Sr₂RuO₄ and Sr₃Ru₂O₇ regions is achieved. The final dimensions of the sample 2 were 5.61 × 0.32 × 0.66 mm³. The composition analyses of the sample 2 have been carried out both by energy dispersive spectroscopy (EDS) and by wavelength dispersive spectroscopy (WDS). EDS analysis performed over different areas of sample 3 away from Sr₃Ru₂O₇ showed few isolated Sr₂RuO₄ clusters visible in SEM on the Sr₂RuO₄ crystal (n=1.333). The deviation from the ideal value in the direction of the monolayer stoichiometry n=1 can be interpreted as a consequence of Sr₂RuO₄ defects.

Spatially resolved WDS presented no signs of gradient concentration of Sr₂RuO₄ or Sr₃RuO₄ approaching the interface. Scans of approximately 10 hours have been carried out in the wavelengths range 0.248–2.379 nm on different regions of the sample to check the presence of spurious elements with Z ranging from 5 (B) to 83 (Br). All the observed peaks have been attributed to X-rays characteristic of Sr or Ru. No magnetically active element has been found. The analysis on the Sr₂RuO₄ and Sr₃Ru₂O₇ domains did not reveal spurious elements within the sensitivity of the experiment. Subsequently, from sample 2 we selected a piece of dimensions 2.82 × 0.32 × 0.66 mm³ nominally consisting only of the Sr₂RuO₄ phase (which we call sample 3). This sample showed few isolated Sr₂RuO₄ clusters of ∼10 μm² on the surface with an average distance among them of ∼30 μm. The amount of Sr₂RuO₄ clusters visible in SEM on the surface of sample 3 has been estimated to be about 0.5% of the Sr₃Ru₂O₇ domain.

To detect the presence of Sr₃RuO₄ nanodomains finely dispersed within the Sr₃Ru₂O₇ layers at an atomic level, we have performed a microstructural analysis by means of transmission electron microscope (TEM) through different areas of sample 3. A representative TEM image of different areas of sample 3 away from Sr₂RuO₄ clusters is shown in fig. 1. It is possible to notice that the Sr₃Ru₂O₇ domain, as extracted from the eutectic, is homogeneous and free of defects over areas of several nanometers. A detailed inspection reveals the presence of one or two unit cells corresponding to the Sr₂RuO₄ structure, occurring as stacking faults monolayers that, along the c-axis, interrupt the Sr₃Ru₂O₇ periodicity. The systematic scan of a large area within sample 3 indicates that such type of stacking faults defects are of nanometric size and their amount can be estimated to be not greater than a few percent of the total area analyzed.

As a confirmation of such dispersion of non-stoichiometric defects with respect to the Sr₃Ru₂O₇ structure, we traced a grid of 20 × 20 points on sample 3 to perform EDS measurements by avoiding the few regions where the clusters of Sr₂RuO₄ are observable. The value obtained for the interpreted Ru-Sr ratio is n = 1.304 ± 0.012, that turns out to be very close to the expected one for an ideal Sr₃Ru₂O₇ crystal (n = 1.333). The deviation from the ideal value in the direction of the monolayer stoichiometry n=1 can be interpreted as a consequence of Sr₂RuO₄ defects.

The structure and crystalline quality of the samples were assessed by a high-resolution triple-axis X-ray diffractometer with a Cu Kα source equipped with a four-circle cradle. The diffraction peaks of sample 1 were identified with the expected (00l) Bragg reflections coming from Sr₃Ru₂O₇ and Sr₂RuO₄, demonstrating both the absence of any spurious phase and the common direction of the c-axes of the two phases. Pole figures proved that the in-plane axes of the two phases were also aligned. It is worth pointing out that XRD spectrum from sample 3 collected with a step size of 0.0002 degrees and time per step of

![Fig. 1: TEM image in the a-c plane of the Sr₃Ru₂O₇ domain as cut from the eutectic. The rectangle indicates the area with the atomic stacking of the Sr₂RuO₄ phase.](image-url)
150 seconds, showed the absence of any Sr₂RuO₄ peak indicating that in the interacting volume, the Sr₂RuO₄ percentage is lower than that one detectable from the surface of the sample.

Hence, by combining the TEM, the X-ray and the EDS/WDS analyses, we may conclude that, the size of Sr₂RuO₄ inclusions dispersed in sample 3, as one, two or three individual layers in a Sr₃Ru₂O₇ matrix, is of the order of a few nanometers in thickness. The upper limit of the volume fraction of these Sr₂RuO₄ layers is 5%. Such percentage can be obtained by combining the systematic TEM analysis [9] and evaluating the difference between the measured average n with respect to that expected for the ideal Sr₃Ru₂O₇ stoichiometry.

We have studied the \( I-V \) characteristics of samples 2 and 3 at low temperatures by using a ³He refrigerator to cool the system down to 0.3 K. All the transport measurements have been performed in Napoli by a standard four-point technique applying the bias current within a-c planes and the magnetic field along the \( c \)-axis. Contact leads have been placed on the crystals to minimize uncontrollable and complicated patterns of the current flow through the sample. For the transport analysis we used two different measurement configurations. The configuration I (sample 1) has two contacts in the Sr₃Ru₂O₇ region and the other two in the Sr₃Ru₂O₇ one at a distance \( d \approx 200 \mu \text{m} \) from the interface (see top side of fig. 2). In the same configuration, other contacts are placed in the Sr₃Ru₂O₇ region at \( d \approx 2000 \mu \text{m} \) away from the interface. In the configuration II (sample 3) the contacts are placed only on the Sr₃Ru₂O₇ region. All the current and voltage leads were attached using an ultrasound bonder. The electrical leads are Al-Si wires, 50 \( \mu \text{m} \) in diameter, and the contact area is about \( 100 \times 100 \mu \text{m}^2 \). The measurement of the resistance vs. temperature in the configuration I below 2 K yields a SC transition at \( T_{c0} = 1.39 \text{K} \). We do not observe extra features, a second transition at lower temperature, as measured by a.c. susceptibility on the sample, below \( 2 \text{K} \). For completeness we mention that, similarly to what has been observed in the eutectic Ru-Sr₂RuO₄ [10], the \( I-V \) spectra are slightly asymmetric by-inverting the sign of the current. We speculate that the origin of such feature is related to the time reversal symmetry breaking of the Sr₃RuO₄ superconducting state [2].

In order to clarify the role played by the occurrence of an anomalous proximity effect induced by the Sr₂RuO₄ macrodomain connected to the Sr₃Ru₂O₇ part, we have performed transport measurements on sample 3 as presented in fig. 3 (configuration II). From the behaviour of the resistance vs. temperature \( R(T) \) it is possible to extract again a SC transition at \( T_{c0} = 1.39 \text{K} \). To prove that the supercurrent flows on a long length scale and only weakly depends on how far the contacts on the Sr₃Ru₂O₇ part are from the interface with the Sr₂RuO₄ region. For completeness we mention that, similarly to what has been observed in the eutectic Ru-Sr₂RuO₄ [10], the \( I-V \) curves measured at different temperatures and magnetic fields show a behaviour similar to the previous one as shown in fig. 2c and fig. 2d. In this case, the \( I_c \) value is about 3.5 A/cm² at 0.3 K. It proves that the supercurrent flows on a long length scale and only weakly depends on how far the contacts on the Sr₃Ru₂O₇ part are from the interface with the Sr₂RuO₄ region.
is comparable to that of Sr in the normal supports, together with the Sr.

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It is possible to obtain the in-plane and out-of-plane normal coherence length in the clean limit for the SrRuO$_7$ of the order of $\xi^N \sim 40 \, T^{-1}$ K nm and $\xi^N \sim 2.5 \, T^{-1}$ K nm ($\xi^N = h v_F / 2 \pi k FY T$, where $i = a, c$ are the axis directions), respectively. Those estimations for the leaking of Cooper pairs in the normal SrRuO$_7$ are not compatible both qualitatively and quantitatively with the observed SC features. Hence, the scenario i) can work only if some sort of unconventional mechanism allows for an enhancing of the proximity length within the Sr$_2$Ru$_2$O$_7$ domain.

One possibility, related to scenario i), is that the presence of the nanometric Sr$_2$RuO$_4$ inclusions can act as resonant pair scattering centers for the electrons scattered across the layers of the Sr$_3$Ru$_2$O$_7$. In this case, areas of RuO$_2$ monolayers, where highly two-dimensional pairing (as seen from $H_{c2}$ in fig. 3b) would occur in the $d_{xy}$ band, act along the c-direction as local centers for pair exchange with the electrons in the bands of the Sr$_3$Ru$_2$O$_7$ having $(d_{xz}, d_{yz})$ character. The latter mechanism is favored because of the specific electronic structure and symmetry of the $t_2g$ bands [13]. The pairs in the RuO$_2$ monolayer would behave like effective pairing impurity for the electron propagation along the c-axis while the interband scattering allows for pair-exchange between electrons in neighbor layers. It emerges a picture similar to the charge Kondo model [14] and a mechanism for long-range order between pairs in different Sr$_2$RuO$_4$ inclusions analog of the RKKY(Ruderman-Kittel-Kasuya-Yosida) interaction between spins. For the charge Kondo model the effective coupling amongst two pair/pseudospins, mediated by particle-particle excitations, at a distance $d$ is $I(d) = (J^2 \rho_f / 8\pi) / d^3$ where $J$ is the effective local pair exchange and $\rho_f$ is the density of states at the Fermi level. Assuming a concentration $x$ of randomly placed Sr$_2$RuO$_4$ inclusions (resonant pairing centers) the mean-field estimation of the SC percolation temperature gives $T_{c-cut} \equiv x J^2 \rho_f \log(D/(x J^2 \rho_f))$ in terms of the bandwidth $D$ [15]. Thus, given for the Sr$_3$Ru$_2$O$_7$ the estimated $\rho_f(\sim 5 \, \text{states/eV})$, $D(\sim 2 \, \text{meV})$ [12] and considering that the rate of interlayer pair scattering $J$ is larger than $D (J/D$ ranges in the interval 4–8) one obtains a value of $T_{c-cut} \sim 1 \, \text{K}$ for a percentage window [2%–8%] of Sr$_2$RuO$_4$ inclusions.

The scenario ii) and iii) invokes the possibility that part of or the entire Sr$_3$Ru$_2$O$_7$ system becomes SC. Such circumstance requires a motivation of the lack of superconductivity in the crystals of Sr$_3$Ru$_2$O$_7$ not grown as eutectic. While iii) is unlikely to occur, it might be possible that the small difference in lattice parameters between the Sr$_2$RuO$_4$ and the Sr$_3$Ru$_2$O$_7$ results in a strain that induces pair formation within the Sr$_3$Ru$_2$O$_7$ domain in the form of inhomogeneous patterns. A more detailed microstructural analysis is required to obtain extra insights in such issues.

In conclusion, we have shown that the Sr$_3$Ru$_2$O$_7$, as extracted from the Sr$_2$RuO$_4$–Sr$_3$Ru$_2$O$_7$ eutectic system, exhibits unusual SC features. The SC state seems to be affine with that of Sr$_2$RuO$_4$ because there occurs no negative interference in the supercurrent flow at the single interface between Sr$_2$RuO$_4$ and Sr$_3$Ru$_2$O$_7$ macrodomains. The microstructural analysis gives a clear indication of the amount and size of Sr$_2$RuO$_4$ or other spurious phases within the Sr$_3$Ru$_2$O$_7$ matrix. Such information excludes a simple percolation via a proximity network of superconducting Sr$_2$RuO$_4$ microdomains to justify the observed supercurrent and points on the role of nanometric RuO$_2$
stacking faults monolayer in assisting (driving) the pair leaking (formation).

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We would like to acknowledge valuable discussions with C. Noce.

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