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μSR studies of superconductivity in eutectically grown mixed ruthenates

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The low-temperature magnetic behavior of the double-layered ruthenate $Sr_3Ru_2O_7$, as grown from a eutectic Sr_2RuO_4-$Sr_3Ru_2O_7$ system, was investigated via zero- and transverse-field muon-spin rotation. The gradual increase of the muon relaxation rate observed below 2.5 K, even in the absence of applied magnetic fields, indicates the occurrence of a spontaneous breaking of time-reversal symmetry. The onset of the latter at a temperature above 1.5 K, the T_c of the single phase Sr_2RuO_4, provides evidence about an unconventional superconducting state in the eutectic phase, which most likely takes place at the interface between the Sr_2RuO_4 and $Sr_3Ru_2O_7$ domains, or even inside the $Sr_3Ru_2O_7$ phase. We show that the superconducting state manifests a two-component behavior in the transverse-field response with change-over at about $T = 2.5$ K and $T = 1.5$ K. The comparison with zero-field μSR data in the Ru-Sr_2RuO_4 eutectic system rules out the possibility of spurious effects due to embedded Ru islands.

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I. INTRODUCTION

Strontium ruthenates of the so-called Ruddlesden-Popper series, described by the formula $Sr_{n+1}Ru_nO_{3n+1}$, display a remarkable variety of unconventional properties. Sr_2RuO_4, the $n = 1$ member,[1] is the only copper-free metal oxide with a layered perovskite crystal structure to become a superconductor and it represents one of the rare examples of odd-parity spin-triplet pairing (below $T_c = 1.5$ K). The $n = 2$ member, $Sr_3Ru_2O_7$, is an enhanced Pauli paramagnet[2] close to magnetic order, which undergoes a low-temperature anisotropic metamagnetic (i.e., field-induced) transition, exhibiting unconventional properties with possible emergent nematic states and quantum criticality.[3,4] Finally, the $n = 3$ member, $Sr_4Ru_3O_{10}$, shows ferro- or metamagnetic behavior depending on the direction of the applied magnetic field,[5–7] and only $SrRuO_3$ ($n = \infty$) is an isotropic ferromagnetic metal with $T_c = 160$ K.[8]

Several studies have pointed out the crucial importance of crystal purity in ruthenate oxides. In fact, both superconductivity in Sr_2RuO_4 and the quantum critical behavior in $Sr_3Ru_2O_7$ emerge only in samples with residual resistivity, ρ_{res}, lower than 1.0 $\mu\Omega$ cm.[9] Extremely pure single crystals of $Sr_3Ru_2O_7$ with ρ_{res} as low as 0.4 $\mu\Omega$ cm have enabled the observation of quantum oscillations in the resistivity, both above and below the critical metamagnetic field.[10]

By now there is an ample experimental evidence concerning the unconventional, non-s-wave superconductivity in Sr_2RuO_4. Transport and resonance studies of Sr_2RuO_4 could not detect a Hebel-Slichter coherence peak in this material,[11] hence offering strong evidence about the non-s-wave nature of its superconductivity. The use of artificial Pb-Sr_2RuO_4-Pb junctions for phase-sensitive studies showed an unexpected drop in the temperature dependence of the critical current,[12] again suggesting that Sr_2RuO_4 is an unconventional superconductor, different from the standard spin-singlet Pb. Evidence about non-s-wave superconductivity has been also provided by small-angle neutron scattering data, which reveal a square rather than a triangular vortex lattice,[13] or by tunneling measurements, showing the existence of Andreev surface-bound states.[14,15]

The hallmark of Sr_2RuO_4 superconductivity is the presence of time-reversal symmetry breaking (TRSB), a phenomenon most likely arising from the chiral p-wave structure of the Cooper pairs. The first experimental evidence for such a configuration in Sr_2RuO_4 came from the observation of a spontaneous magnetic field in muon-spin rotation measurements.[16] Later on, a nonzero Kerr rotation below T_c in high-resolution polar Kerr effect measurements[17] and a nonsymmetric quantum interference pattern in the in-plane Josephson junctions of Pb-Sr_2RuO_4,[18] both confirmed the intrinsic breakdown of the time-reversal symmetry within the superconducting state. Still, surface currents associated with the proposed TRSB paired state have been so far elusive.[19] Recently, the possibility of synthesizing eutectic (i.e., mixed) systems, representing naturally occurring nanoscopic interfaces, has opened new routes for testing the symmetry of the pairing wave function in the proximity of normal or magnetic systems, as well as in searching for novel quantum configurations that could emerge at the interface between the embedded phases. The main drive behind eutectic growth is given by the possibility of developing composite materials with distinct properties

from those of the pure constituents. The first attempt in this direction was represented by the (somehow accidental) fabrication of a eutectic phase where single-crystalline islands of pure Ru metal were embedded in a single-crystal matrix of Sr_2RuO_4.[15,20–22] Surprisingly, the onset of superconductivity in the newly grown eutectic system was found to occur at 3 K (i.e., at twice the original T_c). This increase in T_c was justified as being due to interface states between the Ru metallic islands and the host Sr_2RuO_4 domain, yet the lack of the expected proximity behavior seemed to suggest that the domains hosting the 3-K superconducting state were away from the Ru-Sr_2RuO_4 interface.[23]

Lately, the search for different types of eutectic systems with atomically sharp interfaces, including the Sr_2RuO_4 as a component, has led to the synthesis of Sr_2RuO_4-$Sr_3Ru_2O_7$,[24–26] a material where the stacking of the two phases occurs along their common c axis. Investigations using different techniques have shown that this system exhibits an unusual behavior both in the magnetic and in the transport response.[24–26] Current-voltage measurements have detected critical currents comparable to those found in the pure $n = 1$ member, but with an unusual temperature and field dependence.[26] This result is consistent with a network of coupled Josephson weak links, where the Sr_2RuO_4 grains are responsible for the superconducting behavior, but still require some kind of long-range proximity via the $Sr_3Ru_2O_7$ domains to establish bulk superconductivity. Combined transmission electron microscopy,[27] x-ray diffraction, and compositional analysis, show a Sr_2RuO_4 intergrowth volume fraction in the main matrix of $Sr_3Ru_2O_7$ of only 5%, seemingly confirming this scenario.[28] However, unexpectedly, susceptibility measurements on $Sr_3Ru_2O_7$ regions cut from eutectically grown Sr_2RuO_4-$Sr_3Ru_2O_7$ crystals, show a nearly 100% of superconducting screening fraction.[25] Thus, the origin and the nature of the superconducting state in the eutectic Sr_2RuO_4-$Sr_3Ru_2O_7$ crystals and in the $Sr_3Ru_2O_7$ regions cut from it (hereafter being referred to as eutectic $Sr_3Ru_2O_7$) remain open.

On a general ground, the issue concerning the nature of the superconducting state in the eutectic system is of fundamental interest, both for achieving a deeper insight into the mechanisms responsible for the enhancement of the critical temperature, as well as for the evolution of the TRSB states, including their modification in the presence of different types of interfaces. In particular, the possibility of a transition from a nonchiral (no TRSB) to a chiral superconducting state, as might occur in the vicinity of a Ru-Sr_2RuO_4 interface,[22,29] requires specific probes with a high sensitivity to the local magnetic environment. To this end, muon-spin spectroscopy (μSR) represents the preferred technique[30] for unraveling the complex superconducting behavior of the eutectically grown $Sr_3Ru_2O_7$. Besides the investigation of the superconducting total fraction and of the penetration depth at low temperature in the $Sr_3Ru_2O_7$ domains cut from eutectic crystals, muon-spin rotation analysis has been used to detect the possibility of superconducting states with time-reversal symmetry breaking as compared to the Sr_2RuO_4 case. Indeed, as already shown in the $n = 1$ case,[16] spontaneous currents could show up as a sizable muon-spin relaxation, even in a zero applied magnetic field. The peculiar structure of the eutectic samples makes them particularly suitable for studying the effect of Sr_2RuO_4-$Sr_3Ru_2O_7$ interfaces on the chiral states of the triplet superconductor Sr_2RuO_4, also via a comparison with the pure Sr_2RuO_4 single crystal and, possibly, with the Ru-Sr_2RuO_4 system.

In this work we show that in the eutectically grown $Sr_3Ru_2O_7$ the time-reversal symmetry breaking due to the superconducting state occurs already at 2.5 K (i.e., well above 1.5 K, the critical temperature of the Sr_2RuO_4 single phase). The related relaxation rate varies smoothly with temperature (i.e., with no change in slope) even when passing through 1.5 K. The evolution with the magnetic field of the superconductivity reveals the latter state to be weak and not uniform, while the two-component SC behavior observed in a transverse field hints at a different nature for the superconducting state that nucleates below $T = 2.5$ K and $T = 1.5$ K, respectively.

The current paper is organized as follows: In Sec. II we present the experimental details concerning the growth and characterization of the eutectic samples. In Sec. III we deal mostly with the analysis of the muon-spin rotation/relaxation measurements. Finally, the conclusions are reported in Sec. IV.

II. EXPERIMENTAL DETAILS

High-quality, two-phase crystals of Sr_2RuO_4-$Sr_3Ru_2O_7$ with a residual resistivity lower than 1.0 $\mu\Omega$ cm were grown at the Department of Physics of Kyoto University (Japan) and at the research center CNR-SPIN Salerno (Italy). By using the recently developed flux-feeding floating-zone technique[31] and by changing the Sr:Ru ratio in the starting material, we could obtain crystals with a different amount of Sr_2RuO_4 and $Sr_3Ru_2O_7$.[24] The eutectic compounds were carefully characterized via high-resolution x-ray diffraction and via scanning electron microscopy (SEM), as described in detail elsewhere.[24,32] Additionally, the microstructure of the eutectic samples was checked by electron backscattering diffraction carried out inside the SEM chamber. All investigations show that the two phases, Sr_2RuO_4 and $Sr_3Ru_2O_7$, stack orderly along the shared c axis. For the present work, crystals were cut from eutectic compounds, consisting mainly of $Sr_3Ru_2O_7$ with small regions of Sr_2RuO_4. These were then arranged to form a $14 \times 10 \times 1.5$-mm^3 mosaic configuration.

The selected $Sr_3Ru_2O_7$ crystal fragments employed in the successive μSR experiments were checked once more using the above-mentioned techniques. In particular, their chemical composition was determined via energy- (EDS) and wavelength- (WDS) dispersive spectroscopy which, most importantly, could exclude the presence of any magnetically active elements (within a detection sensitivity of the order of 50 ppm). Figure 1, depicting the x-ray spectrum of a typical sample, shows that almost all of the specimen is in the $Sr_3Ru_2O_7$ phase. As a comparative test and to exclude possible surface effects due to the limited x-ray penetration depth, some of the single crystals used in the measurements were crushed into powders. Rietveld refinement of the powder diffraction pattern did not show appreciable changes in the relative percentage of the two coexisting phases as compared to the single-crystal case.

The joint use of structural, morphological, and compositional data allowed us to estimate a volume fraction of about 5% for the minority Sr_2RuO_4 inclusions in the investigated

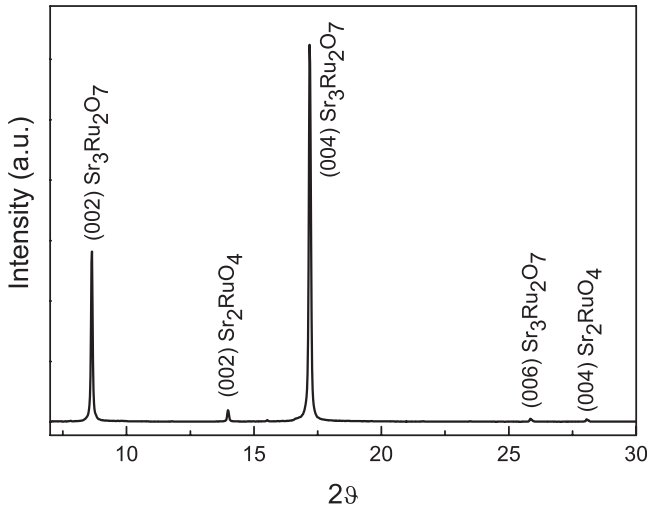

FIG. 1. Typical x-ray diffraction pattern from the cleaved (001) surface of a $Sr_3Ru_2O_7$ crystal, cut from the eutectic system grown via the flux-feeding floating-zone method. A major amount of the $Sr_3Ru_2O_7$ phase is visible.

$Sr_3Ru_2O_7$ crystals. The small amount of Sr_2RuO_4 is consistent also with results from specific heat measurements performed on samples from the same batch as the one employed in the present work.[25,33] Finally, by using a standard four-probe technique and a Heliox ^{3}He refrigerator system, the sample's resistivity could be measured down to 0.3 K.

Zero- (ZF) and transverse-field (TF) μSR measurements were carried out at the GPS and LTF spectrometers of Paul Scherrer Institut, Villigen (Switzerland). The combined use of two instruments allowed us to cover the temperature range from 10 K down to 20 mK. The above-mentioned mosaic of the c-axis oriented eutectic $Sr_3Ru_2O_7$ was glued on top of a silver plate. Due to the large sample thickness (in excess of 1 mm) most of the incoming muons were implanted in the sample, with only an irrelevant fraction missing it and stopping in the Ag substrate (where they produce a temperature-independent and practically nonrelaxing μSR signal). In these conditions, the maximum asymmetry signal from the sample was $A_0 \simeq 0.25$. Since the sought for muon relaxation decay was expected to be rather small, a time window of 10 μs and statistics of the order of 30×10^6 events were needed to unambiguously determine the relaxation value. During the ZF measurements, possible stray magnetic fields at the sample position were dynamically compensated to zero to better than 0.01 mT.

We recall that ZF-μSR represents a unique possibility offered by muon spectroscopy, whereby the precession of the spin-polarized muon ensemble is determined exclusively by the sample's internal magnetic fields. Given the rather large muon gyromagnetic ratio, $\gamma_\mu/2\pi = 135.5$ MHz/T, and a lifetime of $\tau_\mu = 2.2$ μs, fields as low as 0.05–0.1 mT[34,35] can be easily detected. The ZF time-domain data were best fitted using an exponentially decaying function (i.e., Lorentzian in frequency), given by

$$A(t) = A_0 \exp(-\Lambda t) + b_0. \quad (1)$$

Here Λ represents the spin relaxation rate, while b_0 denotes the amplitude of the nonrelaxing background signal.

Similarly to ZF, transverse-field muon-spin rotation (TF-μSR) was employed to measure the internal magnetic field distribution in the mixed superconducting state. In the latter case the magnetic field distribution is not uniform both because of the field penetration as well as due to inhomogeneities arising from the eutectic structure of the crystal. Due to the randomness of the muon implantation process (with respect to the length scale of flux distribution), a magnetic field applied perpendicular to the muon spins would set them in a precessing motion, hence randomly sampling the magnetic field distribution. Therefore, a knowledge of the time evolution of the muon-spin polarization $P(t)$ provides a direct access to the distribution of the local magnetic fields $p(B)$, in turn related to the superconducting penetration depth λ.

III. RESULTS AND DISCUSSION

Transport measurements were carried out both along the c axis and in the ab plane, with resistivities of the order of 0.2 mΩ cm and 0.5 $\mu\Omega$ cm found in the two configurations, respectively.[38] These values confirm the high quality of the $Sr_3Ru_2O_7$ crystals grown from eutectic solidification, with the structural quality being assessed by transmission electron microscopy.[39] More importantly, all the $Sr_3Ru_2O_7$ eutectic crystals employed in the present work show a 30% drop in the c-axis resistance at an onset temperature of 2.5 K, followed by a sharp drop to 5% of the normal state value between 1.5 and 1.3 K (see Fig. 2). Since the onset temperature is higher than that of pure Sr_2RuO_4 ($T_c = 1.5$ K), this suggests that other superconducting states and mechanisms might come into play in the eutectic compound.

In fact, a possible explanation of the superconductivity as being due to a conventional proximity network finds serious difficulties in an almost pure eutectic $Sr_3Ru_2O_7$ crystal. Taking into account the estimated 5% level of Sr_2RuO_4 inclusions, our system is expected to be in a hypothetical percolation regime

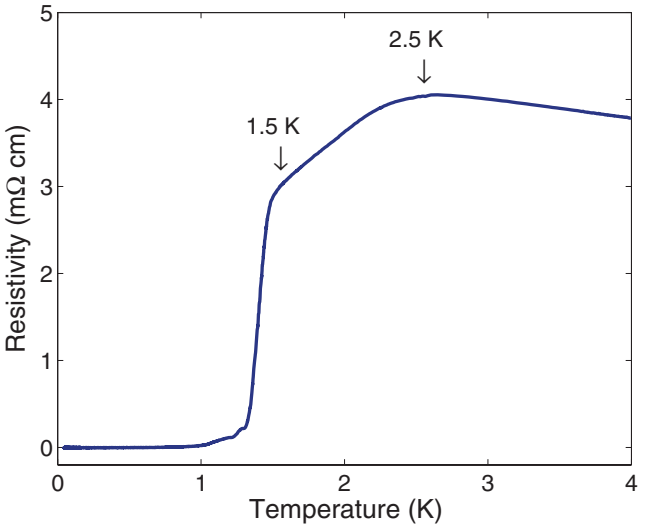

FIG. 2. (Color online) Low-temperature, c-axis resistance of eutectically grown $Sr_3Ru_2O_7$ crystals showing clear resistance drops at 1.5 and 2.5 K.

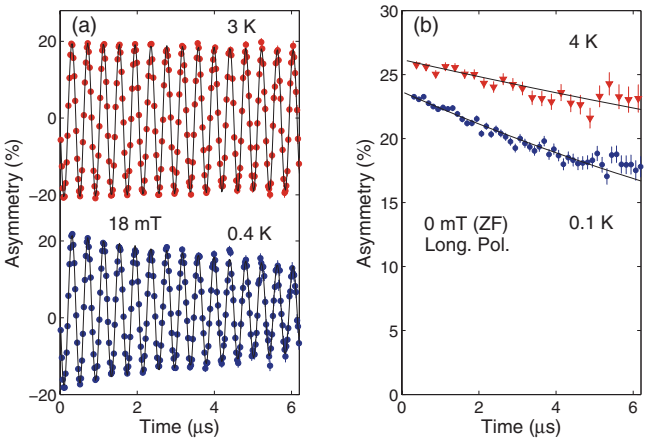

FIG. 3. (Color online) Time-domain μSR data in eutectically grown $Sr_3Ru_2O_7$ for different temperatures at a transverse applied field $\mu_0 H = 18$ mT (a), and without applied fields (b). In both cases, the damping rate of muon asymmetry increases as the temperature decreases. Continuous lines in panel (b) refer to fits using Eq. (1). Data for $T = 0.1$ K in panel (b) shifted by 7% for clarity.

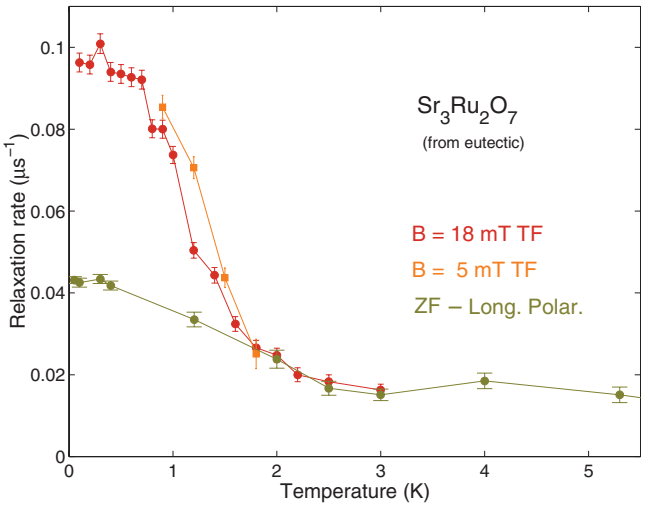

FIG. 4. (Color online) Zero- and transverse-field μSR Lorentzian relaxation rates vs temperature in eutectically grown $Sr_3Ru_2O_7$ at an applied field of 0, 5, and 18 mT, respectively. The low-temperature increase of ZF damping rate indicates the onset of spontaneous magnetic moments.

only for $T \lesssim 0.25$ K,[36] a 10-fold lower temperature than the measured one, 2.5 K.

To shed light on such an intriguing feature of the $Sr_3Ru_2O_7$ crystal cut from the eutectic and to explore the nature of its superconducting behavior, a series of muon-spin rotation/relaxation (μSR) measurements were performed. Figure 3 reports the time-domain μSR data, both in the TF and in the ZF case. In TF-μSR measurements performed above T_c one expects a very small decay of muon-spin polarization, arising from the randomly oriented nuclear moments lying close to a muon stopping site. Below T_c, instead, due to a significant field distribution related to the presence of a vortex lattice, one should detect a considerable decay of polarization. Indeed, as shown in Fig. 3(a), this is exactly our case: The small relaxation rate of 0.016(1) μs^{-1} observed at 3 K, shows an almost fivefold increase at the lowest temperatures.

Similar effects, though much smaller in magnitude, are found also in the ZF case [see Fig. 3(b)]. Their nature, however, differs qualitatively from that of the TF case. In general, when a conventional superconductor is cooled in the absence of magnetic fields, no magnetic response is expected (i.e., there is no creation of a flux-line lattice). The spontaneous appearance of an internal magnetic field below the transition temperature is a clear indication that the superconducting state is characterized by the breaking of the time-reversal symmetry and has been associated with p-wave (odd-parity) superconductivity, as observed in Sr_2RuO_4.[16,45] This is in perfect agreement with the data shown in Fig. 3(b), where the low-temperature muon decay rate [0.043(1) μs^{-1}] is almost 2.5 times higher than the value found above T_c. This key result hints at the presence of *spontaneous* internal fields, arising below the superconducting transition temperature.

Such behavior can have different origins. On one hand, similarly to the phenomenology observed in Ru-Sr_2RuO_4, a novel superconducting state can occur at the interface between the $Sr_3Ru_2O_7$ matrix and the Sr_2RuO_4 inclusions, where due to a c-axis mismatch the reconstruction of the electronic structure and possible modifications of the coupling between the electrons and the bosonic modes responsible for the pairing can lead to an increase of the critical temperature. On the other hand, the superconducting state with an increased T_c might also nucleate inside a $Sr_3Ru_2O_7$ macrodomain in view of the fact that such a system has a higher degree of purity if compared to the $Sr_3Ru_2O_7$ sample not obtained from a eutectic crystal.

The muon polarization decay rates for the ZF- and the TF-μSR case, collected in a temperature range 100 mK to 5 K at different applied fields, are shown in Fig. 4. As already discussed, above $T_c = 2.5$ K all the reported curves share the same baseline, attributed to the presence of nuclear moments. As the temperature is lowered below T_c, there is a clear increase in the relaxation rate, quite significant in the TF case, but clearly noticeable (i.e., much greater than the calculated uncertainty) also in the ZF case. Notice that the TRSB spontaneous fields are most probably quenched by the application of an external magnetic field; even if they were not, their contribution to the TF relaxation would be only half the ZF rate.[46] Therefore in the interval 1.5–2.5 K, although the ZF relaxation rate apparently equals the TF rate, there must be a net superconducting vortex contribution to the latter.

As a further confirmation of the above surprising result, comparative ZF-μSR measurements were carried out also in the pure Sr_2RuO_4, as well as in the eutectically grown Sr_2RuO_4-Ru metal system, with the respective results being shown in Fig. 5. Here too, the normal-state data show very similar, temperature- and sample-independent relaxation rates (of the order of 0.015 μs^{-1}). Things change dramatically once the temperature is lowered: Our eutectically grown $Sr_3Ru_2O_7$ system shows an evident upturn of relaxation already below 2.5 K, while the relaxation of both the Sr_2RuO_4 and the Sr_2RuO_4-Ru metal eutectic system does not change or it has only a weak variation, respectively, until the critical temperature ($T_c = 1.5$ K) associated to the bulk Sr_2RuO_4 is reached.

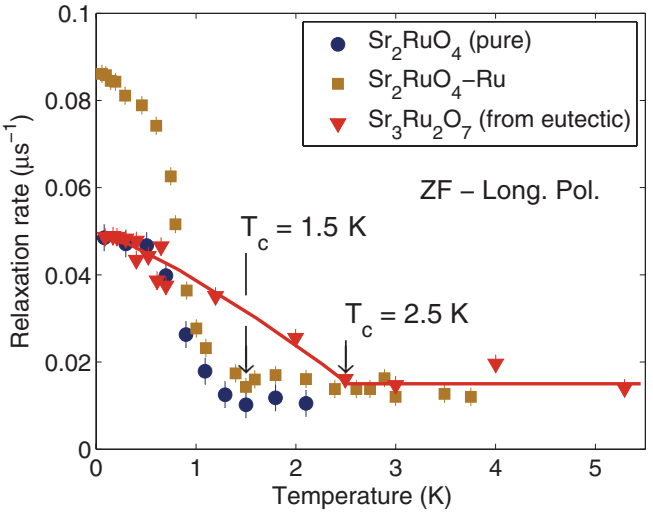

FIG. 5. (Color online) Comparison of zero-field μSR relaxation rates vs temperature in eutectically grown $Sr_3Ru_2O_7$ with those of pure Sr_2RuO_4 and Sr_2RuO_4-Ru system. While the latter two show an upturn of relaxation below $T = 1.5$ K, the relaxation of $Sr_3Ru_2O_7$ starts to increase already at $T = 2.5$ K (the red line is a guide to the eye).

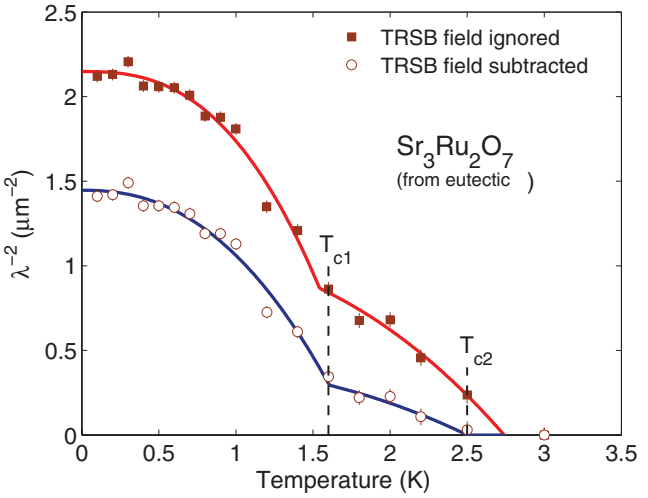

FIG. 6. (Color online) Temperature dependence of λ^{-2} for the eutectically grown $Sr_3Ru_2O_7$, reconstructed from $\sigma_{sc}(T)$ measured at $\mu_0 H = 18$ mT by ignoring (solid squares) or by subtracting (open circles) a possible TRSB-field contribution. The line represents a power-law fit with two components (see text).

The observed increase in the ZF and TF muon-spin relaxation rates could in principle be justified by spin fluctuations in the narrowing limit, $\Lambda \approx \gamma_\mu^2 B_\mu^2 \tau_c$. However, assuming as an upper limit for the instantaneous internal field the static value measured in $SrRuO_3$, $B_\mu \approx 0.25$ T,[37] our measured rates would imply very long fluctuation times, $\tau_c \approx 10^{-12}$ s, that are justified only close to a transition or freezing toward static order and would eventually lead to the detection of a static B_μ at very low temperature, contrary to our observations down to 20 mK. Furthermore, Fig. 2 shows that resistivity decreases below 2.5 K, whereas any spin-scattering mechanism would normally result in a resistivity increase. We must therefore discard the spin fluctuation hypothesis.

Thus superconductivity and TRSB fields must take place already between 1.5 and 2.5 K. This confirms beyond any reasonable doubt that superconductivity in the eutectically grown $Sr_3Ru_2O_7$ cannot be ascribed to possible percolative phenomena due to the presence of Sr_2RuO_4, but it requires a new conceptual framework to be explained.

From the muon-spin relaxation rate $\sigma_{sc}(T)$ measured at $\mu_0 H = 18$ mT one can obtain the temperature dependence of λ^{-2}, as shown in Fig. 6. The first thing to note is that if superconductivity were limited to the marginal Sr_2RuO_4 fraction, 5% in volume, and to its immediate surroundings by conventional proximity, the muon asymmetry would show two components, a large unrelaxed one and a small strongly relaxed one, indeed hardly measurable. The single average decay in the lower part of Fig. 3(a) indicates instead that the volume is entirely occupied by a locally inhomogenous flux distribution, as it results from a flux-line lattice, although probably far from ideal.

We fit the TF-μSR data by using a Gaussian model,

$$A(t) = [A_0 \exp(-\sigma^2 t^2/2) + b_0] \cos(\gamma_\mu B t + \phi), \qquad (2)$$

which, although not perfect (its goodness of fit is only marginally worse than that of a Lorentzian model), empirically provides a commonly accepted measure for the internal field distribution. Here B is the magnetic field (practically the same inside and outside the sample) and $\sigma = (\sigma_{nm}^2 + \sigma_{sc}^2 + \Lambda^2/4)^{1/2}$ the relaxation rate due to contributions, respectively, of the nuclear moments, of the nonuniform magnetic-field distribution in the superconducting state, $p(B)$, and of a possible additional term arising from the TRSB fields (should they not be suppressed by the external magnetic field). Figure 6 shows the temperature dependence of λ^{-2}, when this additional correction is ignored, or is taken into account (i.e., subtracted). We recall that a Gaussian model is well suited for the symmetric distribution observed in samples with a certain degree of disorder [as opposed to an asymmetric $p(B)$ distribution, typical of disorder-free single crystals].[40] Successively, by assuming $\sigma_{sc} \propto \lambda^{-2}$, a relation valid in a broad range of circumstances,[41] one can obtain λ^{-2}. A much larger degree of disorder is expected very close to the surface of a superconductor,[42] but most likely this is not the case for our bulk, single-crystal mosaic sample.

Since the λ^{-2} parameter is well known to be proportional to the superfluid density, $\lambda^{-2} \propto n_s/m^*$, the fact that it is different from zero up to 2.5 K clearly implies a superconducting behavior which persists well above 1.5 K. Strictly speaking, an analysis in terms of λ implicitly assumes the presence of an ordered vortex lattice in the superconducting state. A vortex lattice characterized by disorder, quite likely reflecting also our case, is known to produce less reliable numerical λ values. However, the singularities of the flux-lattice field distribution $p(B)$ depend strongly on intrinsic and local features, such as the core size ξ, the penetration depth λ, and the average flux-lattice parameter, but weakly on the long-range correlations of the flux lattice. Hence the inverse proportionality $\sigma \propto \lambda^{-2}$ is preserved even in moderately to strongly disordered lattices and the functional dependence of λ on temperature (Fig. 6) is hardly affected.

The overall $\lambda^{-2}(T)$ dependence in our case was fitted using different conventional models but, unfortunately, none of them could satisfactorily reproduce the observed features. On the contrary, a phenomenological model contemplating *two* different SC contributions seems to be in excellent agreement with data. In the latter case, each superconducting component was fitted to a power-law dependence $\propto [1-(T/T_c)^{\alpha}]$, reminiscent of a two-fluid model of superconductivity. The resulting best fit parameters are $T_{c1} = 1.54(4)$ K, $T_{c2} = 2.74(9)$ K for the critical temperatures and $\alpha_1 = 2.6(4)$, $\alpha_2 = 2.6(9)$ for the power-law coefficients, respectively. With due caution, related to the limited absolute accuracy of λ as discussed above, the fit provides also a zero-temperature penetration depth $\lambda_{ab}(0) = 670(30)$ nm, which is almost four times longer than that found in the pure Sr_2RuO_4 system.[13,16] If the TRSB fields were not suppressed by the transverse field, a qualitatively similar analysis would apply, yielding $\lambda_{ab}(0) = 870(40)$ nm. We recall once more that the above discussion concerns relatively small changes of the μSR relaxation rate with temperature, in turn reflected in a rather large uncertainty on the extracted penetration depth values.

IV. CONCLUSIONS

We have reported on muon-spin depolarization measurements in the eutectically grown $Sr_3Ru_2O_7$. At a temperature $T_c = 2.5$ K we find the onset of a spontaneous time-reversal symmetry breaking associated with the superconducting behavior of the system. The increased temperature of the superconducting state might be related to interface phenomena between the Sr_2RuO_4 and the $Sr_3Ru_2O_7$ subsystems, but it might also be a feature of the $Sr_3Ru_2O_7$ domain away from the $n = 1$ inclusions. The increase of the superconducting onset temperature is not new for the ruthenium-based eutectic systems, as it occurs also in Ru-Sr_2RuO_4 samples. For the eutectically grown $Sr_3Ru_2O_7$ the presence of spontaneous internal fields is observed at a higher temperature than that of the pure Sr_2RuO_4 crystals. Such a circumstance can be justified by the fact that the interfaces in the eutectically grown $Sr_3Ru_2O_7$ mainly break the translational symmetry along the c axis. Therefore they do not strongly affect the nucleation in the Ru-O planes of a TRSB state of the $k_x \pm ik_y$ type or with a structure similar to that of Sr_2RuO_4.

This seems to be different from the case of the induced superconductivity in the eutectic Ru-Sr_2RuO_4 system, where the onset of superconductivity (at ~ 3 K) is accompanied by a weak change in the relaxation rate, which in turn has a significant variation only at lower temperatures as if arising from the Sr_2RuO_4 internal field contribution (see Fig. 5). Here, the interface between metallic Ru islands and Sr_2RuO_4 domains most probably leads to a breakdown of the translational symmetry in the RuO planes, favoring a single-component superconducting configuration rather than planar TRSB states.

As for the mechanism that justifies the increase of the critical temperature in eutectic $Sr_3Ru_2O_7$, it is worth pointing out that a similar enhancement of superconductivity by almost 2.7 K was obtained already in the 80s by Matthias *et al.*[43] in an eutectic system formed by Ir ($T_c \sim 0.10$ K) with a small amount of YIr_2. In the latter case, a small lattice mismatch between Ir and YIr_2 could induce a change in the frequency of the phonons responsible for the Cooper pair formation,[44] thus leading to an extraordinary enhancement in T_c. Similarly, for the case of eutectic $Sr_3Ru_2O_7$ the c-axis mismatch between the $n = 1$ and $n = 2$ phases of the Ruddlesden-Popper series might be the driving force inducing the change in the electronic structure so as to give the observed enhancement of T_c.

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[45]In both Ref. 16 and here we find that the additional relaxation below T_c is best fitted by an exponential function. In both cases, we expect that this function does not indicate a dynamic process but instead represents the particular static field distribution produced by the spontaneous currents, since in Ref. 16 the field distribution was demonstrated to be static.
[46]Relative to the transverse-field case, the initial depolarization is stronger in zero field by a factor of 2 since for the latter geometry the two components of $B_{\rm loc}$ perpendicular to the z axis contribute to the depolarization, whereas only the one component parallel to the field contributes in the TF case (see p. 106 of Ref. 35).