Critical current of the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic system

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We report critical current measurements of the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic system, a solid mixture of the spin-triplet superconductor Sr_2RuO_4 and the nearly ferromagnetic metal $Sr_3Ru_2O_7$. Our results reveal that this eutectic forms an unusual superconducting state with features distinct from those of the pure Sr_2RuO_4 phase. We observe a crossover from Ambegaokar-Baratoff to Ginzburg-Landau type behavior in the temperature dependence of the critical current, similar to the behavior of Josephson weak-link networks in granular high-T_c cuprates. Our results suggest that large regions of $Sr_3Ru_2O_7$ in this solid mixture may become superconducting by an unusual proximity effect.

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The layered perovskite Sr_2RuO_4, the $n=1$ member of the Ruddlesden-Popper series $Sr_{n+1}Ru_nO_{3n+1}$ of strontium ruthenates, has received considerable attention due to its unusual superconducting characteristics.[1,2] The invariance of the NMR Knight shift across the superconducting transition[3,4] and the results of recent phase-sensitive experiments[5] have provided convincing evidence that Sr_2RuO_4 is an odd-parity, spin-triplet superconductor.

A remarkable doubling of the superconducting transition temperature T_c up to 3 K was observed in the Ru-Sr_2RuO_4 eutectic system, which is formed by the addition of excess Ru to the starting materials used to grow Sr_2RuO_4 (Refs. 6 and 7). This so-called 3-K phase consists of lamellar domains of Ru metal embedded in a matrix of single crystalline Sr_2RuO_4. Measurements of the specific heat and upper critical field indicate that the enhanced superconductivity is not a bulk phenomena, originating rather in the interface region between Sr_2RuO_4 and the Ru inclusions.[8] Tunneling studies indicate that the 3-K phase is also unconventional, but with a different pairing symmetry from the bulk.[9] These results agree qualitatively with a phenomenological theory developed by Sigrist and Monien which proposes a line node *p*-wave state in the interface region around the Ru domains.[10]

Recently, a second eutectic solidification system was reported, Sr_2RuO_4-$Sr_3Ru_2O_7$; these samples contain significant volume fractions of both superconducting Sr_2RuO_4 and the nearly ferromagnetic metal $Sr_3Ru_2O_7$ (Refs. 11 and 12). $Sr_3Ru_2O_7$, which contains two layers of corner sharing RuO_6 octahedra, is pseudotetragonal and shares a common *c* axis with Sr_2RuO_4 in the solid mixture.[11,13] Growth conditions for this eutectic, in terms of the composition of the starting materials, are intermediate between those for pure phase Sr_2RuO_4 (Ref. 7) and $Sr_3Ru_2O_7$ (Ref. 14). AC susceptibility measurements on one of these samples revealed a superconducting transition at 1.43 K, followed by a broad diamagnetic tail in χ' and an unusual dissipation χ'' at low temperatures.[11]

Since both the Ru-Sr_2RuO_4 and the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic systems form connected superconducting pathways even in large samples, it is of interest to compare their superconducting states with those of pure Sr_2RuO_4. Our recent critical current measurements on the 3-K phase suggest that the order parameter of the interface regions on different Ru inclusions overlaps substantially, forming an inhomogeneous superconducting Josephson network with features distinctly different from pure Sr_2RuO_4. This Josephson-type network dominates the critical current of the Ru-Sr_2RuO_4 system even below the bulk superconducting transition.[15] Here we report the measurements of the critical current of Sr_2RuO_4-$Sr_3Ru_2O_7$ single crystals grown by a floating zone method. Our results reveal that the Sr_2RuO_4-$Sr_3Ru_2O_7$ system forms a unusual superconducting state with features distinct from those of the pure or 3-K phases. These features suggest that large regions of $Sr_3Ru_2O_7$ in this eutectic may become superconducting by an unusual proximity effect.

Crystals for this experiment were grown via a floating zone method as reported previously by Fittipaldi *et al.*[11] Critical current measurements were performed on batch C6J04, which was prepared with a feed rod containing 45% excess Ru with respect to Sr_2RuO_4. The resulting eutectic crystal contained interspersed lamellae of Sr_2RuO_4 and $Sr_3Ru_2O_7$ that were on the order of tens of microns in size. A rough estimate based on optical microscopy and the relative intensites of low-angle x-ray diffraction peaks indicates that the volume ratio of Sr_2RuO_4 to $Sr_3Ru_2O_7$ is close to 1. A detailed x-ray analysis of this eutectic system[11] revealed that the two phases share a common *c* axis and retain their single phase lattice parameters.

Transport measurements were performed by a standard four-point method with current applied along the *c* axis; the crystal dimensions were $0.40\ \text{mm}\times 0.31\ \text{mm}\times 0.2\ \text{mm}$, with the shortest dimension along the *c* axis. All of the measurements were performed in a ^{3}He cryostat with a base temperature of 0.3 K, equipped with a 9 T superconducting magnet. To minimize the heating effects, the critical current

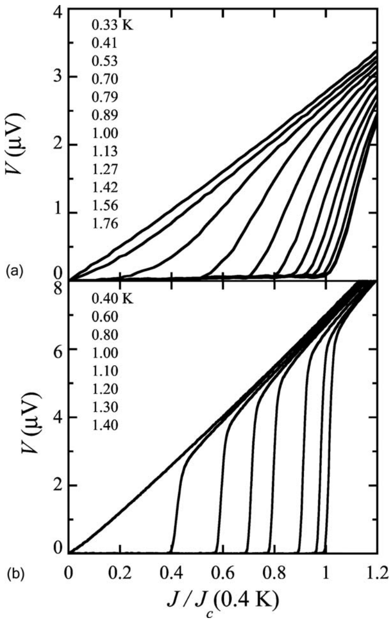

FIG. 1. Voltage versus critical current density for (a) the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic and (b) pure Sr_2RuO_4 with a $T_c = 1.35$ K. Current is applied along the c axis.

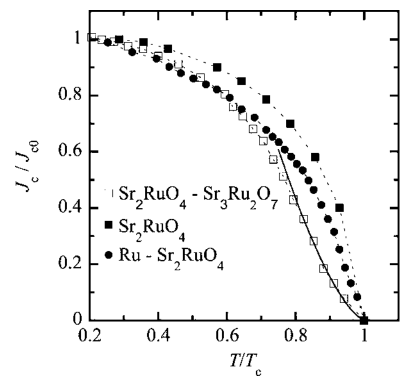

FIG. 2. Critical current density versus temperature for Sr_2RuO_4-$Sr_3Ru_2O_7$, Sr_2RuO_4, and $RuSr_2RuO_4$. Critical current density is normalized to its value at 0.3 K. Temperature is normalized to T_c. The solid line shows a fit to the Ginzburg-Landau temperature dependence of J_c close to T_c in Sr_2RuO_4-$Sr_3Ru_2O_7$.

measurements were made via a dc pulsed-current method with a duration of 1.5 ms and a 0.5 s delay between two successive pulses. The temperature measurements were made using a RuO_2 thermometer mounted in close proximity to the sample.

I-V curves for the Sr_2RuO_4-$Sr_3Ru_2O_7$ system ($T_c = 1.45$ K) are shown in Fig. 1(a), with the data for pure Sr_2RuO_4 ($T_c = 1.35$ K) included for comparison in Fig. 1(b). We chose to measure I_c along the c axis, since this configuration allows us to probe the interconnection of the two phases, which typically stack in layers along the c axis.[11] All of the curves are normalized to the critical current I_c at 0.4 K; an absolute comparison of the critical current densities is not possible, as the path taken by the supercurrent through the crystal is quite complicated and depends on the exact placement of the leads on the crystal. Data are shown in the first quadrant only; all of the curves are symmetric at the negative currents and show no anomalous asymmetry as seen in Ru-Sr_2RuO_4.[15]

Although the volume ratio of Sr_2RuO_4 to $Sr_3Ru_2O_7$ is close to 1, a continuous superconducting path exists through the crystal, giving rise to a zero voltage supercurrent as shown in the *I-V* curves in Fig. 1. The transition out of the normal state occurs over a much broader range of current as compared to the pure sample, which is similar to the behavior observed in the *I-V* curves of 3-K phase crystals.[15] The gradual nature of the transition suggests that the superconducting pathway for our eutectic crystal is not entirely through large, continuously connected Sr_2RuO_4 regions, but rather is likely limited by the Josephson tunnel junctions between the superconducting regions. This manner of broad transition was also observed in granular high-T_c superconductors and used as evidence for the existence of a Josephson network.[16]

The temperature dependence of the critical current $J_c(T)$ (defined as the point where an extrapolation of the *I-V* curve intersects the line $V = 0$, as in Ref. 15) is plotted in Fig. 2 for pure Sr_2RuO_4, and for the Ru-Sr_2RuO_4, and Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectics. J_c is normalized to its value at base temperature (0.3 K), and the temperature is normalized to T_c for easier comparison. Two prominent features suggest the presence of a Josephson weak-link network in the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic. First, similar to the behavior seen in the Ru-Sr_2RuO_4 system, the critical current shows a sharper initial drop than seen in pure Sr_2RuO_4 when the temperature is increased. This would be expected if the superconductivity in the eutectics is dominated by Josephson coupling.

Second, the line shape of $J_c(T)$ for Sr_2RuO_4-$Sr_3Ru_2O_7$ is consistent with other well-known Josephson weak link networks. Granular cuprate superconductors such as $YBa_2Cu_3O_{7-\delta}$ and conventional granular superconductors such as NbN have often been modeled as an array of Josephson-coupled superconducting grains.[17–21] Experimentally, a variety of temperature dependences in the critical current J_c have been reported; for example, three types of behavior have been observed in YBCO thin films: a concave curvature in $J_c(T)$ characteristic of an Ambegaokar-Baratoff mechanism,[17,22] a quasilinear behavior,[22,23] and a convex curvature characteristic of a Ginzburg-Landau mechanism.[19,24]

The critical current in homogeneous superconducting order parameter systems has been successfully modeled using Landau-Ginzburg theories, which predict a $J_c \sim (1 - T/T_c)^{3/2}$ behavior close to T_c.[25] For an array of Josephson-coupled grains, the model of Ambegaokar and Baratoff is frequently used;[17] this predicts a temperature dependence

$$J_c(T) = [\pi\Delta(T)/2e\rho_n d]\tanh[\Delta(T)/2k_B T], \quad (1)$$

where $\Delta(T)$ is the temperature dependent gap function, ρ_n is the normal state tunneling resistivity, and d is the cross section of the Josephson barriers. The scaling behavior of J_c for the granular Josephson weak-link superconductors is often roughly linear close to T_c, which is the limiting behavior of the Ambegaokar-Baratoff expression close to the transition temperature.[17] More precise measurements on YBCO single crystals and thin films (using a Hall probe to detect the magnetic field induced by a persistent supercurrent in ring-shaped samples to estimate the critical current J_c) revealed a crossover between an Ambegaokar-Baratoff-like (AB) temperature dependence in critical current to a Ginzburg-Landau (GL) dependence at some characteristic temperature.[26]

The Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic has a temperature dependence very similar to that seen in YBCO thin films, which has an AB-like dependence at lower temperatures but crosses over into a Ginzburg-Landau $(1-T/T_c)^{3/2}$ behavior close to T_c.[26] In our Sr_2RuO_4-$Sr_3Ru_2O_7$ sample, this crossover occurs at roughly $T=0.8\ T_c$; above this temperature J_c varies almost exactly as $(1-T/T_c)^{3/2}$ (see the solid line in Fig. 2). At lower temperatures it regains the concave line shape typical of AB behavior; a meaningful fit to the AB formula would require the normal state resistance of a junction and the temperature dependent gap function for Sr_2RuO_4, which are currently unknown. Clem *et al.* noted that the standard AB expression for the maximum dc Josephson current in a Josephson array failed to take into account the ability of the supercurrent to suppress the gap; the inclusion of this suppression predicts a crossover from AB to GL dependence when the Ginzburg-Landau coherence length is on the order of the grain size for materials with a strong Josephson coupling. If the grain size is less than the coherence length, the supercurrent will not "see" the junctions and a Ginzburg-Landau temperature dependence will emerge.[27]

The GL coherence length $\xi(0)$ for Sr_2RuO_4 is 660 Å within the *ab* plane and 33 Å along the *c* axis;[2] though current is ostensibly applied along the *c* axis, given the quasi-two-dimensional (2D) nature of the system and the formation of an inhomogeneous network it is very likely that the superconducting path may have considerable in-plane components. Using Clem's picture for the AB to GL transition in the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic, this would suggest the presence of superconducting microdomains of Sr_2RuO_4 on the order of a few tens of nanometers, coupled by Josephson tunnel junctions.[26,27] While it is not possible from our current data to completely rule out the existence of a small granular network, it seems unlikely given the macroscopic scale of the Sr_2RuO_4 and $Sr_3Ru_2O_7$ domains observed in microscopy.[11]

Another more feasible scenario is that $Sr_3Ru_2O_7$ may become superconducting by an unusual proximity effect, a possibility which is hinted at by the unusual extra features in the susceptibility of the Sr_2RuO_4-$Sr_3Ru_2O_7$ system.[11] Similar to the proposed model for the 3-K phase, the interface between Sr_2RuO_4 and $Sr_3Ru_2O_7$ regions could result in internal stress which is compensated for by local structural distortions such as a change in the in-plane rotation of the RuO_6 octahedra.[10] This seems very possible considering the deviation from the ideal Ruddlesden-Popper structure observed in $Sr_3Ru_2O_7$, especially with regard to the rotation of the octahedra.[28] The structural distortion near the interface might result in the occurrence of superconductivity in $Sr_3Ru_2O_7$ over large length scales. This interface-strain induced superconductivity should intrinsically differ from the general proximity effect in normal metals, which in the clean limit occurs over a characteristic length $\xi_n=\hbar v_n/2\pi k_B T$, where v_n is the Fermi velocity in the normal metal; for $Sr_3Ru_2O_7$, ξ_n would be on the order of 0.1 μm.[20,29] A significant penetration of the superconducting order parameter of Sr_2RuO_4 into the nonsuperconducting $Sr_3Ru_2O_7$ regions might suggest that the Ginzburg-Landau coherence length of Sr_2RuO_4 is not the appropriate length scale to use in modeling the AB to GL crossover. An appropriate length would be considerably larger than the coherence length, increasing the coupling between Sr_2RuO_4 regions significantly so that an AB to GL crossover could still be observed even with the micrometer separation between Sr_2RuO_4 domains. There may also be some parallels here with the so-called "giant proximity effect" seen in high-T_c superconductors, where the supercurrent appears to penetrate over distances far beyond the characteristic length ξ_n if the barrier material is nearly superconducting;[30] this is possible considering how similar $Sr_3Ru_2O_7$ is to Sr_2RuO_4 crystallographically.[13]

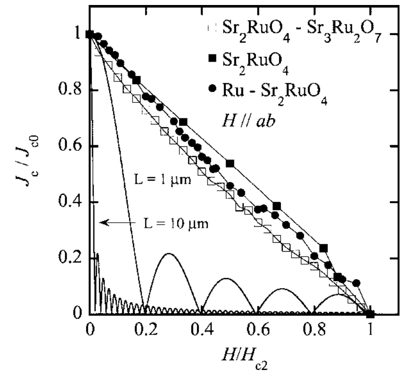

FIG. 3. Critical current density versus field for Sr_2RuO_4-$Sr_3Ru_2O_7$, Sr_2RuO_4, and Ru-Sr_2RuO_4 for field in the *ab* plane. Critical current density is normalized to its value at 0 T, and the field is normalized to the upper critical field H_{c2}. The solid lines show the theoretical expectation for $J_C(H)$ for an array of identical junctions, assuming junction lengths of 1 and 10 μm.

We note that $Sr_3Ru_2O_7$ shares a common *c* axis with Sr_2RuO_4 in their eutectic mixture[11] and is a normal metal at low temperatures.[31] In this situation we might expect that a model for a superconducting-normal metal-superconducting (SNS) array would be most appropriate; the model of DeGennes, which uses a spatially dependent pair potential to describe boundary effects, is often used to interpret critical current data in SNS systems.[20] This model predicts $J_c \sim (1-T/T_c)^2$ near T_c, while at lower temperatures the term $J_c \sim \exp(b\sqrt{T/T_c})$ dominates, where b is a measure of the thickness of the normal metal barrier.[32] SNS junctions made from ceramic YBCO with intergranular silver show a crossover from Ambegaokar-Baratoff temperature dependence to DeGennes-type behavior close to T_c, but the application of a small field quickly transforms this dependence into a Ginzburg-Landau type over a broad temperature range.[26] The reason why this behavior is not observed in our material might also be associated with the possibility of significant penetration of the superconducting order parameter of Sr_2RuO_4 into $Sr_3Ru_2O_7$. In this situation we might not expect a typical SNS Josephson interface.

In Fig. 3 we present the field dependence of critical current $J_c(H)$, with J_c normalized to its zero field value and the field normalized to the upper critical field H_{c2}. Similar to the behavior seen in Ru-Sr_2RuO_4, the field suppresses the critical current in the Sr_2RuO_4 eutectic more quickly than in the bulk material; this is consistent with the existence of superconductivity mediated by weak-link networks, since the field can quickly penetrate the Josephson barriers and suppress the tunneling current.

The line shape of $J_c(H)$ of both eutectic systems differs considerably from that seen in granular superconductors, where a drop in J_c up to two orders of magnitude is often observed at low fields, followed by an asymptotic approach to zero as the field is increased.[16,33] From the Josephson equations we expect for a single Josephson tunnel junction with a length less than the Josephson penetration depth

$$J_c(H) = J_c(0)\left|\frac{\sin(\pi H/H_0)}{\pi H/H_0}\right|, \tag{2}$$

where the characteristic field H_0 is

$$H_0 = \Phi_0/\mu_0 dL. \tag{3}$$

Here Φ_0 is the flux quantum, $d=2\lambda+t$ is the junction thickness, λ is the penetration depth, t is the barrier thickness, and L is the junction length.[33] Equation (2) is a Fraunhoffer pattern, though for a true Josephson array averaging over a large array of randomly oriented junctions will smear out much of the Fraunhoffer structure.[33]

The theoretical expectations for a Sr_2RuO_4 Josephson junction (or an array of exactly identical junctions) with a thickness $t=1$ nm and a length $L=1$ μm and 10 μm are shown in Fig. 3. Due to the large size of the domains in the eutectic, we expect the junction length to be on the order of tens of microns, which should result in a drastic reduction of J_c at low fields. Here instead both eutectics show a quasilinear field dependence of critical current, with only slight differences in the slope as compared to the pure sample. This may suggest that the Josephson junction lengths in the samples are much larger than the Josephson penetration depth and thus the above model for $J_c(H)$ is not applicable. This scenario is feasible given the macroscopic size of the domains in the eutectics and the possibility of a large-scale proximity effect in $Sr_3Ru_2O_7$; this would result in a much more gradual suppression of J_c under field, which is consistent with our experimental results.

The Ru-Sr_2RuO_4 system shows a concave Ambegaokar-Baratoff-like (AB) temperature dependence below $T=0.9$ T_c. Close to T_c, the critical current scales approximately linearly with temperature; no apparent AB to GL transition is observed at zero-field in this eutectic, but as shown in our previous report[15] we do observe such a crossover in $J_c(T)$ around 0.93 T_c when measurements are taken under a magnetic field of 0.1 T. This is similar to the situation for the thin film YBCO, in which the application of a magnetic field reduces the AB to GL crossover temperature.[26] Thus, the quasilinear zero field $J_c(T)$ observed in the 3 K phase samples may correspond to a temperature dependence intermediate between the Ambegaokar-Baratoff and Ginzburg-Landau types. The quasilinear field dependence of J_c in this eutectic might have a similar origin as that for the Sr_2RuO_4-$Sr_3Ru_2O_7$ system.

We have studied the critical current of the Sr_2RuO_4-$Sr_3Ru_2O_7$ eutectic system. These crystalline phases coexist in the sample as macroscopic domains; a connected superconducting pathway is observed in this eutectic, with features distinct from that of the bulk superconducting phase. We observe a crossover from an Ambegaokar-Baratoff to a Ginzburg-Landau temperature dependence in the critical current, suggesting that the Sr_2RuO_4-$Sr_3Ru_2O_7$ system is a strongly coupled Josephson weak-link network in which, similar to granular high-T_c superconductors, current-induced pair breaking is an important limiting mechanism for J_c. Our analysis suggests that large regions of $Sr_3Ru_2O_7$ may become superconducting by an unusual proximity effect when intergrown with Sr_2RuO_4, a possibility which warrants more detailed study by local probe techniques.

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