Alignment of chiral order parameter domains in Sr$_2$RuO$_4$ by magnetic field cooling

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Superconductivity in Sr$_2$RuO$_4$ is unconventional, believed to be of $p_x \pm ip_y$ pairing symmetry. These two degenerate order parameters allow the formation of chiral domains separated by domain walls. In a Josephson junction formed on the edge of a single crystal of Sr$_2$RuO$_4$, the chiral domains can create a variation of the phase in the tunneling direction causing interference effects which suppress and modulate the critical current of the junction. Cooling the junction in a magnetic field lifts the degeneracy between the order parameter states and induces a preferential chirality, significantly modifying the phase interference. We present experimental results on Sr$_2$RuO$_4$/Cu/Pb Josephson junctions cooled in a magnetic field showing a dramatic enhancement of their critical current, giving direct evidence for the presence of chiral domains and their alignment in a magnetic field.

Sr$_2$RuO$_4$ (SRO) was discovered to be superconducting in 1994[1]. Since then it has proven to be a very intriguing and complicated superconductor. A large body of evidence [2] points towards unconventional superconductivity with an order parameter of the form $p_x + ip_y$: the electrons are paired in an odd orbital, spin triplet channel that breaks Time Reversal Symmetry (TRS). The broken TRS, first observed by muon spin rotation[3] and later verified by polar Kerr effect [4], is especially interesting since it can lead to the appearance of order parameter domains of opposite chirality $p_x \pm ip_y$ in the material, much like in a ferromagnet. Texture of the order parameter, has been observed in the Helium 3 A-phase, which is the superfluid analog of SRO, but until recently domains were never directly observed in a superconductor. They have been proposed to explain unusual features in the ultrasound attenuation and the rate of vortex creep dynamics in superconductors thought to break TRS: UPt$_3$, UBe$_13$ and SRO ([5],[6],[7],[8]), but their first unequivocal observation was through Josephson interferometry experiments done on SRO [9]. The presence of order parameter domains opens a whole new field in superconductivity and efforts are being made to understand domains nucleation and dynamics. Many of their properties are still not understood. For example, scanning SQUID and Hall microscopy have repeatedly failed to image the domains ([10],[11]) although theory predicts magnetic fields that are well above the resolution of these instruments.

In this Letter, we aim to further that understanding by manipulating the domains using applied magnetic fields. We find that field cooling increases the probability of one type of domain which translates into a critical current much higher than that observed in the same junction cooled in zero field. We also found an unexpected memory effect with training characteristics.

Superconductors with broken TRS were first studied theoretically by Volovik and Gor’kov [12] who found that they can have unique magnetic properties associated with the variation of the phase of the superconducting order parameter. Since the phase of a $p_x \pm ip_y$ order parameter continuously varies from 0 to $2\pi$ in $k$-space, a spontaneous current will flow within a coherence length of the surface of the material even in the ground state. To cancel the magnetic field in the bulk of the superconductor, additional Meissner screening currents flow within a penetration depth of the surface. Thus, although there is no net magnetization, there should still be a magnetic moment at the surface as well as at sites with a suppressed order parameter such as defects and impurities. To offset the cost of magnetic energy, the discrete degeneracy of order parameters may promote the formation of order parameter domains similar to those in ferromagnetic materials [12], but they may also nucleate spontaneously and become trapped by pinning of domain walls at defects or impurity sites. These so-called chiral domains are defined by the two directions of phase winding associated with the degenerate order parameters $p_x + ip_y$ and $p_x - ip_y$ and they couple to applied magnetic fields ([12],[13],[14]) via the spontaneous currents. The magnetizations associated with the two chiral domains have opposite signs so that an externally applied field causes a difference in their free energies, thus lifting the degeneracy of the order parameters. The chiral domains with a magnetization parallel to the applied field increase in size while the others shrink.

Josephson junctions have largely contributed to the understanding of the material. The determination of allowed and forbidden tunneling directions established constraints on the order parameter[15], SQUID interferometry showed the orbital part of the order parameter to be of odd symmetry[16], and they have proven to be very sensitive probes to the presence of domains ([9],[17]). Here, we use their critical current and interference patterns as a metric for the number and size of order pa-
rameter domains as we cool the junctions in a magnetic field parallel to the c-axis of the crystal. For a junction on the side of a crystal incorporating many domains, the phase difference across the junction can change from one domain to the next causing interference effects that reduce its critical current. The most drastic case is that of a phase difference of $\pi$ where the currents in neighboring domains flow in opposite directions and the measured critical current is the net sum of all of them (Fig. 1). Cooling the crystal in a magnetic field changes the ratio of domains, reducing the amount of cancellation. As one domain type increases in area, the critical current of the junction measured at zero magnetic field correspondingly increases and its diffraction pattern is modified, tending towards a Fraunhofer interference pattern. To illustrate this behavior, we performed computer simulations of critical current modulations in an applied magnetic field when one type of domain increasingly becomes more probable than the other. Here, $P_L$ and $P_R$ are the percentage of left- and right-handed chiral domains in the material. The calculation is done for tunneling from a superconductor with an s-wave order parameter to one with a $p_x \pm ip_y$ order parameter for a junction with 10 domains of random size across its width and assuming a phase difference of $\pi$ between the two polarities of chiral domains. The critical current is normalized: $I_c = 1$ is the maximum current that would flow through a domain free junction at zero applied field. The results are given in Fig. 2 As $P_R$ increases, the magnitude of the current increases and a central peak emerges, reminiscent of the Fraunhofer pattern expected for a domain-free junction. Of course, the detailed shape of the pattern depends on the size and number of domains that are used in the calculation but the qualitative trend is clearly shown by the simulations.

We performed the experiment, using the magnitude of the critical current and its magnetic field dependence as a measure of domain alignment. The high quality single crystals used in the experiment were grown using the floating zone method as described elsewhere [18]. The crystals were glued onto a glass substrate and the junctions defined using a flexible membrane mask. They are parallel to the c-axis, about 200 $\mu$m wide and 50$\mu$m high. The samples were then loaded into a vacuum chamber where they were cleaned by ion milling and then 10nm of Cu and about 1 $\mu$m of Pb were deposited by thermal evaporation. The critical current was measured using a feedback technique where the current output to the sample was automatically adjusted to maintain a set voltage just above the supercurrent regime. We used a SQUID potentiometer circuit to measure the small voltages generated. The magnetic field was applied parallel to the c-axis using a Helmholtz coil. The measurements were done in a $^3$He refrigerator at temperatures between 340mK and 1.3K. The data presented here was taken around 1K. The magnitude of the critical current vary with temperature but the shape of the observed diffraction patterns is relatively independent of temperature. In between cooling cycles, the samples were heated to 10K, which is above the transition temperatures of both superconductors in the device. The samples were then cooled in zero field to 4K to avoid trapping vortices in the Pb film. Then the field was turned on and they were cooled below the transition temperature of the SRO crystal.

This is a challenging experiment because when a junction is cooled in a magnetic field, vortices can be trapped in the superconductors near the junction that suppress
the critical current and distort the diffraction patterns. Hence a field cooled junction will show the combined effects of domain alignment and magnetic field from trapped vortices; depending on whether and where the vortices are trapped, the critical current enhancement might not be observable. Thus we performed the experiment at various field values in the mG range but found that the most significant effects were achieved at fields in the 10 to 100µG range. We attribute these effects to domain alignment. Significant critical current enhancement was observed for four samples for both positive and negative fields. Fig.3 shows the diffraction patterns of zero field cooled (ZFC) and field cooled (FC) junctions for a field of 30µG at which the maximum enhancement was observed. The diffraction patterns are not of the Fraunhöfer type expected for a domain-free junctions: the domains cause phase interference that induces distortions [9]. Even so, a dramatic enhancement of the critical current by more than a factor of 2 and the emergence of a central peak can be observed, both evidence for less interference in the junction. The change in the shape of the diffraction pattern is particularly important as it mostly rules out explanations based on a magnetic field induced increase in coupling between the singlet (Pb) and triplet (SRO) superconductors which would change the magnitude of the critical current but not the interference pattern of the junction. The field cooled junctions are never found to be domain free but this is not surprising given the small fields applied. Out of the four samples measured, the one presented here showed the most significant effect, and the smallest increase observed was 40%. We also cooled some of the samples in a field parallel to the $ab$ plane. No critical current enhancement was observed, additional evidence that the enhancement in perpendicular field is due to order parameter domains in SRO.

Although the data shows the critical current enhancement, in agreement with the theoretical predictions, some aspects of observed enhancement are surprising. First, the optimal enhancement is achieved at extremely small fields, comparable or even smaller than the residual background fields in our system. We are able to observe these effects because the measurement setup has a double layer of mu-metal shielding and a superconducting Pb can that reduces the earth’s magnetic field to about 100µG on average, although this can be significantly lower in the junction area and may not have a large component in the relevant field direction. We should note that the effective field in the junction area is always higher than the nominally applied field because of flux focussing: large areas surrounding the junction are covered by superconductors that expel magnetic field and focus it into the normal regions of the sample. The magnetic field in the junction is self calibrated because the periodicity of the diffraction pattern corresponds to threading one flux quantum through the junction. Although the patterns presented here are complicated, the smallest modulation period corresponds to the largest area, which is the magnetic cross-section of the entire junction. Knowing the junction area, we can then calculate the effective magnetic field. For the sample shown above, we estimate the flux to be focused by a factor of 4: the 30µG externally applied magnetic field value quoted above should translate into about 120µG in the junction area. Despite the rather small value, the fact that we observe comparable critical current enhancement for both positive and negative applied field indicates that we are indeed applying a net field, and not offsetting a residual background field. One would think that a larger field would split the degeneracy more, eventually yielding a domain free junction but we do not observe this and attribute this to the trapping of vortices near the junction which distort the diffraction pattern.

Second, we observed anomalous training and memory effects in field-cooled samples. The critical current enhancement is found to increase with the number of field cooling cycles, reminiscent of the training effect observed in ferromagnet/antiferromagnets multi-layers. The critical current does not reach its peak level in the first iteration of field cooling. Instead, it gradually increases with the number of field coolings, eventually reaching a maximum as can be seen on Fig.4. The three curves shown were measured on ZFC junctions, following the respective number of field cooling noted on the figure. We also observed a memory effect: once the critical current of the junction is enhanced, it retains a high value even when warmed above the transition temperatures of all superconductors in the device. These observations are plotted
The system relaxes with time and temperature, but at temperatures as high as 77K and times as long as 24h, the memory of previous cycles is not fully erased. Fig. 5 shows 3 diffraction patterns taken on a ZFC junction. The first one was taken before any field cooling (black), the second in a cooling cycle immediately following the maximum $I_c$ enhancement, and the third was taken one day after the critical current enhancement is observed. The junction warmed to 77K overnight; the critical current is lower than immediately after the field cooling but still higher than in the original state. Because of the long times required to map the junction behavior, we have not determined the conditions under which the field cooling effects disappears completely.

The memory effect is obviously not caused by the superconductivity in the material since it survives well into the normal state. The most likely explanation would be the presence of isolated ferromagnetic regions at the edge of the junction that would become magnetized during the field cooling and then provide a local magnetic field on the subsequent cooling cycles. A ferromagnetic material far away from the junction wouldn’t provide a credible explanation because magnetic fields larger than the ones used for field cooling are routinely applied to the sample when measuring diffraction patterns and don’t result in a critical current enhancement. Hence whatever causes the memory effect probably originates from inside the superconductor which is shielded by Meissner screening during the field cooling pattern measurements. Magnetic impurities are very unlikely as the superconductivity is very fragile and vanishes with small amounts of impurities, even non-magnetic ones. A transition temperature of 1.5K is only achieved in extremely clean samples. A more likely scenario would be a very small amount of impurity phase of SrRuO$_3$, which is a ferromagnet with a Curie temperature of 150K. Another possibility is that of surface magnetism.

In conclusion, we present experimental results showing a dramatic enhancement of the critical current in SRO/Cu/Pb Josephson junctions cooled in a magnetic field, which is expected to occur in the presence of order parameter domains in the SRO crystal. This increase of the critical current indicates that one of the order parameters, e.g. $p_x + ip_y$, is increasing in size while the other is decreasing, resulting in an increased net current through the junction. We also report memory and training effects in the field cooled SRO samples that have yet to be fully explained but may result from local magnetic inclusions.

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