Signature of a crossed Andreev reflection effect (CARE)
in the magnetic response of $YBa_2Cu_3O_{7-\delta}$ junctions
with the itinerant ferromagnet $SrRuO_3$

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Magnetic properties of SFS and SF ramp-type junctions with $YBa_2Cu_3O_{7-\delta}$ (YBCO) electrodes (S), and the itinerant ferromagnet $SrRuO_3$ (SRO - F), were investigated. We looked for a crossed Andreev reflection effect (CARE) in which an electron from one magnetic domain in F is Andreev reflected as a hole into an adjacent, oppositely polarized, domain while a pair is transmitted into S. CARE is possible in SRO since the width of its domain walls is of the order of the YBCO coherence length (2-3 nm). Our junctions behave as typical magnetic tunneling junctions, as the conductance spectra were always asymmetric, and a few showed bound state peaks at finite bias that shifted with field according to the classical Tedrow and Meservey theory. In many of our SFS junctions with a barrier thickness of 10-20 nm, a prominent zero bias conductance peak (ZBCP) has been observed. This peak was found to decrease linearly with magnetic field, as expected for Andreev and CARE scattering. In contrast, in SF junctions, the observed ZBCP was found to decrease versus field almost exponentially, in agreement with the Anderson-Appelbaum theory of scattering by magnetic states in F. Thus, transport in our SFS and SF junctions depends strongly on the size of the F layer.

We also found that in both cases, the ZBCP height at zero field decreased with increasing magnetic order of the domains in F, in agreement with the CARE mechanism.

Properties of SFS ramp type junctions of $YBa_2Cu_3O_{7-\delta}$ (YBCO) electrodes (S) and $SrRuO_3$ (SRO) barrier (F) have been investigated by two groups more than ten years ago [1, 2]. Both groups have found that the normal resistance values of their junctions show two distinct phenomena. One, that the observed values had a large spread from a few tens of Ohms to a few hundred Ohms, and the other that they were two or more orders of magnitude higher than the expected Ohmic resistance of the SRO film. The high normal resistance was therefore assumed to originate at the Ohmic resistance of the SRO film. The high normal resistance values of their junctions show more orders of magnitude higher than the expected hundred Ohms, and the other that they were two or more orders of magnitude higher than the expected Ohmic resistance of the SRO film. The high normal resistance was therefore assumed to originate at the classical Tedrow and Meservey theory. In many of our SFS junctions with a barrier thickness of 10-20 nm, a prominent zero bias conductance peak (ZBCP) has been observed. This peak was found to decrease linearly with magnetic field, as expected for Andreev and CARE scattering. When the barrier material is fully spin polarized in one direction, no Andreev transport is possible. If however, the ferromagnetic barrier has many domains with opposite polarizations, a crossed Andreev reflection effect (CARE) is possible [4]. This effect can occur at the intersection of the domain walls and the YBCO electrodes at the interfaces, provided the value of the domain walls width is similar to that of the superconductor’s coherence length $\xi$ (2-3 nm for optimally doped YBCO). We have chosen to study junctions with an SRO barrier since the domain wall width of this highly anisotropic ferromagnet is very narrow ($\sim$3 nm only [10, 11]), and fulfils the above condition. As will be described in the following, our conductance spectra results under magnetic fields provide supportive evidence for the existence of CARE in our junctions. It should be noted that recently CARE was observed by Beckmann et al., in conventional FSF junctions made of two closely spaced Fe nanowires in contact with an Al electrode [12]. They found that the resistance difference between parallel and antiparallel magnetization of the Fe electrodes when the Al electrode was in the superconducting state, decay with increasing distance of the Fe electrodes up to about twice the coherence length of Al, in agreement with the CARE phenomena. Technically however, it is impossible to reproduced this kind of study in the high temperature superconductors due to their extremely short coherence length.

In the present study we revisit the same type of junctions of YBCO and SRO, with a special focus on transport properties which are affected by the magnetic nature of the barrier material. In the absence of a critical current, transport in junctions at voltage bias values below the energy gap of the superconductor is controlled by Andreev scattering. When the barrier material is fully spin polarized in one direction, no Andreev transport is possible. If however, the ferromagnetic barrier has many domains with opposite polarizations, a crossed Andreev reflection effect (CARE) is possible [4]. This effect can occur at the intersection of the domain walls and the YBCO electrodes at the interfaces, provided the value of the domain walls width is similar to that of the superconductor’s coherence length $\xi$ (2-3 nm for optimally doped YBCO). We have chosen to study junctions with an SRO barrier since the domain wall width of this highly anisotropic ferromagnet is very narrow ($\sim$3 nm only [10, 11]), and fulfils the above condition. As will be described in the following, our conductance spectra results under magnetic fields provide supportive evidence for the existence of CARE in our junctions. It should be noted that recently CARE was observed by Beckmann et al., in conventional FSF junctions made of two closely spaced Fe nanowires in contact with an Al electrode [12]. They found that the resistance difference between parallel and antiparallel magnetization of the Fe electrodes when the Al electrode was in the superconducting state, decay with increasing distance of the Fe electrodes up to about twice the coherence length of Al, in agreement with the CARE phenomena. Technically however, it is impossible to reproduced this kind of study in the high temperature superconductors due to their extremely short coherence length.
We prepared the YBCO based ramp-type junctions with SRO on (100) \(\text{SrTiO}_3\) (STO) wafers with a ramp angle of \(\sim 35^\circ\). This was done by a multi-step process, where the epitaxial thin film layers are prepared by laser ablation deposition, patterning is done by deep UV photolithography, and etching by Ar ion milling \[13\]. The YBCO films had \(c\)-axis orientation normal to the wafer. The thickness of the base and cover electrodes was kept constant at 80 nm, while the SRO thickness on different wafers ranged between 4 and 80 nm. On each wafer we patterned ten identical junctions along the (100) direction, with a width of 5 \(\mu\)m. Finally, a gold layer was deposited and patterned to produce the 4 \(\times\) 10 contact pads for the 4-probe transport measurements. The YBCO electrodes of our junctions had oxygen content close to optimal doping with a \(T_c\) of 88-89 K. The quality of our junctions fabrication process was tested by measuring the critical current density \(J_c\) of “shorts” (junctions without any barrier). We found \(J_c(77K) \sim 1 \times 10^6 \text{ A/cm}^2\) which is reasonable compared to \(J_c(77K) \sim 3 - 5 \times 10^6 \text{ A/cm}^2\) found in the best blanket films.

Fig. 1 shows the resistance versus temperature of several junctions on a single wafer. The spread of resistance values at temperatures above \(T_c\) is extrinsic and due to the different length of the YBCO leads to the junctions. At low temperatures, the resistance values have an intrinsic large spread of about one order of magnitude. These resistance values are also several order of magnitude higher than the calculated 10 m\(\Omega\) Ohmic resistance of the junction obtained by using the resistivity of the \(\text{SrRuO}_3\) film (see inset (a) to Fig. 1). These observations are similar to the results reported previously on the same kind of junctions by other groups as discussed in the introduction to this paper \[1, 2\]. Unlike previous results however, our junctions generally had a critical current up to a barrier thicknesses of \(\sim 10\) nm, but were resistive at higher barrier thicknesses. Antognazza \textit{et al.} found critical currents with a barrier thickness of up to 20 nm \[1\]. This is possibly due to microshorts or tunneling via oxygen disorder states in their junctions. Dömel \textit{et al.} have found that the inverse resistance difference \(1/R - 1/R_0\) of their junctions (where \(R_0\) is the extrapolated resistance to \(T = 0\)), varies versus temperature as \(T^{4/3}\). They concluded that this indicates tunneling via one and two localized states in the barrier \[2, 13\]. We basically observed a similar behavior as shown in inset (b) to Fig. 1. As one can see however, the results are very sensitive to \(R_0\) and the \(T^{4/3}\) behavior is not obtained with the extrapolated \(R_0\), but a value close to it. Furthermore, in Dömel \textit{et al.} study, there is no data between 5-20 K. If we use our data in this temperature range, and with the extrapolated \(R_0\) value, we find a linear dependence versus \(T\). Thus we believe that tunneling via two localized states is not the dominant transport mechanism in our junctions.

Fig. 2 shows the normalized conductance spectra at low temperature of several junctions on a single wafer with a barrier thickness of 12 nm. One observes that two junctions have a ZBCP inside a tunneling-like structure, while the others have only the tunneling-like behavior. The ZBCP can be attributed to either Andreev reflections or scattering by magnetic states \[13, 17\]. Since our junctions are orientated along the \(a\) or \(b\) axes of the YBCO electrodes, the observed ZBCP is not due to the well known bound states which are formed along the node direction in a d-wave superconductor. In addition, we point out that there is a correlation between the appearance of a ZBCP and the behavior of

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**FIG. 1:** (Color online) Resistance versus temperature of several junctions on a single wafer. Inset (a): resistivity versus temperature of a 90 nm thick \(\text{SrRuO}_3\) film on (100) STO. Inset (b): inverse resistivity of one of the low resistance junctions (J6) versus \(T\) on a log-log scale (solid line - \(T^{4/3}\), dashed line - \(T^1\)).

**FIG. 2:** (Color online) Normalized conductance spectra of several junctions on a single wafer. Inset: zoom up on the resistance versus temperature of a few junctions.
the resistance curves below $T_c$ (see the inset to Fig. 2). For junction J7 and J8, where a ZBCP was observed, the R versus T curves show a change of slope (a cusp) and not a monotonic increase with decreasing temperature as for instance is found in junctions J5 and J6. Another distinct feature in Fig. 2 is that the spectra are clearly asymmetric. This asymmetry is typical of ferromagnetic tunneling junctions due to the opposite shifts of the spectra for up and down spins, and the non zero spin polarization of the magnetic electrode $^{18}$. Further support to the fact that our junctions behave as classical magnetic tunneling junctions, is found in Fig. 3. In this figure one sees the prominent ZBCP and its suppression under applied magnetic fields. But first we shall focus on the bound state peaks observed at about ±10 mV. As can be seen by the zoom up on these parts of the spectra, a positive magnetic field shifts the peak to negative bias, and vice versa. The total measured shift for fields of ±5 T is $\sim 1.2$ mV. The expected shift for an SFS junction is $4\mu H$ (1.28 mV here) $^{18}$. Thus for a bound state of energy $\Delta_1$, the expected shift $4\mu H/2\Delta_1$ should be equal to the measured shift $\sim 1.2/\Delta$ where $\Delta$ is the gap energy. Therefore, $2\Delta_1 \sim \Delta$ and if $\Delta$ of YBCO is $\sim 20$ meV then $\Delta_1 \sim 10$ meV, in agreement with the peaks bias of the bound state in Fig. 2.

Fig. 4 shows a few conductance spectra under different fields and field cooling conditions. There is almost no effect on the spectra at any given field larger than about 0.1T, whether it was obtained under zero field cooling (ZFC) or field cooling (FC). The insets of Fig. 4 show the ZBCP area above the background conductance, and the conductance at zero bias ($G_0 \equiv G(V = 0) = 1/R(V = 0)$) versus field. Surprisingly, both features show a linear decrease with increasing field, except maybe for fields near zero field. The expected decrease of $G_0$ versus field due to scattering by magnetic states in junctions was calculated by Appelbaum and found to be almost exponential $^{19}$. A similar behavior, but with a more gradual decrease versus field, was found also in experiments done in Ta-I-Al tunnel junctions $^{20}$. Thus the linear $G_0$ versus $H$ result in our junctions points to a different scattering mechanism. A theoretical calculation of the current and magnetoresistance in FSF junctions due to CARE was recently published, but it did not include conductance spectra which are relevant in the present study $^{21}$. The closest theoretical calculation we could find for a ZBCP behavior versus $H$ was in a study by Tanaka et al. $^{22}$. They calculated the conductance spectra for the node direction in the cuprates using the extended BTK model for the d-wave superconductors. Clearly, the resulting ZBCP is due to bound states because of the sign change of the order parameter, and not to the CARE process. Nevertheless, the basic scattering mechanism is still Andreev reflections, and therefore a comparison of our data with their results is justified. Extracting $G_0$ from their conductance spectra at different fields, one finds a clear linear decrease with field. Hence, our data is consistent with this behavior, and it is likely that simple Andreev and CARE play a dominant role in the transport of our SFS junctions.

Fig. 5 shows a series of conductance spectra in another junction. These spectra were obtained under various fields starting with ZFC to 4 K, ramping up to 8 T, and going back to zero field. Here again as in Fig. 4, one finds a linear decrease of the ZBCP height versus field as shown in the inset to this figure, but the ZFC data point seems to stand out. Clearly the measured ZBCP height after ZFC is larger than that obtained after field cycling.
to 8 T and back to 0 T. To understand this behavior, we note that during the ZFC process, the SRO barrier layer becomes ferromagnetic with many domains and domain walls as shown schematically in the upper-right corner of the inset to Fig. 5. As a result, the contribution of CARE to the conductance which depends on the number of domain wall intersections with the S electrodes, should be higher than the conductance after field cycling. This is so since the magnetic memory after the field cycling reduces the number of domains as shown schematically in the lower-left corner of the inset to Fig. 5. Because this is the exact observed result, we conclude that CARE is responsible for the excess conductance at zero field under ZFC as shown in the inset to Fig. 5. We stress that this phenomenon is a unique signature of CARE, which can not be explained by the standard Andreev reflections.

Next, we decided to look at the limit of a very thick barrier. Since the Ohmic resistivity of SRO is quite small at 4 K, only ~ 50 $\mu\Omega$cm as shown in inset (a) of Fig. 1, we chose to study SF rather than SFS junctions. In this case, the F electrode "thickness" or size is almost infinite, and therefore its relatively weak itinerant ferromagnetism should be enhanced. The resulting conductance spectra of a typical SF junction are shown in Fig. 6. The spectra and the ZBCP height values versus field were obtained by ZFC to 4 K, ramping to 6 T, and going back to 0 T. The ZBCP inside a gap-like structure is still present, but its magnitude is greatly reduced compared to the previous data in SFS junctions. The interesting feature here is that already at 4 T, the ZBCP is almost fully suppressed. Even more amazing is the field dependence of the ZBCP height as shown in the inset to Fig. 6. This is clearly nonlinear, and rather close to exponential decay. Actually, this decay is very similar to that predicted by the Anderson-Appelbaum (AA) theory of scattering by magnetic states close to the interface with a superconductor [16, 17, 20]. It should be noted that in the AA model this decay is due to the increased Zeeman splitting of the ZBCP [19]. We however, have never observed splitting of the ZBCP, and this could be due to a larger magnetic relaxation rate in SRO which broadens this peak and smears the splitting. It is therefore concluded that the almost exponential decay versus field indicates that the dominant transport mechanism now is not Andreev scattering, but rather magnetic scattering. We conclude that the size of the F electrode plays an important role in determining the transport properties of our junctions. When the F electrode is thin as in the previous results of SFS junctions (10-20 nm), its ferromagnetism is weak and the proximity effect by the S electrodes makes it even weaker. The opposite is true when the F electrode size is large as in the SF junctions case. Then the proximity penetration of superconductivity into F is small compared to the mean free path in the F electrode (which is unlimited now by the junction length), the ferromagnetic order in F is robust, and the transport in the junction is controlled mostly by magnetic scattering.

We note that the value of the ZBCP height after ZFC from room temperature to 4 K is still much larger than its value after field cycling to 6 T and back to 0 T, similar to the result in the SFS junction of Fig. 5. It is tempting to attribute this behavior to CARE as before, but then the absence of a linear decreasing component of the ZBCP height versus field which originates in Andreev scattering, will have to be explained. According
to Yokoyama et al. who calculated the conductance spectra due to magnetic scattering in SN junctions with a d-wave superconductor \[23\], an enhanced magnetic scattering rate (by the higher magnetic disorder after ZFC in the present study) would decrease rather than increase the ZBCP. Since this is opposite to observation, it seems that we are still dealing with suppression of the ZBCP height due to CARE here (from the magnetically disordered ZFC state to the more ordered state after field cycling, similar to the result in Fig. 5). Apparently, in the SF case where the ferromagnetism of $F$ is robust, the linear suppression of the ZBCP height versus field is much enhanced and terminates at a much smaller applied field. This leaves only the exponential decay versus field due to magnetic scattering as the dominant process.

In conclusion, we have found significant magnetic effects in the transport properties of SFS and SF junctions of YBCO and the SRO ferromagnet. i) We observed an asymmetry in the conductance spectra, and shifts of bound state peaks with field, which are typical of magnetic tunneling junctions. ii) In both type of junctions a prominent ZBCP was observed. Its height decreased linearly with increasing field in SFS junctions, but almost exponentially in the SF case. The ZBCP height dependence on $H$ originated in normal Andreev and CARE in the SFS junctions, but was dominated by magnetic scattering in the SF junctions. iii) The observation of a higher ZBCP height at 0 T after ZFC as compared to the value after field cycling is due to the higher magnetic disorder after ZFC in both SFS and SF junctions. This is a strong signature of a CARE phenomenon in our junctions. Finally, we note that a calculation of the conductance spectra under fields in SF and SFS junctions is needed for a more quantitative comparison with the present results.

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