Triplet pair correlations and non-monotonic supercurrent decay with Cr thickness in Nb/Cr/Fe/Nb Josephson devices

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Supercurrents carry charge but not spin in the vast majority of superconductors. This is because the charge carriers are Cooper pairs which are formed of electrons with antiparallel spins (‘singlet pairs’). It is now established that if singlet pairs propagate through a superconductor interface with an inhomogeneous ferromagnet, triplet pair correlations form so the supercurrents can acquire a net spin component. Although the spins at sputter deposited Fe/Cr interfaces can be frustrated due to surface roughness and interdiffusion, an antiferromagnetic spin density wave (SDW) state can still form in Cr close to the interface. Here we show evidence for triplet pair correlations in Josephson junctions with Cr/Fe and Cr/Fe/Cr barriers. Although the exact mechanism of pair conversion is unknown, we propose a simple model in which a SDW state in Cr and frustrated spins at the Cr/Fe interfaces serve as spin-mixers for generating triplet supercurrents and so provide a potential means to generating large spin current densities in superconductors

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Spin-triplet theory[1-4] and experiments[5-20] which provide evidence of triplet pairing have mainly focused on triplet pair formation at magnetically inhomogeneous superconductor-ferromagnet (S-F) (so-called "spin-mixer" interfaces). In S-F-S Josephson devices in which both S-F interfaces serve as spin-mixers, Cooper pairs with parallel rather than antiparallel spins can form meaning that supercurrents in the F layer can potentially carry spin in addition to charge. Consequently, triplet supercurrents could be used as transmitters of spin in logic circuits without ohmic dissipation and so a better understanding of the triplet proximity effect and the discovery of other spin-mixers could lead to the development of superconducting spin-electronics as a major research field.[4]

The basic picture of how spin-polarized triplet pairs are generated in S-F junctions can be summarised as follows. If the magnetization at the interface is collinear to the magnetization of the F layer, spin-zero triplet pairs are induced.[1] Like singlet pairs these are short-ranged in F materials but, unlike singlet pairs, spin-zero triplets are rotationally variant and so if the interface magnetization makes angle to the magnetization of the F layer, spin-polarized triplet pairs form which are long-ranged in F. For a review see Ref. 4.

Spin-triplet theory was initiated[1] more than a decade ago following the report of long-range (>>10 nm) superconducting pair correlations in Al/Co[5] and Al/Ni[6] devices in which singlet pairing seemed impossible. Interferometer measurements also demonstrated evidence of long-ranged superconducting pair correlations in Ho.[7] More direct evidence of triplet pairing was in obtained in Josephson devices with half-metallic ferromagnetic CrO_2 barriers[8] - the absence of minority spin states at the Fermi energy of CrO_2 ruled out singlet pairing and so implied the supercurrents were not only carried by triplet pairs[21] but that the supercurrents were 100% spin-polarized.

More recently, engineered spin-mixers have been demonstrated in Josephson devices with F'-F-F'-type barriers,[9-11,13,14] as theoretically proposed in Ref. 3. Here the F' and F layers are non-collinearly aligned and so the F'-F interfaces serve as spin-mixers while the thickness of the F layer should be larger than the singlet pair coherence length (typically shorter than 1-3 nm[22-25]). Experimental device barriers for generating triplet supercurrents are very complicated and include, for example, Cu/Ni/Cu/Co/Ru/Co/Cu/Ni/Cu,[9,13] Co/Ho/Co,[10] and Ni/$Cr0_2$/Ni.[14] Evidence of triplet pairing has also been reported by Leksin *et al*.[26] and Zdravkov *et al*.[27] from critical temperature measurements of S-F'-F-type structures, as theoretically proposed in Ref. 28.

Identifying and investigating alternative spin-mixer structures for generating triplet pair correlations is important since existing structures contain many interfaces are materials with poorly matched resistivities. These factors limit device performance by reducing pair transmission and spin current density which can be carried by the triplet state. Here we report results using Cr, a spin-density wave (SDW) antiferromagnet (AFM), as a spin-mixer in Josephson junctions with Cr/Fe and Cr/Fe/Cr barriers.

In order to investigate the magnetic properties of our Fe/Cr interfaces, we performed current-in-plane (CIP) magnetoresistance (MR) measurements on Nb/Cr/$(Fe/Cr)_{\times 20}$ films and magnetization vs in plane field hysteresis loops (*M*-*H* loops). The films were grown by d.c. magnetron sputtering in Ar at 1.5 Pa onto unheated single crystal silicon (001) substrates with a 250-nm-thick oxide layer on the surface. Before film growth, the system was cooled via a liquid nitrogen jacket in order to lower the system's base pressure to below 10^{-8} Pa. Targets were pre-sputtered until a constant voltage was achieved. Substrates rested on a circular sample table, which could be rotated below the stationary sputtering targets. Film thicknesses were controlled by the speed of rotation and target power with a resolution better than 1 nm. Growth rates were pre-calibrated by growing films onto patterned substrates and by measuring the height of step edges with an atomic force microscope.

Figure 1(a) shows CIP MR data from a Nb/Cr/$(Fe/Cr)_{\times 20}$ film. The field was applied parallel to the interfaces. For all temperatures, the resistance (R) of a film is largest near zero field, but decreases as the field increases implying a change in the magnetization alignment between neighbouring Fe layers from (approximately) antiparallel in zero field to parallel in high field. However, the low temperature MR data shows no evidence of R reaching a saturation minimum value implying that a fully parallel state is not obtained in the field range investigated. This behaviour contrasts with the magnetization versus applied field loop (M-H) data shown in Fig. 2(b) where, for all temperatures, saturation seems to be achieved below 1T.

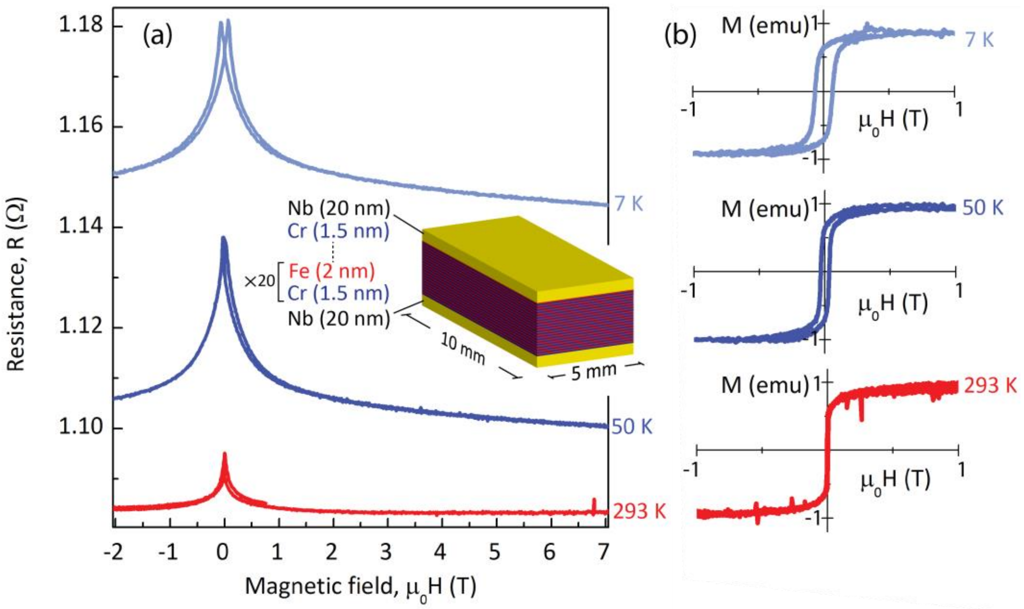

Fig. 1. Magnetic properties of a Nb/Cr/$(Fe/Cr)_{\times 20}$/Nb film. (a) Current-in-plane magnetoresistance data at different temperatures in which the field is applied parallel to the long-axis of the sample and perpendicular to the current direction. (b) Corresponding *M*-*H* loops at different temperatures.

A high field saturation of MR in Fe/Cr multilayers can be explained if we assume that the spins at the Fe/Cr interfaces are frustrated.[29] In Fe/Cr films the following magnetic interactions take place: ferromagnetic Fe-Fe and AFM Cr-Cr interactions within Fe or Cr layers, and AFM Fe-Cr interactions at the interface. For atomically flat an pristing Fe/Cr interfaces the spins point in plane meaning all three interactions coexist while at more realistic Fe/Cr interfaces, roughness and interdiffusion complicate the interactions by introducing frustration. At lower temperatures the anisotropy associated withthe frustrated interface must increase and this necessarily means that larger fields are required to align the interface spins to the applied field. Hence increasingly large fields are required for R to decrease to a minimum value at lower temperatures. The *M-H* loops (Fig. 2(b)) show a much lower saturation field than implied from the MR data meaning that the net moment of the interface is small relative to the total moment of the Fe layers.

To explore the interaction of superconductivity with Fe/Cr, we fabricated Nb/Fe/Cr/Nb Josephson devices. The devices were fabricated from films by first defining 4-μm-wide and 30-μm-long tracks by optical lithography and Ar-ion etching. Samples were transferred to a focused ion beam microscope where current perpendicular-to-plane devices were created (See Ref. 30 for further details). Device measurements were performed in a μ-metal shielded dipstick probe in a liquid helium dewar. The current-voltage (*I-V*) curves were measured in a four-point current-biased configuration using a lock-in amplifier setup. The junction critical current (I_C) is defined as the current corresponding to the peak in the differential resistance. The normal state resistance (R_N) is determined at high bias where the slope of *I-V* is constant. Because the junction areas vary between devices, we calculate the characteristic voltage I_CR_N and use this parameter to compare different devices.

In Figure 2 we have plotted the dependence of the I_CR_N of these junctions on Cr layer thickness (d_{Cr}). The Fe layer thickness was constant for all devices (1.8 nm). A Josephson effect was confirmed by measuring the dependence of I_C on an in-plane magnetic field H (Fraunhofer patters); examples are shown Figs. 2(ii) and 2(iii). The hysteresis in $I_C(H)$ is due to magnetic flux[31] from the Fe layer and is independent of d_{Cr}.

The data in Fig. 2 shows that, although I_CR_N decays as a function of d_{Cr}, it is not a straightforward monotonic decay as expected for an S/AFM/S device. If such behaviour was due to systematic changes in R_N, the resistance-area product (AR_N) of the devices should match the trend of I_CR_N vs d_C. However, as shown in Fig. 2(i) AR_N approximately depends linearly on d_{Cr} without any correlation with the I_CR_N vs d_{Cr} data. We also note that the critical current density (I_C per unit area) has the same behaviour as I_CR_N vs d_{Cr} (data not shown). The non-monotonic behaviour of I_CR_N must, therefore, be due to an interaction of the Cooper pairs with AFM Cr.

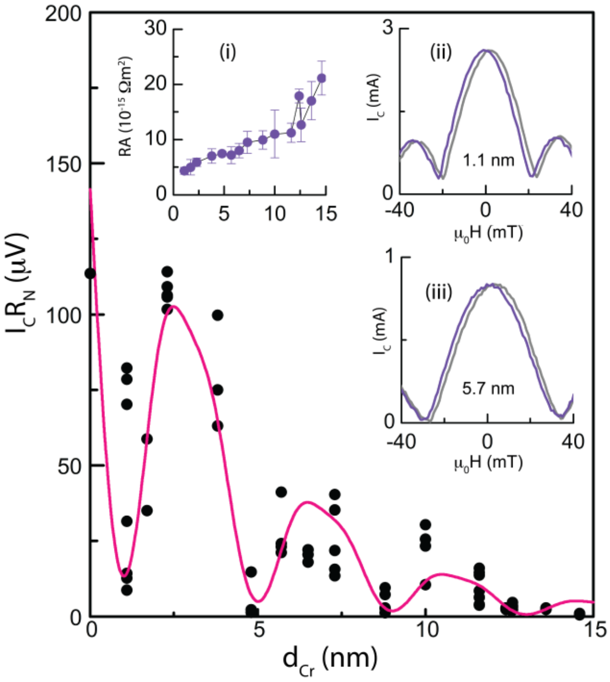

Fig. 2 I_CR_N vs d_{Cr} at 4.2 K. The data at d_{Cr}=0 is taken from Ref. 24. The solid curve is a fit to Eqn. 1. Insets: (i) AR_N vs d_{Cr} and in (ii,iii) oscillations of the Josephson critical current I_C as a function of an in-plane magnetic field H.

A non-monotonic Cr-thickness-dependence of I_CR_N is only expected if the integrated exchange field in Cr is non-zero over the pair coherence length such as in S-F-S Josephson devices.[32] If the Cr is a simple AFM such that spins between adjacent atomic planes are antiparallel then such a modulation is not be possible. However, Cr is a SDW AFM and Josephson devices with Cr show an anomalously short coherence length of ξ_{Cr} ~4 nm,[33, 34] which is likely due to an interaction between pairs and a random incommensurate SDW state.

Based on the previous studies of Josephson devices with Cr, we argue here that the most probable explanation for the non-monotonic behavior of I_CR_N in d_{Cr} is that the electron pairs interact with a SDW state in Cr, which is exchange-coupled to the Fe. To our knowledge this situation has not been microscopically modelled in the context of the superconductor proximity effect. Nevertheless, we are able to capture the form of the experimental data using a simple phenomenological model in which we assume that due to the exchange coupling of the Cr with the Fe the SDW can be described as a coherent modulation of the local magnetic moment with a fixed wavelength λ (in Cr thin films, λ is extended and can be longer than 4 nm).[35] Within the general framework of S-F-S junctions, the net supercurrent across the barrier will be dependent on the net phase difference acquired by the electron pairs as a result of the integrated exchange field in the barrier. Since the Fe is a constant thickness, it induces a constant phase difference ϕ and, during the passage through the Cr, the pairs will acquire an additional phase-shift induced by the effective exchange field associated with the SDW, which we define as $(B\lambda/2\pi)\sin(\alpha+2\pi x/\lambda)$. Here α is the phase of the SDW at the Fe/Cr interface (d_{Cr}=0) and, since on average the Fe/Cr interface is antiparallel, we set $\alpha = -\pi/2$. It can be shown that the characteristic voltage is given by adding the integral of this phase shift with respect to x to ϕ:

$$I_C R_N = A\cos\left(\phi + B\cos\left(\alpha + \frac{2\pi d_{Cr}}{\lambda}\right)\right)\exp\left(\frac{d_{Cr}}{\xi_{Cr}}\right) \quad (1)$$

The final exponential stems from averaging over diffusive trajectories.[2,35]

The values of ϕ and A can be estimated from a plot of $I_C R_N$ versus d_{Fe} for Nb/Fe/Nb junctions. In Ref. 24 we report such data (also shown in Fig. 2) and, for $d_{Fe} \sim 1.8$ nm, $I_C R_N$ is midway between a zero and a peak and so we set $\phi = 7\pi/4$. We assume a singlet coherence length in Cr of ξ_{Cr} = 4 nm, which closely matches the decay envelope of the data in Fig. 2. This value is also consistent with values obtained in Refs. 34 and 35 for Cr thicknesses where a SDW-state is assumed to form. Using the only adjustable parameter B, which corresponds to the relative exchange field of the SDW, a fit using the above equation agrees very well with the experimental data.

It is important to emphasize that in order to describe the complex behaviour of I_C on d_{Cr} our model assumes an effective interaction between the superconducting pairs induced in the Cr and the SDW [proportional to B in Eq. (1)]. This interaction breaks the time-reversal symmetry of the Cooper pairs and therefore necessarily implies the presence of triplet pair correlations as in conventional S-F-S junctions.[1] Furthermore, the frustrated spins at the Fe/Cr interfaces (Fig. 1) should theoretically serve as additional spin-mixers.

If triplet pair correlations are present in devices with Cr/Fe barriers then, in a device with a symmetrical barrier such as Cr/Fe/Cr, it is our hypothesis that spin-one triplets may also form (this is consistent with Ref. 3). To test this hypothesis, we fabricated Josephson devices with Cr/Fe/Cr barriers in which the Fe thickness d_{Fe} was systematically varied. In Fig. 3(a) we have plotted $I_C R_N$ for a series of junctions as a function of d_{Fe}. Each Cr layer has a thickness of 2.5 nm – this thickness was chosen since at this thickness we obtained the maximum value of $I_C R_N$ in Fig. 2.

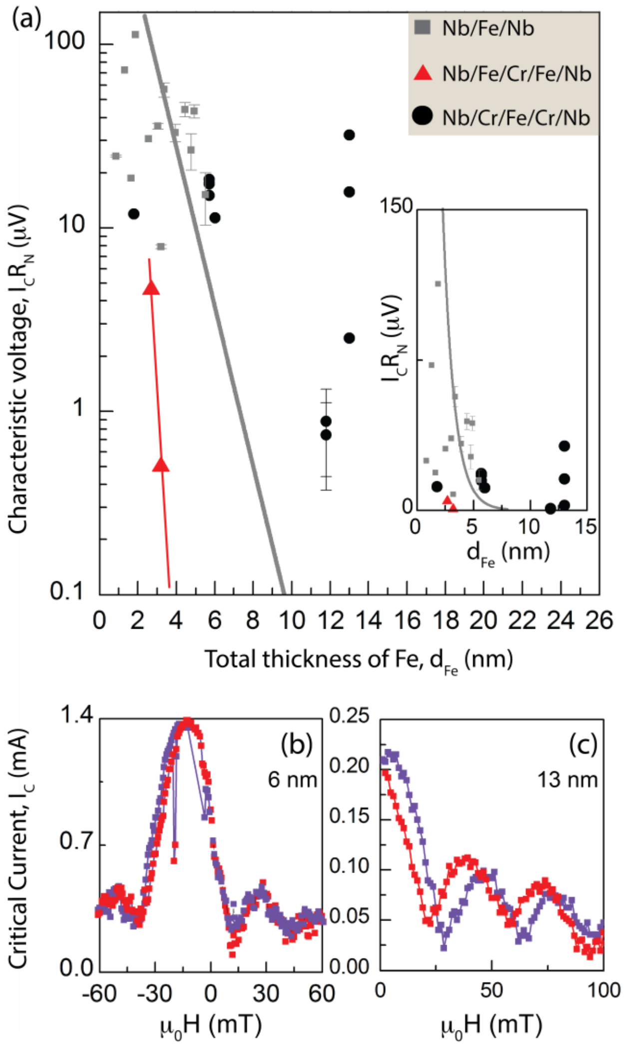

Fig. 3 (a) $I_C R_N$ dependence on d_{Fe} at 4.2 K for different barrier configurations: Cr(2.5)/Fe/Cr(2.5) (●); Fe only (■) with an exponential decay (greyline) assuming a coherence length of 3 nm, taken from Ref. 24; and Fe(1.35)/Cr(2.5)/Fe(1.35) barriers (▲) (estimated from Ref. 36) with parallel aligned Fe layers (the red curve is a guide to the eye). (b,c) Fraunhofer Oscillations of I_C with an in-plane magnetic field for two devices with different Fe layer thickness (labelled) – hysteresis in I_C(H) is due to barrier flux with varies depending on junction dimensions and d_{Fe}.

Although there is a lot of scatter in $I_C R_N$ vs d_{Fe}, the pair coherence length in Fe for some devices is much longer than the known coherence length for singlet pairs in Fe of only 1 nm.[24] This is clear if we compare the Nb/Cr/Fe/Cr/Nb data to values of $I_C R_N$ obtained from Nb/Fe/Cr/Fe/Nb devices[36] which also include the additional decay due to the presence of Cr and Fe/Cr interfaces (reproduced in Fig. 3(a)). For all Fe layer thicknesses investigated, the magnitude of $I_C R_N$ of the Nb/Cr/Fe/Cr/Nb devices is significantly larger than values obtained for Nb/Fe/Cr/Fe/Nb devices. For comparison, we also plot $I_C R_N$ data obtained Nb/Fe/Nb devices,[24] which do not include Cr or Fe/Cr interface.

The fact that we observe enhanced supercurrents in Nb/Cr/Fe/Cr/Nb is compelling evidence for the generation of spin-triplet correlations in Cr/Fe/Cr barriers. The scatter in $I_C R_N$ vs d_{Fe} is interesting as it may indicate that the generation of spin-one triplets is dependent on differences between the Cr layers on either side of the Fe layer. In Ref. 3 it is theoretically shown that the generation of spin-one triplets in devices with F’/F/F’ barriers is strongly dependent on the symmetry of the two F’ layers – while the F’/F interfaces need to be non-collinear, the two F’ layers should be collinear in order to maximise the generation of triplet pairs. In our Cr/Fe/Cr devices, the orientation of the spins in one Cr layer are unlikely to match the orientation of the spins in the second Cr layer and so the efficiency of triplet pair generation is likely to vary between devices.

On the basis of the data in Figs. 2 and 3 and our phenomenological analysis, we suggest that spin-zero and spin-one triplets are generated due to the SDW state in Cr and frustration at the Fe/Cr interfaces. This necessarily implies a novel method of triplet pair generation. Spectroscopic measurements, which can probe the local density of states in Fe/Cr in the superconducting state should now be performed in order to understand better the ‘nature’ of the triplet state. Scanning tunnelling spectroscopy on S/half-metallic junctions has revealed features such zero-bias conductance peaks which are associated with an odd frequency spin-triplet state (see, e.g., Ref. 37) while Andreev spectroscopy on similar structures has shown evidence for equal-spin Andreev reflection. Similar experiments on Nb/Cr/Fe structures could also reveal similar results.

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