Coupling of two superconductors through a ferromagnet: evidence for a \( \pi \)-junction

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Abstract

We report measurements of the temperature dependence of the critical current in Josephson junctions consisting of conventional superconducting banks of Nb and a weakly ferromagnetic interlayer of a Cu\(_x\)Ni\(_{1-x}\) alloy, with \( x \) around 0.5. With decreasing temperature \( I_c \) generally increases, but for specific thicknesses of the ferromagnetic interlayer, a maximum is found followed by a strong decrease down to zero, after which \( I_c \) rises again. Such a sharp cusp can only be explained by assuming that the junction changes from a 0-phase state at high temperatures to a \( \pi \)-phase state at low temperatures.

Almost all of the presently known superconductors contain conventional Cooper pairs, two electrons with opposite spin and momentum \((+k_\uparrow,-k_\downarrow)\). Such a system is described by an isotropic excitation gap and / or order parameter. The exceptions are found notably in high \( T_c \) (oxide) superconductors, and in some Heavy Fermion systems, in which cases the exact pairing mechanism is not yet fully understood. Still, it is surprising that of the many possible ways to form a pair, so few are actually realized. For instance, it is not imperative that the net momentum of the pair is zero. It was predicted long ago by Larkin and Ovchinnikov \cite{1} and by Fulde and Ferrel \cite{2} that pairing still can occur when the electron energies and momenta at the Fermi energy are different for the two spin directions, for instance as the result of an exchange field in magnetic superconductors. The resulting 'LOFF'-state is qualitatively different from the zero-momentum state: it is spatially inhomogeneous and the order parameter contains nodes where the phase changes by \( \pi \). The LOFF state was never observed in bulk material, but below we present evidence that it can be induced in a weak ferromagnet (F) sandwiched between two superconductors (S). Such an SFS junction can yield a phase shift of \( \pi \) between the superconducting banks, as was also predicted \cite{3, 4, 5}. The \( \pi \)-state offers fundamentally new ways for studying the coexistence of superconductivity and magnetism and may also be important for superconducting electronics, in particular in quantum computing: several schemes for the realization of the necessary qubits (quantum two level systems) rely on the use of phase shifts of \( \pi \) in a superconducting network \cite{6, 7}. The spatial variation of the superconducting order parameter in the ferromagnet arises as a response of the Cooper pair to the energy difference between the two spin directions in ferromagnet. The electron with the energetically favorable spin increases its momentum by \( Q \propto E_{ex}/v_F \), where \( E_{ex} \) is the exchange energy and \( v_F \) is the Fermi velocity, while the other electron decreases...
its momentum by the same amount. Since the original momentum of each electron can be positive or negative, the total pair momentum inside the ferromagnet is $2Q$ or $-2Q$. Combination of the two possibilities leads to a spatially oscillating superconducting order parameter, $\psi(z)$, in the SFS junction along the direction normal to the SF interfaces: $\psi(z) \propto \cos(2Qz)$. The same picture applies in the diffusive limit. In this case, the oscillation is superimposed on the decay of the order parameter due to pair breaking by impurities in presence of the exchange field. In the regime $E_{ex} \gg kBT$, the decay length $\xi_{F1}$ is given by the usual expression $(hD/E_{ex})^{1/2}$, where $D$ is the electron diffusion coefficient in the ferromagnet, while the oscillation period $2\pi\xi_{F2}$ is equal to $2\pi(hD/E_{ex})^{1/2}$. Due to the oscillations, different signs of the order parameter parameter can occur at the two banks of the SFS junction when the F-layer thickness $d_F$ is of the order of half a period. This is the so-called $\pi$-phase state, which competes for existence with the ordinary 0-phase state. Fig. 1a shows the result of a Ginzburg-Landau free-energy calculation consisting of negative condensation energy and positive gradient energy for either state in the F-layer. One can see that the $\pi$-phase is more favorable in the range $d_F/(2\pi\xi_{F2})$ between 0.4 and 0.8. Fig. 1b shows the behaviour of $\psi(z)$ in the F-layer below and above $d_{F,cr}$ calculated using the formalism of ref. 3. The crossover from the 0-phase to the $\pi$-phase state should manifest itself in an anomalous thickness dependence both of the superconducting transition should manifest itself in an anomalous thickness crossover from the 0-phase to the calculated using the formalism of ref. [5]. The systems (no coupling) especially was shown that also in bilayer Pb/Fe [15] but the results are not conclusive. The approach we choose is to induce the crossover as function of temperature, not of thickness, and to use a unique signature of the junction $I_c$ according to the Josephson relation $I_c = I_{0}\sin(\phi)$, with $\phi$ the phase difference across the junction, biasing the junction at $\phi = \pi$ should lead to a negative current response upon a small increase of the phase. In other words, $I_c$ becomes negative. A change of state from 0 to $\pi$ will lead to a zero-crossing of $I_c$, and even if only the absolute value of the current is measured, a sharp cusp will be observed. The condition for having the temperature as parameter is $kBT \approx E_{ex}$. The exchange field and the temperature then are equally important and the behaviour of the order parameter should be written as

$$\psi(z) \propto e^{-z/\xi_F} \propto e^{-z/\xi_{F1}} e^{-iz/\xi_{F2}},$$  \hspace{1cm} (1)

with $\xi_F$ given by

$$\xi_F = \sqrt{\frac{hD}{2(\pi kBT + iE_{ex})}},$$  \hspace{1cm} (2)

which yields for $\xi_{F1}$ and $\xi_{F2}$:

$$\xi_{F1,2} = \sqrt{\frac{hD}{(E_{ex}^2 + (\pi kBT)^2)^{1/2} \pm kBT}}.$$  \hspace{1cm} (3)

Note that this reverts to $\xi_{F1} = \xi_{F2}$ for $E_{ex} \gg kBT$ as discussed above. This is the case encountered with classical ferromagnets (Fe, Co, Ni), where $E_{ex}$ is of the order of 1 eV and much larger than the critical temperature $T_c$ of conventional superconductors. In the case $kBT \approx E_{ex}$ the decay length $\xi_{F1}$ increases with decreasing temperature whereas $\xi_{F2}$ decreases (see Eq. 2). This is how varying the temperature provides the possibility to cross from a 0-phase to a $\pi$-phase state [16]. Moreover, a small value for $E_{ex}$ ensures a large decay length $\xi_{F1}$, making it possible to fabricate Josephson SFS sandwiches with homogeneous and continuous ferromagnetic interlayers. Thus, the basic task is to find and prepare such weak ferromagnets.

The junctions we studied consisted of superconducting Nb (S) banks with an interlayer of a ferromagnetic Cu$_{1-x}$Ni$_x$ alloy (F). The onset of ferromagnetism in these alloys is around $x = 0.44$; above this concentration the Ni magnetic moment increases with about 0.01 $\mu_B$/at.% Ni, which allows precise tuning of the magnetism. An insulating SiO-layer was used between the top electrode and the bottom SF sandwich. The window in this layer determined the junction area of 50x50 $\mu$m$^2$. A schematic sample cross-section is given in Fig. 2 (upper panel). Because of the low junction resistance $R_n \approx 10^{-5}\Omega$ the
transverse transport characteristics were measured by a SQUID picovoltmeter with a sensitivity of $10^{-11}$ V in the temperature range of 2.2 K to 9 K. Junctions were fabricated with $x$ between 0.40 and 0.57. Upon crossing from the paramagnetic to ferromagnetic regime the junction critical currents dropped sharply but the $I-V$ characteristics and magnetic field dependence $I_c(H)$ were still similar to those for standard SNS junctions (N is a normal metal). In Fig. 2 (middle panel) $I-V$ data are shown for a junction with $x = 0.5$, $d_F = 14$ nm at a temperature of 4.2 K. The voltage onset at $I_c$ is sharp and well defined. Fig. 2 (lower panel) shows that $I_c(H)$ for this junction yields the classical ‘Fraunhofer’ pattern. Note that the central peak is at zero field, even though the alloy is ferromagnetic. This is due to the fact that the net magnetization of the sample is zero after a careful cooldown, resulting in a small-scale magnetic domain structure in the F-layer and zero average phase change over the junction. Our central result was obtained for junctions with Cu$_{0.48}$Ni$_{0.52}$ alloys. At this concentration the ferromagnetic transition temperature $T_{Curie}$ is about 20 K to 30 K. SQUID magnetometry for single thin films of thickness in the range 20 nm to 100 nm showed a small hysteresis loop close to 10 K with a coercive field of about 8 mT and a saturation moment of 0.07 $\mu_B$/Ni at. Fig. 3 shows $I_c(T)$ in zero magnetic field for two junctions with $d_F = 22$ nm [18]. The curve marked a) shows that $I_c$ increases with decreasing temperature, goes through a maximum, returns to zero, and rises again sharply. For all data points, it was ascertained that the zero-field value was the maximum value for $I_c$. The curve marked b) shows the same characteristic behaviour although the zero value for $I_c$ lies at a different temperature. In this case $I_c(H)$ characteristics were measured at three different temperatures to ascertain that $I_c$ was determined correctly. The data, shown in the inset of Fig.3, prove that the $I_c(T)$ oscillations are not associated with residual magnetic inductance changes. The sharp cusp in $I_c(T)$ can be explained only by the transition from a 0-phase state to a $\pi$-phase state. This can also be demonstrated by the thickness dependence of the effect. Shown in Fig. 4a are a series of measurements for junctions of different thicknesses in the range 23 nm to 27 nm. At 23 nm only positive curvature is visible, an inflection point is observed for 25 nm, a maximum for 26 nm, and the full cusp now at 27 nm. Fig. 4b shows a set of calculations based on the formalism of the quasiclassical Usadel equations [13], with reasonable parameters for $E_{ex}$ and $d_F/\xi_F$, where $\xi_F = (\hbar D/(2\pi k_B T))^{1/2}$, demonstrating how the crossover moves into the measurement window upon increasing the F-layer thickness.

A final remark concerns qualitative and quantitative reproducibility. Qualitatively, the cusps can be observed for certain thickness intervals in all sample batches with ferromagnetic layers which are presently fabricated, both for concentrations of 52 at.% Ni (with $T_{Curie}$ about 20 K - 30 K) and 57 at.% Ni (where $T_{Curie}$ is around 100 K). Quantitatively, there are still variations in the values of thickness interval and temperature, as well as in the magnitude of the critical current for different batches even with the same nominal F-layer content. In both of these respects typical batch-to-batch variation is demonstrated in the differences between Figs. 2 and 3. We believe this is due to small variations in the magnetic properties of the F-layers. In single films, $T_{Curie}$ shows a spread of about 10 K; the weak magnetism is apparently sensitive to the details of the preparation procedure.

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Figure 1: (a) Calculations of the Ginzburg-Landau (GL) free energy in the F-layer for the 0- and π-phase states. (b) The spatial distribution of the order parameter in the F-layer of the SFS junction calculated for various ratios of \( d_F/(2\pi\xi_{F2}) \): for \( d_F/(2\pi\xi_{F2}) = 1/(2\pi) \) and 1 the lowest energy corresponds to the 0-phase, while for \( d_F/(2\pi\xi_{F2}) = 1/2 \) and 3/2 the π-phase is energetically favorable. Shown for comparison is the 0-phase for \( d_F/(2\pi\xi_{F2}) = 1/2 \) (dotted line), which has higher energy than the π-phase.

Figure 2: (Upper) Schematic cross-section of the sample. (Middle) Typical I-V characteristic. (Lower) Magnetic field dependence of the critical current \( I_c \) for the junction with Cu0.5Ni0.5 and \( d_F = 14 \) nm.
Figure 3: Critical current $I_c$ as a function of temperature $T$ for two junctions with $Cu_{0.48}Ni_{0.52}$ and $d_F = 22$ nm [17]. The inset shows the dependence of $I_c$ on magnetic field $H$ for the temperatures around the crossover to the $\pi$-state as indicated on curve b: (1) $T = 4.19$ K, (2) $T = 3.45$ K, (3) $T = 2.61$ K.

Figure 4: Left: critical current $I_c$ as function of temperature for $Cu_{0.48}Ni_{0.52}$ junctions with different F-layer thicknesses between 23 nm and 27 nm as indicated. Right: model calculations of the temperature dependence of the critical current in an SFS junction for $E_{ex} = 0.9\pi T_c$ and various ratios of $d_F/\xi^*$, where $\xi^* = \sqrt{\hbar D/(2\pi k_B T_c)}$. 