Coupling of Two Superconductors through a Ferromagnet: Evidence for a π Junction

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We report measurements of the temperature dependence of the critical current, I_c, in Josephson junctions consisting of conventional superconducting banks of Nb and a weakly ferromagnetic interlayer of a Cu_xNi_{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 be explained only by assuming that the junction changes from a 0-phase state at high temperatures to a π phase state at low temperatures.

Almost all of the presently known superconductors contain conventional Cooper pairs, two electrons with opposite spin and momentum ($+\mathbf{k}\uparrow$, $-\mathbf{k}\downarrow$). Such a system is described by an isotropic excitation gap or an order parameter. The exceptions are found notably in high T_c superconductors, and in some organic superconductors and heavy fermion systems, in which the exact pairing mechanism is not yet fully understood. Still, it is surprising that of 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 [1] and by Fulde and Ferrel [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 "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 π. It was never observed in bulk materials, but below we present evidence that it can be induced in a weak ferromagnet (F) sandwiched between two superconductors (S). Such a SFS junction can yield a phase shift of π between the superconducting banks, as was also predicted [3–5]. The π state offers new ways for studying the coexistence of superconductivity and magnetism and may also be important for superconducting electronics, e.g., in quantum computing: several schemes for the necessary qubits (quantum two-level systems) rely on phase shifts of π in a superconducting network [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. 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 Q. 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 an oscillating order parameter $\psi(z)$ in the junction along the direction normal to the SF interfaces: $\psi(z) \propto \cos(2Qz)$ [8,9]. The same picture applies in the diffusive limit. Now the oscillation is superimposed on the decay of the order parameter due to pair breaking by impurities in the presence of the exchange field. In the regime $E_{ex} \gg k_BT$, the decay length ξ_{F1} is given by $(\hbar D/E_{ex})^{1/2}$, where D is the electron diffusion coefficient in the ferromagnet, while the oscillation period $2\pi\xi_{F2}$ is equal to $2\pi(\hbar D/E_{ex})^{1/2}$. Because of the oscillations, different signs of the order parameter can occur at the two banks when the F-layer thickness d_F is of the order of half a period. This is the so-called π-phase state, which competes for existence with the ordinary 0-phase state. Figure 1a shows a Ginzburg-Landau free-energy calculation consisting of negative condensation energy and positive gradient energy for either state in the F layer. The π phase is more favorable in the range $d_F/(2\pi\xi_{F2})$ between 0.4 and 0.8. Figure 1b shows the behavior of $\psi(z)$ in the F layer below and above $d_{F,cr}$. The crossover from the 0 phase to the π phase state should manifest itself in an anomalous thickness dependence both of the superconducting transition temperature T_c of the junction [10,11] and of the critical current I_c [4]. Experiments on $T_c(d_F)$ have been performed in systems such as Nb/Gd [12], Nb/Fe [13], V/Fe [14], and Pb/Fe [15] but the results are not conclusive. Especially, it was shown that also in bilayer systems (no coupling) $T_c(d_F)$ can behave in an anomalous fashion [15].

Our approach is to induce the crossover as a function of temperature, not of thickness, and to use a unique signature of the junction I_c: according to the Josephson relation $I_s = I_c \sin(\phi)$, with ϕ the phase difference across the junction, biasing with $\phi = \pi$ 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 π will lead to a zero crossing of I_c, and if only the absolute value of the current is measured, a sharp cusp will be observed. The condition for having the temperature as a parameter is $k_BT \approx E_{ex}$. The exchange field and the temperature then are equally important and the behavior of
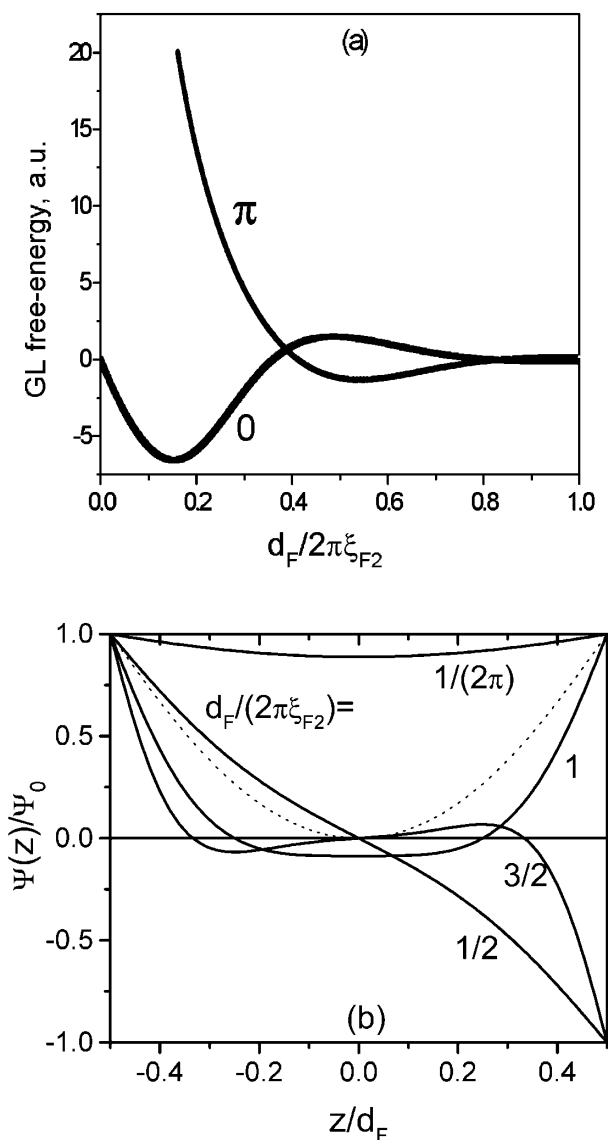
FIG. 1. (a) Calculations of the Ginzburg-Landau (GL) free energy in the F layer for the 0- and π-phase states. (b) The spatial distribution of the order parameter in the F layer of the SFS junction calculated for various ratios of $d_F/(2\pi\xi_{F2})$: for $d_F/(2\pi\xi_{F2}) = 1/(2\pi)$ and 1 the lowest 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.

the order parameter should be written as

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

with ξ_F given by

$$\xi_F = \sqrt{\frac{\hbar D}{2(\pi k_B T + iE_{ex})}}, \qquad (2)$$

which yields for ξ_{F1} and ξ_{F2}:

$$\xi_{F1,2} = \sqrt{\frac{\hbar D}{[E_{ex}^2 + (\pi k_B T)^2]^{1/2} \pm k_B T}}. \qquad (3)$$

This reverts to $\xi_{F1} = \xi_{F2}$ for $E_{ex} \gg k_B T$ as discussed above, and encountered with classical ferromagnets (Fe, Co, Ni) with E_{ex} of the order of 1 eV. In the case $k_B T \approx E_{ex}$ the decay length ξ_{F1} increases with decreasing temperature whereas ξ_{F2} decreases. This is how varying the temperature provides the possibility to cross from a 0-phase to a π-phase state [16]. Moreover, a small value for E_{ex} ensures a large decay length ξ_{F1}, making Josephson SFS sandwiches with homogeneous and continuous ferromagnetic interlayers possible.

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 is around $x = 0.44$; above this concentration the Ni magnetic moment increases with about 0.01 μ_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 50×50 μm^2. A schematic sample cross section is given in Fig. 2 (upper panel). Because of the low junction resistance $R_n \approx 10^{-5}$ Ω the transverse transport characteristics were measured by a SQUID picovoltmeter with a sensitivity of 10^{-11} V in the temperature range of 1.2 to 9 K. Junctions were fabricated with x between 0.40 and 0.57. Upon crossing to the ferromagnetic regime the junction critical currents dropped sharply but the I-V characteristics and magnetic field dependence $I_c(H)$ (H in the plane of the junction) 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. Figure 2 (lower panel) shows that $I_c(H)$ for this junction yields the classical "Fraunhofer" pattern. The oscillation period is in reasonable agreement with the cross section of the junction. Note that the central peak is at zero field, even though the alloy is ferromagnetic. This signifies that on average there is no change in the phase difference over the junction along the different directions in the plane of the junction, presumably due to a small-scale magnetic domain structure of the magnetic layer with zero net magnetization. The peak was found shifted when the sample was heated above T_c (but below the ferromagnetic transition temperature, T_{Curie}) and a small field briefly applied, leading to a finite magnetization. Sometimes the peak was found shifted in zero applied field, probably due to flux trapping in the superconducting banks during cooling down. This could be remedied by reheating and recooling. The starting point for all measurements was a central peak at zero field.

Our main result was obtained for junctions with $Cu_{0.48}Ni_{0.52}$ alloys. At this concentration T_{Curie} is about 20 to 30 K. The resistivity of such alloys at 10 K is of the order of 50 $\mu\Omega$ cm, indicating a mean free path of about 1 nm. The magnetization of the films is in-plane. SQUID magnetometry at 10 K on single alloy films of thickness 20 to 100 nm, and on Nb/alloy/Nb trilayers with similar
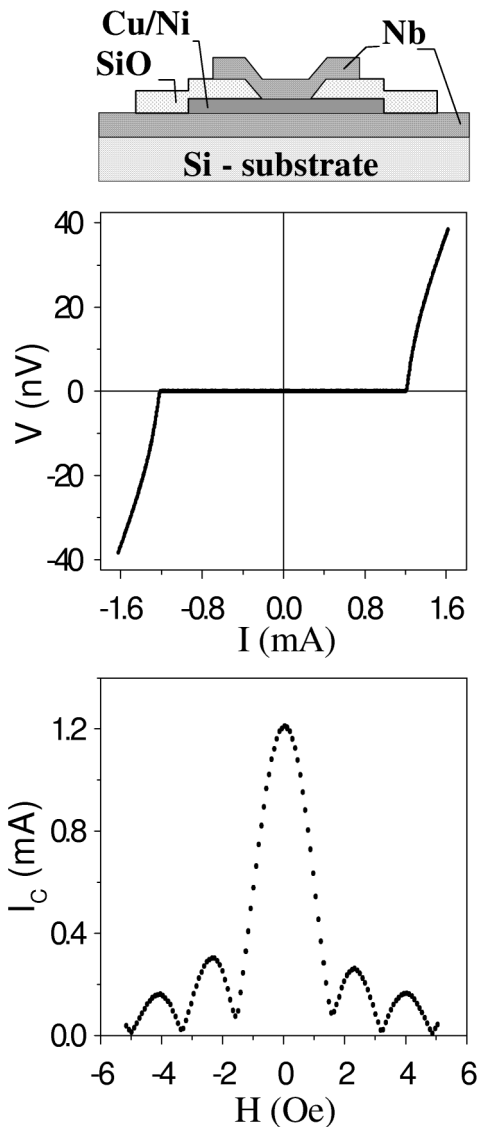
FIG. 2. (Top) Schematic cross section of the sample. (Center) Typical I-V characteristic. (Bottom) Magnetic field dependence of the critical current I_c for the junction with $Cu_{0.5}Ni_{0.5}$ and $d_F = 14$ nm.

FIG. 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]. Inset: I_c versus magnetic field H for the temperatures around the crossover to the π state as indicated on curve b: (1) $T = 4.19$ K, (2) $T = 3.45$ K, (3) $T = 2.61$ K.

alloy thickness, showed a small hysteresis loop with a coercive field of about 8 mT and a saturation moment of $0.07\mu_B/Ni$ atom. We found no significant difference between single layers and trilayers, from which we conclude that also in the junctions the alloy layer is ferromagnetic and that a supercurrent can be sustained even through a ferromagnetic layer. Figure 3 shows $I_c(T)$ in zero magnetic field for two junctions with $d_F = 22$ nm [17]. 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 behavior 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
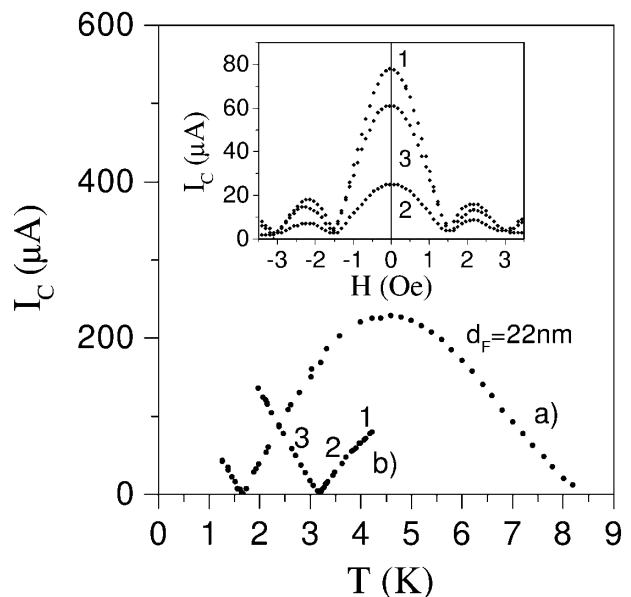
was determined correctly. The data, shown in the inset of Fig. 3, prove that the $I_c(T)$ oscillations are not associated with residual magnetic inductance changes which would change the position of the central peak. It is important to realize that the phase difference in zero applied field is uniform in the plane of the junction, either 0 or π. The Fraunhofer pattern *will not shift* when the phase turns from 0 to π, but the zero-field I_c goes from positive to negative. In a current-driven experiment, this leads to the sharp cusp observed in $I_c(T)$. The π state can also be demonstrated by the thickness dependence of the effect. Shown in Fig. 4a is a series of measurements for junctions of different thicknesses in the range 23 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. Figure 4b shows a set of calculations based on the formalism of the quasiclassical Usadel equations [18], with reasonable parameters for E_{ex} and d_F/ξ^*, where $\xi^* = [\hbar D/(2\pi k_B T_c)]^{1/2}$. They qualitatively demonstrate how the crossover moves into the measurement window upon increasing the F-layer thickness. Quantitatively, the thickness dependence in the calculations is much weaker than in the experiments. Parameters such as the spin flip scattering length probably also play a role. Still, the appearance of the crossover is mimicked correctly. If we estimate it around $d_F/2\pi\xi_{F2} \approx 0.4-0.5$, it follows that $\xi_{F2} \approx 10$ nm, as expected for the low magnetic moment and justifying the assumption of dirty limit conditions.

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
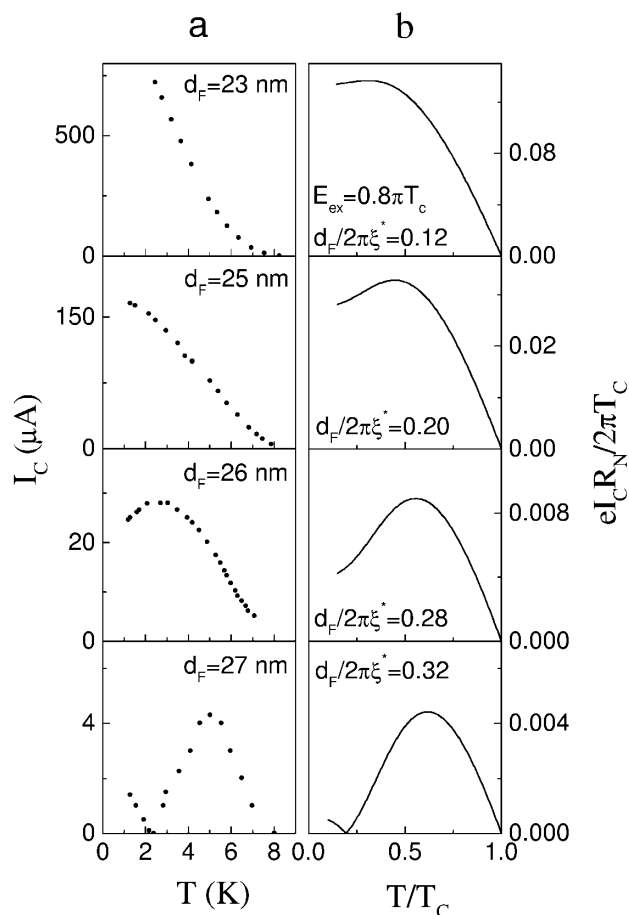
FIG. 4. (a) Critical current I_c as a function of temperature for $Cu_{0.48}Ni_{0.52}$ junctions with different F-layer thicknesses between 23 and 27 nm as indicated. (b) Model calculations of the temperature dependence of the critical current in a SFS junction for $E_{ex} = 0.8\pi T_c$ and various ratios of $d_F/2\pi\xi^*$, where $\xi^* = \sqrt{\hbar D/(2\pi k_B T_c)}$.

concentrations of 52 at. % Ni (with T_{Curie} about 20–30 K) and 57 at. % Ni (with T_{Curie} around 100 K). Moreover, for higher Ni concentration the crossovers are at lower thickness, reflecting the decrease in $\xi_{F1,F2}$. Quantitatively, there are still variations in the values of thickness interval and crossover temperatures, and in the magnitude of the critical current for different batches, even with the same nominal F-layer content. Typical batch-to-batch variations are demonstrated in the differences between Figs. 3 and 4. 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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