Oscillations of the superconducting order parameter in a ferromagnet

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Planar tunneling spectroscopy reveals damped oscillations of the superconducting order parameter induced into a ferromagnetic thin film by the proximity effect. The oscillations are due to the finite momentum transfer provided to Cooper pairs by the splitting of the spin up and down bands in the ferromagnet. As a consequence, for negative values of the superconducting order parameter the tunneling spectra are capped ("\( \pi \)-state"). The oscillations' damping and period are set by the same length scale, which depends on the spin polarization.

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The quantum character of superconductivity arises from the existence of phase coherence in the electron condensate. In conventional superconductors, where pairing is provided by the exchange of virtual phonons, the phase is a constant. On the other hand, phase sensitive experiments in high temperature superconductors have shown that the wavefunction of Cooper pairs with perpendicular quasiparticle momenta presents a \( \pi \)-phase shift suggesting unconventional pairing. Here, we show that a \( \pi \)-phase shift can also occur in the order parameter of conventional superconductors when superconducting correlations coexist with ferromagnetic order.

More than 30 years ago, Fulde and Ferrel \cite{FuldeFerrel}, and Larkin and Ovchinnikov \cite{LarkinOvchinnikov} (FFLO), showed independently that the superconducting order parameter may be modulated in real space by an exchange field. A Cooper pair, in the singlet state, acquires a finite momentum \( Q = 2E_{ex}/\hbar v_F \), where \( 2E_{ex} \) is the exchange energy corresponding to the difference in energy between the spin-up and spin-down bands, and \( v_F \) the Fermi velocity. The superconducting phase grows linearly with the spatial coordinate \( x \), \( \varphi = Q \cdot x \) and a \( \pi \)-phase shift was expected for translations of \( \Delta x \approx \hbar v_F / 4E_{ex} \).

Unlike high temperature superconductors where \( \varphi \) is a \( 2\pi \)-multiple of 0 and \( \pi \), in the FFLO-state \( \varphi \) varies continuously.

The FFLO-state only occupies a tiny part of the superconducting phase diagram close to the normal state \( \xi \text{F} \). The fragility of singlet superconductivity in a finite exchange field that removes the degeneracy of the ground state with respect to the spin degrees of freedom makes its experimental evidence still an open question. In homogeneous superconductors the normal state is recovered when \( E_{ex} > \sqrt{2}/2\Delta_s \) (Clogston criterion) \( \xi \text{F} \), where \( \Delta_s \) is the superconducting energy gap. The situation is more favorable if Cooper pairs are injected from a superconductor into a ferromagnet \( F \) by the proximity effect. Assuming that the superconductor is weakly affected by the exchange field, superconducting correlations persist in \( F \) even for exchange energies much higher than \( \Delta_s \). The physical reason is that Cooper pairs are not instantaneously broken when they penetrate into the ferromagnet. They survive for a time corresponding to a traveled length on the order of \( \xi_F = \hbar v_F / 2E_{ex} = 1/Q \), the coherence length scale in \( F \), \( \xi \text{F} \), which is independent on the energy gap. The breakdown of the Clogston criterion turns out to be very significant since \( E_{ex} \) is typically at least two orders of magnitude larger than \( \Delta_s \).

When a Cooper pair moves into a ferromagnet, the phase shift produces oscillations of the real part of the superconducting order parameter on a length scale given by \( \xi_F \), as shown in Fig. 1a \text{[6]}. However this artificially generated FFLO-state vanishes on the same length scale, which is typically of the order of a few nm. Unlike bulk superconductors where the gap equation allows the FFLO-state to occur only for exchange fields close to the critical field, this state exists for any exchange energy. Furthermore, the oscillating behavior of the order parameter in \( F \) is only due to the phase factor, while the number of Cooper pairs decays monotonically. We shall call the states corresponding to a positive sign of the real part of the order parameter the "\( 0 \)-state" and those corresponding to a negative sign the order parameter the "\( \pi \)-state".

An induced superconducting order parameter in \( F \) modifies the quasiparticle density of state (DOS). In the "\( \pi \)-state", i.e. when the thickness of the ferromagnet is larger than \( \xi_F \), the features in the superconducting DOS are reversed with respect to the normal state (see inset a of Fig. 1a). This can be explained considering the microscopic mechanism that allows superconducting correlations to propagate into \( F \), i.e., Andreev reflections \( \xi \text{F} \). The process is illustrated in Fig. 2b using the energy-momentum dispersion law of the normal metal: an incoming electron in a normal metal \( N \) with energy lower than \( \Delta_s \) from the Fermi level, is reflected into a hole at the S/N interface. The incoming electron and the outgoing hole accumulate a phase difference \( \varphi = \Delta p \cdot x \) depending on their traveled distance, \( x \), and on the difference between their momenta, \( \Delta p \). Note that \( \Delta p \) is a function of the quasiparticle energy. If the normal layer is very thin, the phase difference is small and, roughly speaking, the DOS in \( N \) is close to that of the Cooper pair reservoir. The situation is strongly modified if the normal metal is ferromagnetic. As Andreev reflections invert
spin-up into spin-down quasiparticles and vice-versa, the total momentum difference includes the spin-splitting of the conduction band: \( \Delta p_F = \Delta p + Q \) (see Fig. 1). If the exchange energy is much larger than the energy gap, which is usually the case, \( \Delta p_F \approx Q \) and the phase difference between the electron and hole wavefunction is almost energy independent. The DOS is modified in a thin layer on the order of \( \xi_F \). In particular, the interference between the electron and hole wavefunction produces an oscillating term in the superconducting DOS with period \( \approx x E_{xx}/h v_F \). A phase-induced oscillating term in the superconducting DOS is a natural consequence of the proximity effect. It has already been observed in the past either in the clean or the dirty limits. However, oscillations usually appear as a function of energy since, in a normal metal, the phase is energy dependent. Differently, in a ferromagnet the oscillations turn the overall energy-dependent DOS up side down with respect to the normal state.

We measure the superconducting DOS by tunneling spectroscopy. The conductance vs. bias of a tunnel junction between a superconductor and a normal metal probes the energy-dependent quasi-particle DOS of the superconductor, convoluted by the thermal broadening. The DOS is obtained after normalizing the spectra by the background conductance measured when both electrodes are in the normal state. Planar junctions provide unsurpassed energy resolution and even more importantly, in our case, large magnitude resolution. We fabricated planar junctions completely "in-situ" by thin film deposition in a typical base pressure of \( 10^{-9} \) Torr. The junction area \( 100 \mu m \times 100 \mu m \) is defined by evaporating 500 Å of insulator (SiO) through shadow masks. A thin layer of PdNi (hereafter called PdNi) backed by 500 Å of Nb (\( T_c = 8.8 \) K) is deposited just after oxidation defining a four-terminal cross-junction geometry. The Nb and the PdNi respectively provide the Cooper pair reservoir and the ferromagnetic thin film. The mean free path in the PdNi thin films is limited by surface scattering. The Ni concentration is kept on the order of 10 % and measured after fabrication by Rutherford Back scattering Spectrometry (RBS). The junction resistance is typically between 500 and 1kΩ while the interface resistance between PdNi and Nb is \( \approx 10^{-5} \)Ω. The bias-dependence of the tunneling conductance is measured using a standard AC-modulation technique. Combining Lock-in detection with an ultra-low noise DC/AC mixer, we can directly resolve structures in the DOS as small as \( 10^{-4} \) of the background conductance.

Ferromagnetic order in PdNi alloys results from indirect exchange between the Ni magnetic moments provided by the large spin susceptibility of Pd Ni. At low Ni concentrations, the total magnetic moment is mainly due to the spin polarized electrons of the host at the Fermi level. Long-range itinerant ferromagnetism provides an almost ideal system where Cooper pairs are suddenly polarized when they enter into the ferromagnet. The main advantage of using a ferromagnetic alloy, instead of pure Ni, for instance, is that the exchange energy can be kept suitably small. \( E_{xx} \) may be estimated from the magnetization \( M \approx \mu_B \chi E_{ss} \), where \( \mu_B \) is the Bohr magneton and \( \chi \) the host susceptibility. In PdNi alloys with 10 % of Ni, \( E_{xx} \) is of the order of 10meV resulting in \( \xi_F \approx 50 \) Å, which corresponds to an order of magnitude increase with respect to pure ferromagnetic elements such as Fe, Ni or Co. This coherence length is accessible to standard thin film technology. Of course, decreasing the Ni concentration closer to the paramagnetic-ferromagnetic transition would further increase the penetration length of Cooper pair into the ferromagnet. However, we observed that lowering the Ni concentration results in a reduced magnetic homogeneity.

In Fig. 3 the superconducting DOS at \( T = 300 \) mK is presented for two different thickness of PdNi. The Al counter-electrode is driven into the normal state by applying a magnetic field of 100 Gauss perpendicular to the film. For the thinner ferromagnetic layer (50Å) the phase factor is positive ("0-state") and the DOS displays a maximum at the Nb gap edge and a minimum at the Fermi level set to zero in our spectra. As a result of the finite interface resistance between PdNi and Nb, the pair amplitude is small, corresponding to a few per cent difference from the background conductance. To stress that in our geometry the relevant energy scale for the proximity effect is the Nb gap energy \( \Delta_{Nb} = 1.40 \) meV, the DOS of Nb measured in a junction without PdNi is also plotted on the r.h.s. of Fig. 2a. Increasing the thickness of the ferromagnetic layer (75Å), the phase factor becomes negative ("\( \pi \)-state") and the DOS is flipped with respect to the normal state. When both electrodes are in the superconducting state the structures are amplified by the Al BCS singularity and shifted in energy by the aluminium gap as expected for elastic tunneling.

A check on the magnetic properties of F is shown in Fig. 2b and Fig. 2c, which presents the normalized Hall resistivity, \( \rho_{Hall}/\rho^2 \), vs. applied field, of the 50 and 75 Å thick PdNi layers respectively, corresponding to the "0 and \( \pi \)-state" measured by tunneling spectroscopy. The Hall resistivity is sensitive to magnetic scattering through the spin-orbit coupling and provides a suitable probe of weak magnetic moments in thin films. In ferromagnetic materials scattering by defects produces a net asymmetry in the transverse current density that is compensated, at equilibrium, by the anomalous Hall field. The Hall resistivity shows a fast variation at low magnetic field when the magnetic domains order and a linear dependence at higher field corresponding to the ordinary Hall effect. As the anomalous Hall effect is proportional to the magnetization and to the square
of $\rho$, the film resistivity, the extrapolation of $\rho_{\text{Hall}}/\rho^2$ at zero field is directly proportional to the saturation magnetization \[13\]. Complementary measurements by MOKE (Magneto-Optical-Kerr-Effect) on junctions presenting the same structure also show ferromagnetic ordering with a typical coercive field, $H_c$, of 1500 Gauss close to that measured by the anomalous Hall effect ($H_c = 1200$ Gauss). Finally, from the direct measurement of the saturation magnetization by SQUID we can extract the exchange energy and hence verify the estimated coherence length in the ferromagnet. We obtain $M = 0.21\mu_B$ which gives $E_{ex} = 15meV$ and $\xi_F = 45$ Å.

Increasing the thickness of the ferromagnetic layer, i.e. for $x >> \xi_F$, the proximity effect disappears and the normalized tunneling conductance becomes equal to unity. The DOS at zero energy, $N(0)$, as a function of $x$ is simply found from the spatial dependence of the order parameter, being $N(0) = \Re(\sqrt{1 - \frac{\pi^2}{4} - \cos(x\sqrt{2}/\xi_F)e^{-x\sqrt{2}/\xi_F}})$. In Fig. 3 we present the DOS at zero energy vs. $x/\xi_F$. We observe quantitative agreement with the $\cos(x\sqrt{2}/\xi_F)e^{-x\sqrt{2}/\xi_F}$ dependence of the order parameter \[13\] presented in Fig. 3. Here $\xi_F$ is the coherence length in the dirty limit and is obtained by measuring the exchange energy from the saturation magnetization as indicated above. The only fitting parameter is the finite interface transparency, $\gamma_B = 0.035$, which accounts for the reduced pair amplitude in $F$. A shift of 15 Å in the thickness of the ferromagnetic layer was also included to account for interface roughness or interdiffusion as shown by X-ray reflectivity measurements \[24\].

Our results show that the superconducting order parameter induced into a ferromagnet by proximity effect oscillates with a period given by the exchange energy. They suggest that S/F nanostructures offer a unique way to investigate the interplay between superconductivity and magnetic order since they do not require comparable energy scales. Furthermore they indicate that the proximity effect may indeed be used to fabricate Josephson junctions with a $\pi$-phase shift, as recently proposed \[21\].

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**FIG. 1.** a) Exponentially damped oscillations of the real part of the superconducting order parameter induced into a ferromagnetic material by proximity effect. The space coordinate $x$ denotes the distance from the superconductor/ferromagnet interface. The period of the oscillations is set by the coherence length $\xi_F$. "0-state" and "\(\pi\)-state" correspond to a positive and negative sign of the real part of the order parameter, respectively. For the sake of simplicity, the superconductor is assumed unaffected by the exchange field of the ferromagnet $F$. Inset : superconducting density of states at zero temperature in the "0-state" and "\(\pi\)-state" for an\(\pi\)-state for an $1800$ Gauss lower order parameter amplitude. b) Schematic of the Andreev reflection process: an electron in the normal metal with momentum, $k_+$, is elastically reflected as a hole, $k_-$, at the superconductor/normal metal interface ($S/N$). c) If $N$ is spin polarized the momentum shift, $\Delta k_F$, is dominated by the spin-splitting of the up and down bands.
FIG. 2. a) Differential conductance vs. bias for two Al/Al$_2$O$_3$/PdNi/Nb tunnel junctions corresponding to two different thickness (50 Å and 75 Å) of PdNi. The spectra have been taken at T=300 mK and H=100 Gauss and normalized by the normal state conductance obtained applying a magnetic field higher than the Nb critical field. The tunneling spectra show the "0" and "π" state shape expected from Fig.1a when the thickness of the ferromagnetic layer is respectively smaller or larger than $\xi_F$. Note that the induced superconducting density of states (DOS) is small. The normalized conductance for a tunnel junction without PdNi is also reported on the r.h.s. The field dependence of the normalized Hall resistivity at T=1.5 K for the same PdNi films as in the tunnel junctions corresponding to the "0-state" (50 Å) and to the "π-state" (75Å) is shown in Fig. 3 b) and c) respectively. Long range magnetic order leads to saturation of the magnetic domains and field-induced hysteresis.

FIG. 3. Tunneling conductance at zero energy vs. the PdNi thickness normalized by the coherence length $\xi_F$. The data taken at T=300 mK and H=100 G, are shown as solid symbols. The theoretical curve (dotted line) obtained solving the Usadel equations in the presence of an exchange field takes into account a finite interface resistance as fitting parameter. The dashed line denotes the transition from the "0-state" to the "π-state".
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