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2001 J. Phys.: Condens. Matter 13 6445

(http://iopscience.iop.org/0953-8984/13/30/301)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 6445–6451 PII: S0953-8984(01)20903-5

Superconducting cross-correlations in ferromagnets: implications for thermodynamics and quantum transport

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Received 15 January 2001, in final form 5 June 2001
Published 13 July 2001
Online at stacks.iop.org/JPhysCM/13/6445

Abstract
It is demonstrated that non-local Cooper pairs can propagate in ferromagnetic electrodes with opposite spin orientations. In the presence of such cross-correlations, the superconducting gap is found to depend explicitly on the relative orientation of the ferromagnetic electrodes. Non-local Cooper pairs can in principle be probed by means of direct-current (dc) transport. For two ferromagnetic electrodes, we propose a 'quantum switch' that can be used to detect correlated pairs of electrons. For three or more ferromagnetic electrodes, the Cooper pair-like state is a linear superposition of Cooper pairs which could be detected in the dc transport. The effect also induces an enhancement of the ferromagnetic proximity effect on the basis of superconducting cross-correlations propagating along domain walls.

Ferromagnetism and superconductivity are antagonist correlated states of matter. In ferromagnetism, one spin population is favoured because of spin symmetry breaking, while in s-wave superconductivity, electrons with the opposite spin are bound into Cooper pairs because of the attractive electron–electron interaction. Determination of to what extent these two orders can coexist in the same system has been a long-standing problem. As first proposed 40 years ago by Anderson and Suhl, the coexistence is possible if the ferromagnet acquires a cryptomagnetic [1] or cryptomagnetic-like [2] domain structure. On the other hand, in superconductor/ferromagnet heterostructures, a Cooper pair penetrating into a *single-domain* ferromagnet acquires a finite kinetic energy due to the coupling to the exchange field. This results in a spatial oscillation of the induced superconducting order parameter [3–6], giving rise to the so-called π-state, which has been probed recently in two experiments [7, 8]. In this letter, we consider Cooper pair penetration in a *multi-domain* ferromagnet. It has already been shown theoretically that crossed Andreev reflections can arise in a heterostructure in which two ferromagnets with opposite spin orientations are connected to a superconductor [9]. Such Andreev reflections do not exist when a single-domain ferromagnet is in contact with a superconductor [10–12]. We demonstrate here that quasi-long-range superconducting correlations can propagate in two magnetic domains with opposite magnetizations. These correlations correspond to non-local Cooper pair-like objects in which the spin-up (spin-down) electron propagates in a spin-up (spin-down) ferromagnetic domain.

This implies several consequences that may be tested in future experiments. First, considering the problem from the point of view of a superconductor order parameter coupled to a ferromagnetic environment, we show that the transition temperature of the superconductor depends explicitly on the relative spin orientations of the electrodes. The superconducting gap is smaller when the electrodes are misoriented.

The second implication of the model is that ferromagnetic domain walls can propagate superconducting cross-correlations, in which the two electrons making up a Cooper pair reside in neighbouring magnetic domains. This may explain the enhancement of the proximity effect observed in ferromagnet/superconductor heterostructures [13–16].

The third implication of the model is related to the production and measurement of linear superpositions of non-local Cooper pairs. It was stressed by Einstein, Podolsky and Rosen in 1935 [17] that non-locality is a deep feature of quantum mechanics. Non-locality [18] has been probed experimentally with photons [19, 20]. Condensed matter systems will perhaps provide the opportunity to fabricate entangled states with electrons, which are massive particles, and to fabricate quantum bits, which could be the building blocks of a quantum computer [21–24]. Two proposals have been made recently: one is based on tunnelling in a double quantum dot [25] and the other is based on noise correlations of Cooper pairs emitted in a beam splitter [26]. We show that superconducting cross-correlations in ferromagnets provide the possibility of manipulating linear superpositions of Cooper pairs. For two ferromagnetic electrodes, we propose a 'quantum switch' device that can be used to detect correlated pairs of electrons. Linear superpositions can be obtained with three or more ferromagnetic electrodes, and can be probed by means of dc transport.

Let us now consider a microscopic model in which a superconductor is connected to external electrodes. The superconductor is represented by the single-site effective Nambu Green's function [27]

$$\hat{g}^{R,A}(\omega) = g(\omega \pm \mathrm{i}\eta)\hat{I} + f(\omega \pm \mathrm{i}\eta)\hat{\sigma}^x$$

with

$$g(\omega) = -\pi\rho_N\omega/\sqrt{\Delta^2-\omega^2} \qquad f(\omega) = \pi\rho_N\Delta/\sqrt{\Delta^2-\omega^2}$$

and where ρ_N, having the dimension of an inverse energy, is the normal-state density of states, $\hat{I}$ is the 2×2 identity matrix, and $\hat{\sigma}^x$ is a Pauli matrix. We assume that N ferromagnetic electrodes are in contact with the superconductor (see figure 1(a)), with a hopping Hamiltonian

$$W = \sum_{k=1}^{N} t_{x,\alpha_k}\left[c_{\alpha_k}^{+}c_x + c_x^{+}c_{\alpha_k}\right].$$

The electrode k having a spin polarization $P_k = (\rho_{k,\uparrow} - \rho_{k,\downarrow})/(\rho_{k,\uparrow} + \rho_{k,\downarrow})$ is represented by the Green's function $\hat{g}_k^{A,R} = \pm\mathrm{i}\pi[\rho_{k,\uparrow}(\hat{I}+\hat{\sigma}^z)/2 + \rho_{k,\downarrow}(\hat{I}-\hat{\sigma}^z)/2]$. We use a perturbation theory based on the tunnel amplitude W, which we sum up to infinite order [27, 28]. The Dyson equation takes the form

$$\hat{G}_{x,x}^{R,A} = \left[\hat{I} - \sum_{k=1}^{N}\hat{g}_{x,x}^{R,A}\hat{t}_{x,\alpha_k}\hat{g}_{\alpha_k,\alpha_k}^{R,A}\hat{t}_{\alpha_k,x}\right]^{-1}\hat{g}_{x,x}^{R,A} \tag{1}$$

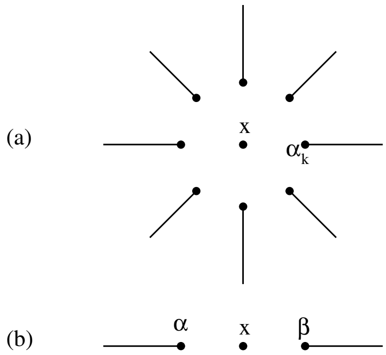

Figure 1. A schematic representation of the models. (a) The model with a coupling of the superconducting site x to N ferromagnetic electrodes. (b) The model with a coupling to two ferromagnetic electrodes.

where $\hat{t}_{\alpha_k,x}$ is the Nambu representation of the tunnel matrix element: $\hat{t}_{\alpha_k,x} = t_{\alpha_k,x}\hat{\sigma}^z$. The relevant parameters appear to be the spectral linewidth associated with spin-σ electrons:

$$\Gamma_\sigma = \sum_{k=1}^{N} \Gamma_{k,\sigma} \qquad \text{with } \Gamma_{k,\sigma} = (t_{\alpha_k,x})^2 \rho_{k,\sigma}.$$

Solving equation (1) leads to

$$\hat{G}^A_{x,x} = \frac{1}{\mathcal{D}}\left\{ g\hat{I} + f\hat{\sigma}^x + \mathrm{i}\pi(f^2 - g^2)\left[\frac{\Gamma_\downarrow}{2}(\hat{I} + \hat{\sigma}^z) + \frac{\Gamma_\uparrow}{2}(\hat{I} - \hat{\sigma}^z)\right]\right\} \tag{2}$$

with

$$\mathcal{D} = 1 - \mathrm{i}\pi g(\Gamma_\uparrow + \Gamma_\downarrow) + \pi^2(f^2 - g^2)\Gamma_\uparrow\Gamma_\downarrow.$$

To calculate the superconducting order parameter, we need to solve the Dyson–Keldysh equation

$$\hat{G}^{+-} = (\hat{I} + \hat{G}^R \otimes \hat{W}) \otimes \hat{g}^{+-} \otimes (\hat{I} + \hat{W} \otimes \hat{G}^A)$$

where the convolution includes a sum over the labels x and α_k.

Noting that $X_\sigma = (1 - \mathrm{i}\pi g\Gamma_{-\sigma})/\mathcal{D}$, $Y_\sigma = \mathrm{i}\pi f\Gamma_\sigma/\mathcal{D}$, and using equation (2), we obtain the exact expression for the Nambu component of the Keldysh Green's function:

$$\begin{aligned}\left[G^{+-}_{x,x}\right]_{2,1} = 2\mathrm{i}\pi n_F(\omega)\Big\{&\rho_g(X_\uparrow\overline{Y}_\uparrow + Y_\downarrow\overline{X}_\downarrow) + \rho_f(X_\uparrow\overline{X}_\downarrow + Y_\downarrow\overline{Y}_\uparrow)\\ &+ \frac{1}{\pi^2\Gamma^\downarrow}\overline{Y}^\downarrow(X^\uparrow - 1) + \frac{1}{\pi^2\Gamma^\uparrow}Y^\uparrow(\overline{X}^\downarrow - 1)\Big\}\end{aligned} \tag{3}$$

where $n_F(\omega)$ is the Fermi distribution, and we used the notation $\hat{\rho} = \rho_g\hat{I} + \rho_f\hat{\sigma}^x = \mathrm{Im}[\hat{g}^A]/\pi$. The superconducting gap is obtained by imposing the self-consistent equation [29]

$$\Delta = U\int_{-\infty}^{+\infty}[\mathrm{d}\omega/(2\mathrm{i}\pi)][\hat{G}^{+-}(\omega)]_{2,1}$$

with U the microscopic attractive interaction. The dominant contribution arises from the large-$|\omega|$ behaviour and we obtain a BCS-type relation:

$$\Delta = D\exp\left[-\frac{1}{\rho_N U}(1 + \pi\rho_N\Gamma_\uparrow)(1 + \pi\rho_N\Gamma_\downarrow)\right] \tag{4}$$

with D the bandwidth of the superconductor. As an example, we consider a coupling to two ferromagnets. With a parallel alignment of the magnetization in the electrodes, we have $\Gamma_\uparrow = 2\gamma$ and $\Gamma_\downarrow = 0$. With an antiparallel alignment, we have $\Gamma_\uparrow = \Gamma_\downarrow = \gamma$. The ratio of the two gaps is found to be

$$\frac{\Delta_{AP}}{\Delta_P} = \exp\left(-\frac{\pi^2 \rho_N \gamma^2}{U}\right) \tag{5}$$

which shows that the spin-polarized environment generates a reduction of the superconducting gap that depends explicitly on the spin orientation of the environment. The transition temperature of the superconductor is larger if the electrodes are in an antiparallel alignment. This behaviour should be contrasted with another model proposed recently [30].

As we show now, the gap variation equation (5) arises from the possibility that superconducting pairs can delocalize in the ferromagnetic electrodes having opposite spin orientations. Let us consider the problem with two electrodes only. The two electrodes are labelled by the Greek indices $\alpha_1 = \alpha$ and $\alpha_2 = \beta$. We use equation (3) to calculate exactly the crossed Keldysh Green's functions:

$$\left[G^{+-}_{\alpha,\beta}\right]_{2,1} = \mathrm{i}\langle c^+_{\beta,\uparrow} c^+_{\alpha,\downarrow}\rangle = \pi^2 t_\alpha t_\beta \rho_{\alpha,\downarrow}\rho_{\beta,\uparrow}\left[G^{+-}_{x,x}\right]_{2,1} \tag{6}$$

$$\left[G^{+-}_{\alpha,\beta}\right]_{1,2} = \mathrm{i}\langle c_{\beta,\downarrow} c_{\alpha,\uparrow}\rangle = \pi^2 t_\alpha t_\beta \rho_{\alpha,\uparrow}\rho_{\beta,\downarrow}\left[G^{+-}_{x,x}\right]_{1,2} \tag{7}$$

with $[\hat{G}^{+-}_{x,x}]_{2,1} = \overline{[\hat{G}^{+-}_{x,x}]_{1,2}}$ given by equation (3). The density-of-states prefactors in equations (6), (7) appear to be a direct consequence of the Pauli exclusion principle. To show this, we consider equations (6), (7) in the limit of fully polarized ferromagnets. In the parallel alignment ($\rho_{\alpha,\uparrow} = \rho_{\beta,\uparrow} = 1$, $\rho_{\alpha,\downarrow} = \rho_{\beta,\downarrow} = 0$), all pair correlations are vanishing: $\langle c^+_{\beta,\uparrow} c^+_{\alpha,\downarrow}\rangle = \langle c_{\beta,\downarrow} c_{\alpha,\uparrow}\rangle = 0$. This is fully expected because one cannot add or destroy a spin-down electron in the presence of a spin-up band only. For the same reason, one has $\langle c^+_{\beta,\uparrow} c^+_{\alpha,\downarrow}\rangle = 0$ in the antiparallel alignment ($\rho_{\alpha,\uparrow} = \rho_{\beta,\downarrow} = 1$, $\rho_{\alpha,\downarrow} = \rho_{\beta,\uparrow} = 0$). The remaining non-vanishing cross-correlations are $\langle c_{\beta,\downarrow} c_{\alpha,\uparrow}\rangle$ and $\langle c^+_{\beta,\downarrow} c^+_{\alpha,\uparrow}\rangle$. This shows the possibility of generating superconducting cross-correlations in two ferromagnets with opposite magnetizations. To characterize the propagation of cross-correlations, we calculate the Gorkov function $\hat{G}^{+-}_{i,j}$, with i and j two sites in the ferromagnetic electrodes α and β such that $x_i = -x_j$. Assuming that the ferromagnetic electrodes behave like a three-dimensional metal, we find

$$\left[\hat{G}^{+-}_{i,j}\right]_{1,2} \sim \frac{1}{|x_i|}\pi^2 t_\alpha t_\beta \rho_{\alpha,\uparrow}\rho_{\beta,\downarrow}\left[\hat{G}^{+-}_{x,x}\right]_{1,2}.$$

By comparison, there is a density-of-states prefactor $\rho_{\alpha,\uparrow}\rho_{\alpha,\downarrow}$ in the *local* superconducting correlation in electrode α. As a consequence, in strongly spin-polarized ferromagnets, superconducting cross-correlations can propagate while ordinary superconducting correlations cannot propagate. It is well known that there is an oscillation-induced order parameter associated with Cooper pair penetration in partially spin-polarized ferromagnets [2–6]. There are no such oscillations in the case of cross-correlations because Cooper pairs do not acquire a centre-of-mass momentum when entering the ferromagnetic electrodes.

The model can be considered from the point of view of propagation of cross-correlated Cooper pairs along domain walls in a multi-domain ferromagnet. Such cross-correlations can generate an enhancement of the ferromagnetic–superconducting proximity effect, which does not contradict the results of recent experiments on ferromagnet/superconductor heterostructures [13–15]. Another proposal based on spin accumulation has been made recently [16], but appears to be incompatible with some experiments [14]. Our scenario and the spin-accumulation picture both contribute to the same effect, but in a different situation: cross-correlations can propagate only in multi-domain ferromagnets, while the spin-accumulation mechanism is valid even with single-domain ferromagnets.

Now we show that superconducting cross-correlations can be used to produce correlated pairs of electrons. Let us consider two ferromagnets α and β in contact with a superconductor. The cross-correlated degrees of freedom are represented by the Cooper pair-like wave function $|\psi\rangle = [u_0 + v_0 c^+_{\alpha,\uparrow} c^+_{\beta,\downarrow}]|0\rangle$, with u_0 and v_0 the BCS coherence factors. Let us consider two additional ferromagnetic electrodes α' and β' having spin orientations $\Sigma_{\alpha'}$ and $\Sigma_{\beta'}$ connected to the electrodes α and β (see figure 2). The electrodes α' and β' are considered to be reservoirs in which all inelastic processes take place. For the sake of obtaining the basic physics of such systems, we restrict ourselves to fully polarized ferromagnets and high-transparency contacts [31]. If $\Sigma_{\alpha'} = \uparrow$, $\Sigma_{\beta'} = \downarrow$, the correlated pair can be transmitted into the reservoirs α' and β' and a finite current is flowing into the superconductor (see figure 2(a)). If $\Sigma_{\alpha'} = \Sigma_{\beta'} = \downarrow$, the spin-up electron making the correlated state is backscattered at the interface with the spin-down ferromagnet α'. Coming back onto the superconductor interface it undergoes a crossed Andreev reflection [9] in which a Cooper is formed in the superconductor and a spin-down hole is transferred into electrode β. The whole process does not carry electrical charge: there is no current transmitted into the superconductor (see figure 2(b)). The 'quantum switch' device in figure 2 can therefore be used to produce and detect correlated pairs of electrons by means of dc transport.

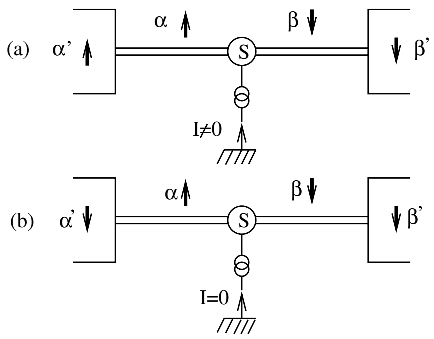

Figure 2. A schematic representation of the quantum switch used to probe correlated pairs of electrons. A current source is connected to the superconductor. In (a), there is a finite current flowing. In (b), there is no current flowing.

Now we discuss the production of linear superpositions in a three-terminal device (see figure 3). The three ferromagnetic electrodes are labelled by the indices $\alpha_1 = \alpha$, $\alpha_2 = \beta$, and $\alpha_3 = \gamma$. With fully polarized ferromagnets having a spin orientation $\sigma_\alpha = \sigma_\beta = \uparrow$, $\sigma_\gamma = \downarrow$, the exact form of the cross-correlations is given by $[\hat{G}^{+-}_{\alpha(\beta),\gamma}]_{1,2} = \pi^2 t_{\alpha(\beta)} t_\gamma [\hat{G}^{+-}_{x,x}]_{1,2}$. This means that Cooper pairs can delocalize over several electrodes. The corresponding wave function is a linear superposition of Cooper pairs

$$|\psi\rangle = \lambda_\alpha [u_0 + v_0 c^+_{\alpha,\uparrow} c^+_{\gamma,\downarrow}]|0\rangle + \lambda_\beta [u_0 + v_0 c^+_{\beta,\uparrow} c^+_{\gamma,\downarrow}]|0\rangle.$$

The coefficients λ_α and λ_β are such that the Cooper pair wave function contains the same pair correlations as the Gorkov function:

$$\langle c^+_{\beta,\uparrow} c^+_{\gamma_\downarrow}\rangle / \langle c^+_{\alpha,\uparrow} c^+_{\gamma_\downarrow}\rangle = \lambda_\beta/\lambda_\alpha = t_\beta/t_\alpha$$

from which we deduce

$$\lambda_{\alpha(\beta)} = t_{\alpha(\beta)} \Big/ \sqrt{t_\alpha^2 + t_\beta^2 + 2u_0^2 t_\alpha t_\beta}.$$

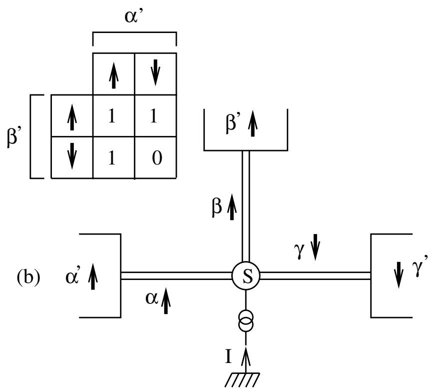

Figure 3. A schematic representation of the three-terminal device used to probe the linear superposition of Cooper pairs. The inset shows the presence/absence of a current flowing into the superconductor as a function of the spin orientation in the ferromagnetic reservoirs.

As a direct consequence of the linear superposition, the current flowing into the superconductor is vanishing if $\Sigma_{\alpha'} = \Sigma_{\beta'} = \downarrow$ and finite in the three other spin orientations (see figure 3). Now the linear superposition associated with the magnetization of the electrodes $\sigma_\alpha = \uparrow$, $\sigma_\beta = \sigma_\gamma = \downarrow$ is

$$|\psi\rangle = \lambda'_\beta[u_0 + v_0 c^+_{\alpha,\uparrow} c^+_{\beta,\downarrow}]|0\rangle + \lambda'_\gamma[u_0 + v_0 c^+_{\alpha,\uparrow} c^+_{\gamma,\downarrow}]|0\rangle$$

with

$$\lambda'_{\beta(\gamma)} = t_{\beta(\gamma)} \Big/ \sqrt{t_\beta^2 + t_\gamma^2 + 2u_0^2 t_\beta t_\gamma}.$$

The current flowing into the superconductor is vanishing in the two spin orientations $\Sigma_{\alpha'} = \downarrow$, $\Sigma_{\beta'} = \uparrow, \downarrow$ and finite otherwise. Therefore, a dc-current measurement can make the distinction between the linear superpositions associated with the spin orientations $\sigma_\alpha = \sigma_\beta = \uparrow$, $\sigma_\gamma = \downarrow$ and $\sigma_\alpha = \uparrow$, $\sigma_\beta = \sigma_\gamma = \downarrow$.

To conclude, we have shown that quasi-long-range superconducting cross-correlations can propagate in ferromagnets having opposite spin orientations. The superconducting cross-correlations are much stronger than the local ones. Such cross-correlations can propagate along ferromagnetic domain walls, and contribute to an enhancement of the ferromagnetic proximity effect, which may be present in recent experiments [13–15]. The superconducting gap depends explicitly on the spin orientation of the ferromagnetic electrodes, which could be used as an experimental probe of superconducting cross-correlations. We have shown that cross-correlations can be used to produce correlated pairs of electrons and linear superpositions of correlated pairs. Such states can in principle be detected by means of dc transport. The microscopic calculation of the current will be the subject of future work.

Acknowledgments

The author acknowledges invaluable discussions with P Degiovanni, D Feinberg and M Giroud.

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