Odd triplet superconductivity in superconductor/ferromagnet multilayered structures.

A.F. Volkov\textsuperscript{1,2}, F.S. Bergeret\textsuperscript{1}, and K.B. Efetov\textsuperscript{1,3}

\textsuperscript{(1)} Ruhr University Bochum, D-44780 Bochum, Germany
\textsuperscript{(2)} Institute of Radioengineering and Electronics of the Russian Academy of Sciences, Moscow 103907, Russia
\textsuperscript{(3)} L.D. Landau Institute for Theoretical Physics, 117940 Moscow, Russia

We demonstrate that in multilayered superconductor-ferromagnet structures a non-collinear alignment of the magnetizations of different ferromagnetic layers generates a triplet superconducting condensate which is odd in frequency. This triplet condensate coexists in the superconductors with the conventional singlet one but decays very slowly in the ferromagnet, which should lead to a large Josephson effect between the superconductors separated by the ferromagnet. Depending on the mutual direction of the ferromagnetic moments the Josephson coupling can be both of 0 and π type.

It is well known that ferromagnetism and superconductivity are antagonistic phenomena; ferromagnetism strongly suppresses superconductivity. This suppression in superconductor/ferromagnet (S/F) layered structures is caused mainly by the exchange interaction. This means in particular that the singlet Cooper pairs cannot penetrate into the F layers over a noticeable length, since the exchange energy is by orders of magnitude larger than the Cooper binding energy. Thus, the singlet pairs are destroyed by the exchange field because the spins of the electrons cannot be antiparallel anymore. This suppression of superconductivity can be reduced in S/F structures if the ferromagnetic layers are ultrathin, but, generally, the destruction of the singlet superconductivity by a homogeneous exchange field can hardly be avoided.

The situation may be different if the spins of the superconducting pairs are parallel to each other. It is clear that such a triplet superconductivity is not sensitive to the exchange field and the coexistence of the superconductivity and ferromagnetism becomes possible. Unfortunately, the triplet pairing is a rather exotic phenomenon and has been observed until now only in superfluid He\textsuperscript{3} and in a superconducting material Sr\textsubscript{2}RuO\textsubscript{4} \cite{1}. It is expected to be very sensitive to disorder \cite{1,2}, which makes its observation even more difficult. In order to satisfy fermionic commutation relations the condensate function, even in frequency, should be odd in the momentum of the pair and this is the reason why it is so sensitive to impurities. Another possibility was suggested by Berezinskii long ago \cite{3}. He conjectured that the triplet superconductivity might be possible if the condensate function were even in the momentum but odd in the frequency. Attempts to find conditions for the existence of such an odd superconductivity were done in several works much later \cite{4} but the results were not encouraging (in Ref. \cite{4} a singlet pairing odd in frequency and in the momentum was considered).

In this Letter, we demonstrate that the odd triplet superconductivity is not exotic at all and is \textit{inavoidable} in multilayered S/F structures with conventional superconductors if the directions of the magnetization of the different F-layers are neither parallel nor antiparallel to each other. In this case the triplet condensate (TC) can easily penetrate into the ferromagnetic layers over long distances and result in supercurrents through the ferromagnets. The conditions for the realization of the odd triplet superconductivity do not seem to be problematic from the experimental point of view and we hope that proper measurements will be done in the nearest future.

A generation of a triplet condensate by a non-homogeneous magnetization (domain walls) and its penetration into a ferromagnet has been discussed recently \cite{5,6}. In the structures considered in these works only changes in the conductivity of the system were analyzed. However, the detection of the triplet component in such structures is a quite difficult task. In contrast, we predict in this Letter a new type of \textit{superconductivity} (odd triplet) in S/F structures and discuss how to identify it experimentally. To be specific, we consider a S/F multilayered structure (Fig. 1) in which the new superconducting state might be observed. In this state, the superconductivity in the S regions is caused by the singlet component (SC) and takes place in the plane of the layers, while the transverse superconductivity through the F layers is mainly due to the TC. Moreover, the relation between the condensate current $I_S$ and a phase difference $\varphi$ depends in a crucial way on chirality of the magnetic moment $\mathbf{M}$ in space.

The multilayered S/F structure we consider is represented in Fig.1. The (in-plane) magnetizations $\mathbf{M}$ of the neighboring F layers are not parallel to each other and the angle between them is $2\alpha$. In order to achieve such a non-collinear alignment one can employ an exchange-biased spin-valve or ferromagnets with strong anisotropy and different easy-axis of magnetization. It will be shown that in such a structure the TC may arise if the thickness of the superconducting layers $2d_S$ is less or comparable with the coherence length $\xi_S = \sqrt{D_S/2\pi T}$. The TC penetrates into the F layer over a long distance $\xi_T = \sqrt{D_F/2\pi T}$ and ensures a Josephson coupling between the nearest $S$ layers. In the case under consideration the relation between the condensate current $I_S$ and the phase difference $\varphi$ has the conventional form $I_S = I_c \sin \varphi$. However, the sign of the critical current
\( I_c \) depends on the chirality, namely, it is positive if the rotation angle \( 2\alpha_0 \) of the magnetization at the \( S_i \) layer has the same sign for neighboring \( S \) layers (\( S_i \) and \( S_{i+1} \)) and it is negative if the signs of the rotation are opposite for neighboring \( S \) layers (\( \alpha_i\alpha_{i+1} < 0 \)).

We consider the simplest case of a dirty system for which the thickness of the \( \text{condensate function} \) needs to make further assumptions. First, we assume that the \( M \) vector lies in the \( (y, z) \) plane and \( \alpha \) is the angle between the \( z \)-axis and \( M \). Eqs. (1-2) have to be supplemented by boundary conditions. In the case of a perfect \( S/F \) interface (the reflection coefficient is very small) they have the form [9]

\[
\begin{align*}
\dot{f}(d_S + 0) - \dot{f}(d_S - 0) &= 0, \\
\gamma &\frac{\partial_x}{\tau} \dot{f}(d_S + 0) - \partial_x \dot{f}(d_S - 0) = 0,
\end{align*}
\]

where \( \gamma = \sigma_F/\sigma_S, \sigma_{F,S} \) are the conductivities in the \( F \) and \( S \) regions (for simplicity we do not take into account a dependence of the conductivity \( \sigma_F \) on the spin directions). If the SF interface resistance \( R_b \) is finite the r.h.s of the first equation equals \( \gamma_\varepsilon \frac{\partial_x}{\tau} \dot{f}(d_S + 0) \), where \( \gamma_\varepsilon \) is proportional to \( R_b \) [10]. For simplicity we set \( \gamma_\varepsilon = 0 \). A generalization of the results for the case of a finite \( \gamma_\varepsilon \) does not lead to qualitative changes.

The condensate matrix function \( \dot{f} \) has following form

\[
\dot{f} (x) = i \left( \dot{f}_1 (x) \hat{\sigma}_1 + \dot{f}_2 (x) \hat{\sigma}_2 \right),
\]

where \( \dot{f}_{1,2} \) are matrices in the spin space. In the \( F \) layers they can be written as

\[
\dot{f}_1 = B_1 (x) \hat{\sigma}_1 + B_2 (x) \hat{\sigma}_2, \\
\dot{f}_2 = B_0 (x) + B_3 (x) \hat{\sigma}_3.
\]

In the \( S \) layers the functions \( \dot{f}_{1,2} \) have the same form, but the coefficients \( B \) should be replaced by \( A \). The functions \( B_i (x) \) have the form: \( B_i (x) \sim b_i \exp(\mp \kappa (x \mp d_S)) \). Substituting these expressions for \( B_i (x) \) into Eqs. (1-2) one obtains a set of algebraic equations that determine the eigenvalues \( \kappa \). In the \( S \) region all condensate components (SC and TC) are decoupled and there is only one solution for \( \kappa: \kappa^2 = 2|\omega|/D_S \). In the \( F \) region the SC and TC are coupled by the exchange interaction (if \( \alpha \) is not zero) and there are three eigenvalues for \( \kappa \). Two of them are equal to \( \kappa_ \pm = \kappa_J(1 \pm i) \), and determine a fast decay of the condensate in the \( F \) layers (dashed line in Fig. 2), here \( \kappa_J = \sqrt{J/D_F} \). This result demonstrates the well known short-range penetration of the superconducting condensate into the ferromagnet (see for example the review article [11]). However, the third solution for \( \kappa \) is completely different and is given by \( \kappa_\omega = \sqrt{2|\omega|/D_F} \).

Thus, we see that the huge exchange energy \( J \) entering the solution for \( \kappa_+ \) is replaced by a small energy \( \omega \) of the order of \( T \). At large distances it is the solution with \( \kappa_\omega \) that determines the penetration of the superconducting condensate into the \( F \) regions (see Fig. 2). It corresponds to the triplet component and, as it has been discussed in the introduction, there is no wonder that the exchange field does not influence it.

Having determined the solutions for \( \kappa_+ \), we can write the solutions for \( B_i (x) \). For example, the coefficient \( B_1 (x) \) may be written at \( x > d_S \) in the form
\[ B_1(x) = \sum_i b_{1i} \exp[-\kappa_i(x - d_S)] , \quad (6) \]

where \( b_{1i} = b_{1\omega}, b_{1\pm} \) and \( \kappa_i = \kappa_x, \kappa_{1\pm}. \) The coefficients \( b_{1\pm} \) are related to the coefficients \( b_{1\pm} \) that determine the magnitude of the SC by \( b_{1\pm} = \mp \sin \alpha \sgn \omega \cdot b_{1\pm}. \)

What remains to be done is to determine the coefficients \( a_i, b_i, \) using the boundary conditions (3) for an arbitrary angle \( \alpha. \) In a general case the expressions for the coefficients are cumbersome, and for clarity we present here only results for small \( \alpha. \) In zero order in \( \alpha, \) only terms \( \alpha/|\kappa_x| \sim \alpha/\kappa_S \) contribute. The terms \( \sim \alpha/\kappa_x \) are finite in the main approximation. Secondly, in order to fulfill the matching conditions (3), one has to take into account the main approximation. For example, for \( b_{1\pm} \) we obtain

\[ b_{1\pm} = \frac{\Delta}{2|\omega|} \frac{1}{1 + \gamma \kappa_{1\pm}/(\kappa_S \tan \theta_S)} \quad (7) \]

where \( \kappa_S = \sqrt{2|\omega|/D_S}, \theta_S = \kappa_S d_S. \) This solution corresponds to those obtained earlier (see [11] and references therein) where the case of a parallel or antiparallel magnetization alignment was analyzed. It is valid in the limiting cases \( \theta_S << 1 \) and \( \theta_S >> 1 \) (if \( T \) is close to the critical temperature \( T_c \)).

If the magnetization vectors in the neighboring \( F \) layers are inclined with respect to each other by an angle \( 2\alpha, \) the situation changes qualitatively. First, the function \( \hat{f}_0 \) that describes the TC is no longer zero because the terms proportional to \( b_{1\pm} \) in the expression for \( B_1(x) \) (Eq. (6)) are related to \( b_{1\pm}, \) and hence they are finite in the main approximation. Secondly, in order to fulfill the matching conditions (3), one has to take into account the first long-range term in Eq.(6). Using the boundary conditions, we obtain for the coefficient \( b_{1\omega} \) the following expression

\[ b_{1\omega} = -\frac{\Delta}{\omega} \sin \alpha \frac{\kappa_J \tan \theta_S}{\cos^2 \theta_S \gamma \kappa_x/\kappa_S + \tan \theta_S} \frac{1}{(\kappa_{1\omega} \tan \theta_S + \kappa_S/\gamma)} \quad (8) \]

Contrary to the SC determined by the solution \( B_0(x) \) and \( A_0(x), \) the TC is an odd function of \( x. \) This is seen from Eqs. (6), (8) and the fact the \( \alpha \) has different signs at \( x > d_S \) and \( x < -d_S. \) According to the boundary conditions (3) (continuity of the condensate function and current conservation), the TC induced in the \( F \) layer penetrates into the superconductor. The corresponding solution in the \( S \) layer has the form: \( A_1(x) = a_1 \sin (\kappa_S x). \) In the limit \( T << J, \) the TC penetrates into the \( F \) layer over the length of the order \( 1/\kappa_T = \sqrt{D_F/2\pi T} \) which is much greater than the SC penetration length \( 1/\kappa_x. \) One can see from Eq. (8) that the amplitude \( b_{1\omega} \) of the long-range TC is an odd function of the Matsubara frequency \( \omega \) and is symmetric in the momentum space as in the case of the TC which arises in a ferromagnet with a non-homogeneous \( M \) near a \( S/F \) interface [5]. This new type of the condensate, odd in \( \omega \) and even in the momenta \( p, \) has been proposed by Berezinskii [3] in order to explain the pairing mechanism in He\(^3\) (it was proven later that the condensate in He\(^3\) is in fact even in \( \omega \) and odd in \( p \)). The solution we present here corresponds to this hypothetical pairing, which means that we have found a concrete realization of the idea. It follows from the geometry of the structure we consider that the odd triplet superconductivity we found exists in the transverse direction.

It is seen from Eqs. (7) and (8) that the amplitudes of the SC and TC at the \( S/F \) interface are comparable if \( \xi_F << \xi_T \) and \( \alpha \) and \( \theta_S \) are of the order of 1. If the latter condition is not satisfied (that is, the thickness of the \( S \) layer \( d_S \) is large in comparison with \( \xi_F \)), the TC decays exponentially in \( S \) and its amplitude is small. Therefore we calculate the Josephson current between the neighboring \( S \) (\( S_1 \) and \( S_2 \)) layers in the case \( \theta_S << 1 \) and assuming that the condition \( \xi_F << \xi_T < 2d_F \) is fulfilled. Then, the Josephson current is due to the overlap of the TC induced near each \( S/F \) interface. In this case the TC in \( F \) is described by the expression

\[ \hat{f}_\text{trip}(x) = i \hat{\tau} \hat{\sigma}_1 b_{1\omega} \exp(-\kappa_{1\omega}(x - d_S)) + \hat{S} \cdot i \hat{\tau} \hat{\sigma}_1 \cdot \hat{S}^+ b_{1\omega} \exp(\kappa_{1\omega}(x - d_S - 2d_F)) \quad (9) \]

Here, the matrix of a gauge transformation \( \hat{S} = \cos(\varphi/2) + i \hat{\tau} \sin(\varphi/2) \) allows us to take into account the phase difference \( \varphi \) between the neighboring \( S \) layers (we assume that the phase of the \( S_1 \) layer is zero). The coefficients have different signs \( (b_{1\omega} = -b_{1\omega}) \) if the magnetization \( M \) at both \( S \) layers rotates in the same direction, and \( b_{1\omega} = b_{1\omega} \) in the opposite case (different signs of chiralities). The condensate current \( I_S \) between \( S_1 \) and \( S_2 \) is given by the formula

\[ I_S = (L_y L_z) \sigma_F (\pi t/4e) T \tau \hat{\tau} \hat{\sigma}_1 \sum_\omega \hat{f}_\text{trip} \quad (10) \]

From Eqs.(9-10), we get \( I_S = I_c \sin \varphi, \) where

\[ I_c = -(2\pi \epsilon R_F) 2d_F \sum_\omega \kappa_\omega b_{1\omega} b_{1\omega} \exp(-2d_F \kappa_\omega) \quad (11) \]

and \( R_F = 2d_F/((L_y L_z) \sigma_F) \) is the resistance of the \( F \) layer in the normal state. Substituting Eq.(8) into Eq.(11), we find for the critical current in the limit \( \theta_S << 1 \)

\[ \epsilon R_F I_c = \frac{4T}{\pi} \left( \frac{\Delta}{T} \right)^2 \frac{(\kappa_J d_S)^2 (d_F \kappa_F \epsilon e^{-2d_F \kappa_F} \sin \alpha_1 \sin \alpha_2}{|\kappa_S d_S + \gamma \kappa_J (1 + i/\kappa_S)|^2 (\kappa_T d_S + 1/\gamma)^2} \quad (12) \]

where \( \kappa_S = \sqrt{2\pi T/D_S}. \)

If the magnetization vector at each \( S \) layer rotates in the same direction \( (\alpha_1 = \alpha_2), \) then the critical Josephson current \( I_c \) is positive (0-contact). If \( \mathbf{M} \) rotates at \( S_1 \) and \( S_2 \) in opposite directions \( (\alpha_1 = -\alpha_2), \) then the critical current \( I_c \) is negative (\( \pi \)-contact). In the latter case a
phase difference $\pi$ is established between neighboring $S$ layers in a multilayered $S/F$ structure. We would like to note that the mechanism of the $\pi$--contact considered here is completely different from that suggested in Ref. [12] and observed in Ref. [13]. In our case, the negative critical current is caused by the TC, but not by the SC as in Ref. [12], and, in addition, is realized only if the chiralities at the $S_1$ and $S_2$ are different. The possibility of switching between the $0$ and $\pi$-states by changing the angle $\alpha$ may find applications in superconducting devices.

The effect described above exists for all temperatures below the critical temperature $T^*_c$ which is determined from the self-consistency equation. In the main approximation in the small angle $\alpha$ it agrees with that obtained in Refs. [10], [11] for the case of the parallel orientation of the magnetization. A correction to $T^*_c$ due to a small misalignment of the magnetizations in neighboring $F$ layers, i.e. due to the TC, is proportional to $\sin^2 \alpha$, but we are not interested in this correction here. We note that the critical temperature $T^*_c$ for the case of arbitrary $\alpha$ was analyzed in Ref. [14]. However, the form of the condensate function presented in that work is not correct because the authors started from an equation different from Eq. (2) (instead of the commutator, they wrote the product $\hat{J} \cdot \hat{f}$). As a result, the long range triplet component was completely lost.

In conclusion, we have predicted a new type of superconductivity. It was demonstrated that in superconductor-ferromagnet structures a non-collinear alignment of the exchange fields in the ferromagnetic layers generates a triplet component of the superconducting condensate and this component is odd in the frequency. The odd triplet condensate penetrates into the ferromagnet over long distances and is not sensitive to impurities.

Superconductors separated by the ferromagnet is possible and the critical current can be measured. The Josephson contact can be both of $0$ and $\pi$-type depending on the arrangement of the magnetic moments. We hope that the effects considered in this paper will be observed experimentally in the nearest future.

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