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Giant Mesoscopic Fluctuations and Long-Range Superconducting Correlations in Superconductor-Ferromagnet Structures

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The fluctuating superconducting correlations emerging in dirty hybrid structures under the conditions of the strong proximity effect are demonstrated to affect the validity range of the widely used formalism of Usadel equations at mesoscopic scales. In superconductor ferromagnet structures these giant mesoscopic fluctuations originating from the interference effects for the Cooper pair wave function in the presence of the exchange field can be responsible for an anomalously slow decay of superconducting correlations in a ferromagnet even when the noncollinear and spin orbit effects are negligible. The resulting sample to sample fluctuations of the Josephson current in superconductor ferromagnetic superconductor junctions and the local density of states in superconductor ferromagnetic hybrid structures can provide an explanation of the long range proximity phenomena observed in mesoscopic samples with collinear magnetization.

The successful development of modern experiments in a wide class of superconductor-ferromagnet (S - F) hybrid structures has opened a completely new research area, i.e., superconducting spintronics [1–4]. Despite the growing number of various exciting experimental and theoretical results in this field there remains still a very important and puzzling contradiction between the experimental data and the understanding of the physics of the proximity effect in these systems. This contradiction relates to the length of the decay of superconducting correlations, L_S , in a dirty ferromagnet. The standard Usadel theory gives us the exponential suppression of superconducting correlations at the length $L_S \sim \xi_F = \sqrt{\hbar D/2h}$, where D is the diffusion coefficient and h is the exchange field (see Ref. [5] for a review). For most typical ferromagnets and alloys one can get the estimate $L_S \sim 1$ – 10 nm while the existing experimental data [6–15] show that superconducting correlations survive for much larger distances from the S - F interface comparable with the coherence length in the normal metal (N) $\xi_N \sim \sqrt{D/T}$. This puzzling discrepancy between the theoretical estimate and experiment stimulated researchers to suggest various fascinating mechanisms of such a long-range proximity phenomenon. Most of these explanations exploit the assumption of the inhomogeneous exchange field, which generates the long-range triplet pairing component in the anomalous Green functions (see, e.g., Ref. [16]). The resulting increase in the L_S length has been confirmed experimentally for a Josephson junction with a composite F layer containing a region with noncollinear magnetic moments [11,12]. Thus, the noncollinearity of the magnetic structure nicely explains the overwhelming majority of the above experiments. An alternative source of the long-range effect originating from

the spin-orbit interaction has been recently discussed in Refs. [17–20]. However, up to now there are no satisfactory explanations of the experiments [6–8,13] where no traces of a noncollinear magnetization were reported and the strength of the spin-orbit effects can be a subject of debate.

Motivated by the above discrepancy between experiment and theory we suggest to reexamine the standard Usadel-type model and search for possible shortcomings of this model, which can reveal themselves in the L_S estimates. One of the most important assumptions that form the basis of the Usadel theory is that we operate with the ensemble-averaged Green functions neglecting, thus, possible fluctuations of the measurable quantities due to the random distribution of impurities [21–23]. In the case of the dirty ferromagnet this assumption is crucial to get the exponential decay of the anomalous Green function at the length ξ_F . Indeed, the motion of quasiparticles in a ferromagnetic metal occurs along random quasiclassical trajectories, which experience sharp turns at the impurity positions (see Fig. 1). The exchange field is responsible for the relative phase γ gained between the electronic and hole parts of the quasiparticle wave function along these trajectories. Averaging the Green functions we average

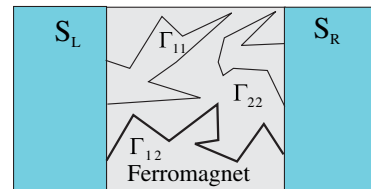


FIG. 1. Random quasiparticle trajectories in the S - F - S Josephson junction.

in fact the exponential phase factor $e^{i\gamma}$ with the random phase γ depending on the trajectory length, obtaining naturally an exponentially decaying quantity $\propto e^{-x/\xi_F}$, where x is the distance from the S - F interface. This destructive interference cannot play such a dramatic role when we calculate root-mean-square (rms) values due to a partial phase gain compensation in squared quantities. Considering, e.g., the supercurrent I of the S - F - S Josephson junction we can introduce the rms value of the current as follows: $\delta I = \sqrt{\langle I^2 \rangle - \langle I \rangle^2}$. The compensation of the phase factor γ can occur only for correlated random trajectories passing at a distance not exceeding the Fermi wavelength $\lambda_F = 2\pi/k_F$. This restriction causes the reduction of the δI value by a factor of \sqrt{N} , where N is the number of transport channels in the junction. Finally, we obtain $\delta I / \langle I \rangle \sim e^{d/\xi_F} / \sqrt{N}$, where d is the distance between the S electrodes. The number of channels can be of course pretty large: $N \sim k_F L$ for two-dimensional and $N \sim (k_F L)^2$ for three-dimensional junctions with the transverse dimension L . Nevertheless, the current fluctuations can strongly exceed the average value at large distances d well above the coherence length ξ_F . In this sense these fluctuations are giant compared to the ones in S - N - S junctions where the value $\delta I \sim e\Delta_0/\hbar$ for short junctions with $d \ll \xi_S$ [24] is known to be determined by the universal conductance fluctuations [25,26] or even smaller for long junctions with $d \gg \xi_S$ [27]. Here, Δ_0 is the gap in the bulk superconductor and ξ_S is the superconducting coherence length. Experimentally, in each particular sample we can expect to measure a random critical current value, which should exhibit giant sample-to-sample fluctuations. Thus, in a given experiment one can easily obtain the critical current well above the limit imposed by the Usadel theory, which can give us only the average current value. The above arguments and standard Landauer relation between the normal junction resistance R and the N number make it possible to guess a simple estimate for the fluctuating critical current:

$$\delta I \sim \Delta_0 / \sqrt{\hbar R}. \quad (1)$$

Note that this inverse square root dependence differs strongly from the standard relation $I_c \sim \Delta_0 / (eR)$ for the S - N - S junction. Our further calculations nicely confirm the above δI estimate and, thus, the observation of this unusual relation between the supercurrent and normal junction resistance could provide a verification of the long-range proximity mechanism caused by mesoscopic fluctuations. The ensemble averaging lying in the basis of the derivation of the Usadel equations from the quasiclassical Eilenberger theory overlooks the above fluctuation effects emerging at mesoscopic scales. As we show below these fluctuation effects reveal themselves even in the quasiclassical limit $\lambda_F \rightarrow 0$ when we can neglect the corrections found in Refs. [22,23], which vanish in this limit corresponding to a large junction conductance.

We proceed with a detailed consideration of the critical current fluctuations in the S - F - S junction using an approach based on the averaging over the random quasiparticle trajectories passing in the field of point scatterers (see Ref. [28] for a review). For each random trajectory inside the F layer one can consider the 1D problem for propagating electrons and holes experiencing Andreev reflection at the point where the trajectory touches the left or right S electrode. We start from the case $d \ll \xi_S$ and assume the superconducting gap (exchange field) to vanish inside (outside) the F layer. Thus, we neglect the so-called inverse proximity effect, i.e., the mutual influence of the order parameters at the interface. The current-phase relation for the short junction limit can be defined only from the spectra of the subgap Andreev states at the trajectories ending at both the left and right S electrodes $\epsilon = \pm\Delta_0 \cos[(\varphi \pm \gamma)/2]$, neglecting the contributions from the states above the gap. Here, φ is the phase difference between the S electrodes, and $\pm\gamma$ is the spin-dependent phase shift between the electronlike- and holelike parts of the total wave function along the quasiclassical trajectory Γ_{12} (see Fig. 1). Each trajectory Γ can touch each of the S electrodes only once; otherwise, part of the trajectory Γ touching the same electrode two times can be considered separately and the corresponding spectrum does not depend on the phase difference φ (trajectories Γ_{11} and Γ_{22} in Fig. 1). Certainly, there exist trajectories of the length exceeding ξ_S with the quasiparticle spectrum consisting of several subgap branches but the probability to get such trajectories vanishes for short junctions. According to the procedure suggested in Ref. [17], the phase shift γ can be determined from the equations that formally coincide with the Eilenberger-type equations written for the singlet and triplet parts of the anomalous quasiclassical Green function $f = f_{\text{sing}} + \mathbf{f}_t \hat{\sigma}$ and zero Matsubara frequencies

$$\begin{aligned} -i\hbar V_F \partial_s f_{\text{sing}} + 2\mathbf{h}\mathbf{f}_t &= 0, \\ -i\hbar V_F \partial_s \mathbf{f}_t + 2f_{\text{sing}}\mathbf{h} &= 0. \end{aligned} \quad (2)$$

Here, V_F is the Fermi velocity, s is the trajectory coordinate, and the function $f_{\text{sing}}(s_R) = \cos \gamma$ taken at the right S electrode determines the phase gain γ along the trajectory. The boundary conditions at the left electrode read $f_{\text{sing}}(s_L) = 1$, $\mathbf{f}_t(s_L) = 0$. Let us emphasize here that contrary to the standard consideration the Eilenberger-like equations in our approach are written along a random trajectory with many sharp turns and therefore they do not contain the impurity terms.

Summing up over all trajectories Γ we find the current:

$$I = \sum_{\Gamma} [j(\varphi + \gamma) + j(\varphi - \gamma)](\mathbf{n}_F, \mathbf{n}_L). \quad (3)$$

Here, $j(\chi) = \sum_{n \geq 1} (j_n/2) \sin(n\chi)$ is the trajectory contribution at a zero exchange field,

$$j_n = \frac{2eT}{\pi\hbar} \sum_{m=0}^{\infty} \int_0^{2\pi} d\chi \frac{\sin\chi \sin(n\chi)}{\mu_m + \cos\chi}, \quad (4)$$

and $\mu_m = 2\pi^2 T^2 (2m+1)^2 / \Delta_0^2 + 1$. The vectors \mathbf{n}_L and \mathbf{n}_F are the unit vectors normal to the left electrode surface and parallel to the trajectory direction, respectively. The vector \mathbf{n}_F parametrizes the trajectories outcoming from the left electrode. The random phase γ depends on the whole path between the electrodes and not just on the distance between the starting and ending points of the trajectory. Taking for simplicity the case of a homogeneous exchange field we find $\gamma = 2h(s_R - s_L) / \hbar V_F = \Omega t$, where t is the time of flight of the electron along the trajectory and $\Omega = 2h/\hbar$.

To average the above current expression over the random time of flight t we need to introduce the distribution function describing the probability density $w(\mathbf{r}_2, \mathbf{r}_1, t)$ to get the trajectory starting at a certain point \mathbf{r}_1 at the left electrode at the time $t_1 = 0$ and touching the right electrode at an arbitrary point \mathbf{r}_2 at the time $t_2 = t$. In the diffusion limit this probability density is almost independent on the quasiparticle velocity direction at the electrodes and satisfies the diffusion equation:

$$\frac{\partial}{\partial t} w = D \frac{\partial^2}{\partial \mathbf{r}_2^2} w + \delta(\mathbf{r}_2 - \mathbf{r}_1) \delta(t). \quad (5)$$

Here, we assume the elastic mean free path ℓ to be less than all the relevant length scales so that, in particular, $\ell \ll \xi_F$. The boundary condition should be defined from the fact that the trajectory that touches the S electrodes should be removed from the game. An obvious reason is that the corresponding electron moving along the trajectory experiences in this case the full Andreev reflection. Thus, at the surfaces of both S electrodes we should put $w = 0$. Choosing $\mathbf{r}_{1,2}$ at the left and right electrodes, respectively, we find the probability distribution $P(t)$ for the first-passage time between two electrodes:

$$P(t) = - \int_{S_R} D \left(\mathbf{n}_R \frac{\partial}{\partial \mathbf{r}_R} \right) w(\mathbf{r}_R, \mathbf{r}_L, t) dS_R, \quad (6)$$

where the integral is taken over the surface of the right electrode and \mathbf{n}_R is the unit vector normal to this surface. The value $P(t)$ gives the probability of the trajectory starting at the point \mathbf{r}_L at $t_1 = 0$ to leave the junction in the time interval from t to $t + dt$. The average current can be written as follows:

$$\langle I \rangle = \sum_{n \geq 1} N j_n \sin n\varphi \langle \cos n\gamma \rangle, \quad (7)$$

where $\langle \cos n\gamma \rangle = \text{Re} \int_0^\infty e^{-in\Omega t} P(t) dt = \text{Re} P(n\Omega)$. We assume here the surfaces of the S electrodes are flat and obtain a one-dimensional problem along the coordinate x perpendicular to these surfaces. Introducing the function $W(x, t)$ satisfying the 1D diffusion equation $DW''_{xx} - in\Omega W = 0$ with the boundary conditions $DW(x=0) = \ell$

and $W(x=d) = 0$ one can find $P(n\Omega) = DW'_x(x=d, n\Omega)$. Substituting the solution of the above diffusion equation into the current we obtain

$$\langle I \rangle = \text{Re} \sum_{n \geq 1} N j_n \sin n\varphi \frac{\ell \sqrt{in}}{\xi_F} \frac{1}{\sinh[\sqrt{in}d/\xi_F]}. \quad (8)$$

One can see that this expression reproduces the result of the Usadel theory only for the first harmonic $I_1 \propto \sin\varphi$ in the current-phase relation [5]. The length L_n of the exponential decay of higher harmonics $I_n \propto \sin n\varphi$ appears to exceed the appropriate length in the Usadel-type calculation: we obtain here $L_n = \xi_F/\sqrt{n}$ instead of $L_n = \xi_F/n$. This result indicates an obvious increase of the range of superconducting correlations due to mesoscopic fluctuations and originates from the incorrect calculation of the ensemble averages of the product of the anomalous Green functions in the ferromagnet within the Usadel theory. This failure of the Usadel-type consideration is caused by the appearance of the random interference phase γ and occurs only in the nonlinear regime of rather strong superconducting correlations. Indeed, considering, e.g., the value $\langle \cos 2\gamma \rangle$ in the above derivation we calculate the average $\langle |f_{\text{sing}}|^2 - |f_t|^2 \rangle$, which definitely differs from the product of averages $\langle f_{\text{sing}} \rangle \langle f_{\text{sing}}^* \rangle - \langle f_t \rangle \langle f_t^* \rangle$. The mesoscopic fluctuations affect the validity of the nonlinear Usadel equations also for S - N - S junctions [29], providing an exponentially small contribution to the density of states (DOS) below the minigap. Note that the above approach describes the fluctuation contributions that do not vanish in the limit $\lambda_F \rightarrow 0$ and can, thus, exceed the subminigap DOS corrections found previously in Ref. [22] on the basis of the nonlinear sigma model. Our contributions are caused by the quantum interference effects associated with a much larger wavelength of the quasiparticle wave function envelope: $\hbar V_F/E$ or $\hbar V_F/h$ for S - N - S and S - F - S systems, respectively.

To find the rms value of the supercurrent we evaluate now the expression

$$\langle I^2 \rangle = \sum_{\Gamma, \tilde{\Gamma}, n, m} j_n j_m A_{nm}(\mathbf{n}_F, \mathbf{n}_L)(\tilde{\mathbf{n}}_F, \tilde{\mathbf{n}}_L) \sin n\varphi \sin m\varphi, \quad (9)$$

where $A_{nm} = \langle \cos n\Omega t \cos m\Omega \tilde{t} \rangle$. The calculation of the above double sum can be done similarly to the calculation of the conductance $R^{-1} = G(d, \ell)$ in a dirty wire above T_c . Assuming the normal layer thickness to be rather large ($d \gg \xi_F$) and omitting the averages of the fast oscillating phase factors (which should give the short-range terms decaying at the length ξ_F) we get

$$\langle I^2 \rangle \approx [G(\tilde{d}, \tilde{\ell})/4G_0] \sum_{n \geq 1} j_n^2 \sin^2 n\varphi, \quad (10)$$

where $G_0 = e^2/\pi\hbar$, $\tilde{d} = \Omega d/k_F V_F$, and $\tilde{\ell} = \Omega \ell/k_F V_F$. Taking the Drude-type conductance $G/G_0 = N\ell/d$ for a disordered wire of the length d we find the estimate

$$\sqrt{\langle I^2 \rangle - \langle I \rangle^2} \sim \sqrt{\frac{N\ell}{d}} \sqrt{\sum_{n \geq 1} j_n^2 \sin^2 n\varphi}. \quad (11)$$

The deviations from the Drude result arise naturally from the so-called interference or localization corrections to the conductance [28]. Perturbatively, they can be estimated as terms arising from the paths with self-crossings in the above double sum over the trajectories. According to the Thouless criterion [34], the localization effects in a disordered wire are small provided the effective number $N\ell/d$ of conducting modes is large. Thus, one can expect our Drude-type estimate to hold in the case $N\ell/d \gg 1$. In the opposite limit, the wire conductance in Eq. (10) and, thus, the rms value of the critical current decay exponentially at the length $N\ell$.

Comparing the rms value with the average current taken in the same limit $d \gg \xi_F$ we find

$$\delta I / \langle I \rangle \sim \sqrt{\frac{\xi_F^2}{N\ell d}} \exp\left(\frac{d}{\xi_F \sqrt{2}}\right). \quad (12)$$

This expression for the current fluctuations definitely cannot be obtained within the averaged Usadel theory and results from the partial cancelation of the interference contributions in the product of the anomalous Green functions. Note that turning to the limit $d \ll \xi_F$, i.e., to the case of the S - N - S junction, our consideration should give a vanishing δI value since we disregarded the quantum interference of random semiclassical trajectories responsible for standard mesoscopic fluctuations [24]. Despite the small factor $N^{-1/2}$ in Eq. (12) the current fluctuations for $d \gg \xi_F$ appear to be giant compared to the current average value, which decays exponentially at the small distance ξ_F . The rms value can well exceed the Josephson current quantum $e\Delta/\hbar$ in S - N - S junctions [24]. It is also important to note that contrary to the average current the fluctuating contributions to higher harmonics of the current-phase relation are not suppressed exponentially compared to the first harmonic. This strong anharmonicity probably relates to the experimental data on the large second harmonics in S - F - S junctions [35,36]. Certainly, in realistic junctions the above assumption of the full Andreev reflection at the S - F boundaries can be broken due to the effect of the interface potential barriers, which certainly suppress the higher current harmonics. Still the main effect, namely, the partial compensation of the phases γ in the rms values should exist even in the presence of the barriers.

One can easily see that the above long-range behavior of the critical current fluctuations holds also beyond the short junction approximation (i.e., for $d > \xi_S$) at least for the first harmonic $I_1(\varphi)$. Indeed, the critical current in this limit is determined by the singlet component of the anomalous Green function $\sum_{\Gamma} f_{\text{sing}\Gamma} = \sum_{\Gamma} \cos \gamma_{\Gamma}$. The average current, therefore, decays exponentially as $\langle I \rangle \propto (\ell N / \xi_F) e^{-d/\xi_F \sqrt{2}}$ while the rms average becomes long

ranged because of the partial phase compensation at close trajectories: $\langle (\delta I)^2 \rangle \propto \langle f_{\text{sing}}^2 \rangle \propto N\ell/d$. Thus, the above calculations confirm the estimate (1) both for short and long junctions. Certainly, a further increase in the distance d will give us the exponential decay of the supercurrent but at the distances $\gtrsim \xi_N$.

The mesoscopic fluctuations considered in our work should be most easily observed in ferromagnetic wires because their relative contribution decays with the increase of the N number. Let us perform the quantitative comparison of our theory with the experimental data of Ref. [13], which has reported on the observation of the critical current $I_c \sim 10 \mu\text{A}$ for a Co nanowire with length $d = 0.6 \mu\text{m}$ and diameter $2r = 40 \text{ nm}$ at the temperature $T = 1.8 \text{ K}$. This temperature is much smaller than the critical temperature of W electrodes $T_c \sim 5 \text{ K}$ so we may use the low temperature limit for j_n and Eq. (10) transforms into the estimate $\delta I \sim \Delta_0 / 2\sqrt{\pi\hbar R}$. We take the resistivity $\rho = 10 \mu\Omega \text{ cm}$ obtained in Ref. [13] for a Co nanowire with the same diameter and the length $d = 1.5 \mu\text{m}$ and find $R \sim 4 \Omega$. Taking the gap $\Delta_0 \sim 1.74T_c \sim 8 \text{ K}$ we finally get the value $\delta I \sim 1 \mu\text{A}$, which is only an order of magnitude less than the critical current observed in Ref. [13]. The remaining discrepancy is probably caused by the overestimating of the wire resistance R due to the presence of contact resistances due to the interdiffusion of W at the distance $\sim 200 \text{ nm}$. It is also useful to compare the fluctuation contribution with the possible effect of spin-triplet correlations, which can still appear, e.g., due to some noncollinearity of the magnetic moments at the interfaces. Introducing such thin noncollinear domains of thickness $d_i \ll \xi_F$ at the left and right ends of the ferromagnetic wire one obtains the long-range current contribution in the form [37] $I_{\text{tr}} \sim (d_i/\xi_F)^4 \Delta_0 / eR$. One can see that for the wires with rather large resistances $R > G_0^{-1} (d_i/\xi_F)^4$ the fluctuation contribution dominates.

We now briefly comment on the effect of mesoscopic fluctuations on the local DOS (LDOS) at the Fermi level. In the ballistic system for straight linear trajectories one can easily obtain an Eilenberger-type expression for this quantity as a sum of contributions from different quasiclassical paths. This expression can be simplified applying the normalization condition for quasiclassical Green functions and taking the perturbation expansion in powers of the f function (see, e.g., Ref. [38] for convenient notations). Generalizing this expression for the trajectories experiencing many sharp turns one can get $\delta\nu/\nu_F \propto -N^{-1} \sum_{\Gamma} (|f_{\text{sing}}|^2 - |f_t|^2)$, where ν_F is the normal-metal LDOS. The ensemble average of this value certainly decays exponentially, $\langle \delta\nu/\nu_F \rangle \propto -\langle \cos 2\gamma \rangle \propto -(\ell/\xi_F) e^{-d/\xi_F} \cos(d/\xi_F + \pi/4)$, with the increase in the distance d from the S electrode. The fluctuating LDOS contains a long-range contribution similar to the one calculated above for the critical current: $\sqrt{\langle (\delta\nu/\nu_F)^2 \rangle} \propto \sqrt{\ell/dN}$. This nonexponential behavior of the fluctuating

superconducting contribution to the LDOS could be measured by a local conductance probe at different points of a ferromagnetic nanowire placed in contact to a superconductor providing, thus, a possible explanation of the long-range proximity effect observed in Refs. [6–8].

The direct observation of the giant sample-to-sample fluctuations assumes the measurements of the critical current or LDOS on different junctions. It would be much more convenient to find the way to change the interference phases γ in a given sample and measure the junction “fingerprints” in analogy to the observation of universal conductance fluctuations versus an applied magnetic field [39]. Indeed, such a type of experiment in the S - F - S junctions may become possible provided we apply a magnetic field that can affect the domain structure in the F layer without producing noncollinear magnetic regions to avoid the admixture of the long-range triplet correlations.

To sum up, we suggest a theoretical model describing the mesoscopic fluctuations in S - F and S - N systems. This model allows us to obtain a huge fluctuation contribution in the supercurrent in S - F - S junctions and the LDOS, which survives the destructive effect of the exchange field and could, thus, provide an explanation of the experimental data on the long-range proximity phenomena in ferromagnetic wires even in the absence of the exchange field inhomogeneity. The resulting fluctuating supercurrent is a sign-changing quantity and, thus, the ensemble fluctuations cause the appearance of both zero and π junctions. Our analysis reveals also that fluctuations can be responsible for anomalously large values of the second and higher harmonics in the current-phase relation.

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