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LETTERS TO THE EDITORS

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A comment on the paper "Competition between superconductivity and magnetism in ferromagnet/superconductor heterostructures" by Yu A Izyumov, Yu N Proshin, and M G Khusainov

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In their recent review [1], Izyumov, Proshin, and Khusainov examined the proximity effect between a superconductor and a ferromagnet.

Ordinary (singlet) superconductivity and ferromagnetism are two competing types of ordering: while singlet superconductivity is accompanied by formation of Cooper pairs, in which the electron spins are antiparallel, ferromagnetic order presupposes that the electron spins are parallel.

In view of this, the coexistence of both orders in a single substance is possible within a very narrow range of parameters [2, 3]. At the same time, the study of the interrelationship of superconductivity and ferromagnetism is possible in a situation where their 'sources' are separated in space, which leads to the proximity effect in FM/S systems (FM stands for a ferromagnetic metal and S for a superconductor). In such a system, due to separation of the spin subbands in the ferromagnet, a superconducting state that is nonuniform (along the normal to the boundary) and is similar to the Larkin – Ovchinnikov – Fulde – Ferrell (LOFF) state [2, 3] is realized.

One of the fundamental problems is that of T_c, the critical temperature of the FM/S junction, in the case of finite layer thicknesses. The review [1] focuses on this problem. Certain remarks concerning the methods used in solving it are in order, however.

1. 3D LOFF states

In studying the problem of the critical temperature of an FM/S junction, the authors of Ref. [1] pay much attention to

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the so-called *three-dimensional* (3D) LOFF states, which were proposed by them in earlier papers (see Refs [4, 5]).

However, the assumption that such states exist is based on an erroneous interpretation of the Kupriyanov – Lukichev (KL) boundary conditions [6] imposed on the Green's functions on both sides of the mirror boundary. Near T_c, the KL boundary conditions are linearized and assume the form (in the notation of Ref. [1])

$$\frac{4D_s}{\sigma_s v_s}\frac{\partial F_s(\boldsymbol{\rho},z,\omega)}{\partial z}\bigg|_{z=+0} = \frac{4D_f}{\sigma_f v_f}\frac{\partial F_f(\boldsymbol{\rho},z,\omega)}{\partial z}\bigg|_{z=-0} = F_s(\boldsymbol{\rho},z=+0,\omega)-F_f(\boldsymbol{\rho},z=-0,\omega)\,, \qquad (1)$$

where F is the anomalous Green's function, $\boldsymbol{\rho}$ is a two-dimensional vector in the plane of the junction, and the z axis is directed along the normal to the boundary. Conditions (1) must be met at all points of the boundary, i.e., for all $\boldsymbol{\rho}$. Performing a Fourier transformation in $\boldsymbol{\rho}$, we obtain

$$\frac{4D_s}{\sigma_s v_s}\frac{\partial F_s(\mathbf{q},z,\omega)}{\partial z}\bigg|_{z=+0} = \frac{4D_f}{\sigma_f v_f}\frac{\partial F_f(\mathbf{q},z,\omega)}{\partial z}\bigg|_{z=-0} = F_s(\mathbf{q},z=+0,\omega)-F_f(\mathbf{q},z=-0,\omega)\,, \qquad (2)$$

where the arguments of the Green's functions contain the *same* two-dimensional wave vector $\mathbf{q}$.

At the same time, the 3D LOFF states proposed by the authors of the review were derived from conditions (3.23) of Ref. [1], which differ from the KL boundary conditions by the assumption that the two-dimensional momenta on both sides of the boundary may be different: $\mathbf{q}_s = 0$ and $\mathbf{q}_f \neq 0$. A microscopic derivation of these boundary conditions is not given in Ref. [1]; moreover, as far as we know, it is not given in the earlier works of the authors of the review.

The assumption that $\mathbf{q}_s \neq \mathbf{q}_f$ means that we reject the local nature (in coordinate $\boldsymbol{\rho}$) of the boundary conditions (1). Although in their paper [6] Kupriyanov and Lukichev examined a quasi-one-dimensional situation, where all quantities vary only along the z axis, the KL boundary conditions can easily be generalized to a general three-dimensional case. Indeed, the Zaĭtsev boundary conditions [7] for the Eilenberger equations are three-dimensional and local; they can be interpreted as one-dimensional boundary conditions (along the z axis) that must be met at *each point* $\boldsymbol{\rho}$ of the boundary. The KL boundary conditions for the Usadel equations are derived from the Zaĭtsev boundary conditions. Hence, although Kupriyanov and Lukichev studied the one-dimensional case, it is clear that the generalization of the boundary condition that they derived to the three-dimensional case is trivial: we must only require that the one-dimensional KL boundary conditions be met at each point of the boundary, which leads to equations (1).

Thus, the boundary conditions (3.23) from Ref. [1] in which $\mathbf{q}_s \neq \mathbf{q}_f$ contradict the KL boundary conditions and are invalid, which makes the 3D LOFF states discussed by the authors of the review a corollary of this error.

The fact that the boundary conditions (3.23) of Ref. [1] are incorrect for $\mathbf{q}_s \neq \mathbf{q}_f$ manifests itself most vividly in the case of an ideal boundary (i.e., for $\sigma_s, \sigma_f \to \infty$). Here, as a result of an inverse Fourier transformation, we arrive at a situation where the anomalous Green's function experiences a discontinuity (whose size varies along the boundary) as the boundary is crossed. However, from quantum mechanical ideas it follows that in the case where the transparency of the boundary is equal to unity the Green's functions must be continuous in view of the continuity of the electron wave functions from which the Green's functions are built.

The true critical temperature T_c was defined in Ref. [1] as the highest critical temperature of the ordinary (1D) and 3D LOFF states. The authors of the review found that it is highly important to take into account the 3D states, since in the overwhelming range of parameters the superconducting transition occurs exactly in a 3D LOFF state (e.g., see Figs 5 and 10 in Ref. [1]).

However, as shown in our commentary, the 3D LOFF states with $\mathbf{q}_s \neq \mathbf{q}_f$ follow from invalid boundary conditions and cannot be physically implemented, with the result that they should be ignored in the definition of T_c. Thus, the results concerning the critical temperature obtained by the authors of the review [1] with allowance for 3D LOFF states are erroneous.

2. Complex-valued diffusion coefficient

The authors of the review [1] examined what is known as the 'dirty limit,' so that the critical-temperature problem is described by linearized Usadel equations [8], which contain the isotropic part of the anomalous Green's function, F_0. For the Usadel equations to be valid in a ferromagnet, the following necessary condition must be met:

$$2I\tau_f \ll 1\,, \tag{3}$$

where I is the exchange energy, and τ_f is the mean free time. Within this range, the authors of the review use a complex-valued diffusion coefficient (see Eqn (3.7) in Ref. [1], with only positive Matsubara frequencies considered), which has the form

$$D_f(I) = \frac{D_f}{1 + \mathrm{i}2I\tau_f}\,, \tag{4}$$

where

$$D_f = \frac{v_f l_f}{3} \tag{5}$$

is the ordinary diffusion coefficient. Expression (4) has been used by the authors of the reviews in Refs [4, 5, 9–11] and by other researchers (see Refs [12, 13]).

However, in Ref. [14] it was shown that although expression (4) for the diffusion coefficient can be formally obtained through the standard derivation of the Usadel equations from the Eilenberger equations (for $I \gg \pi T_{cs}$), nevertheless the complex-valued correction to the diffusion coefficient leads to excessive accuracy and must be discarded.

Indeed, the Usadel equation contains F_0, the isotropic part of the Green's function. At the same time, the total angular dependence of the Green's function is given by an expansion in Legendre polynomials

$$F(\mathbf{r}, \omega, \theta) = \sum_{k=0}^{\infty} F_k(\mathbf{r}, \omega)\, P_k(\cos\theta) \tag{6}$$

(where the angle θ describes the direction of the momentum of the relative motion of the electrons in a Cooper pair), and to derive the Usadel equations from the Eilenberger equations we must keep the isotropic part F_0 and the first angular harmonic F_1. The higher harmonics beginning with F_2 are discarded, since their order of smallness (at $I \gg \pi T_{cs}$) [14] is

$$\left|\frac{F_2}{F_0}\right| \sim 2I\tau_f\,. \tag{7}$$

Thus, the complex-valued correction to the denominator in (4) is of the same order of smallness as the higher angular harmonic discarded in the standard derivation of the Usadel equations. Hence, in the standard derivation of the Usadel equations this correction should also be discarded.

This statement, however, is not the final word on the problem of the complex-valued diffusion coefficient in a ferromagnet [15]. The thing is that in a more exact derivation that allows for the second harmonic F_2, in the case where $I \gg \pi T_{cs}$ we indeed arrive at a Usadel equation

$$\frac{D_f(I)}{2}\,\frac{\mathrm{d}^2}{\mathrm{d}\mathbf{r}^2}\,F_0 - \mathrm{i}IF_0 = 0 \tag{8}$$

with a complex-valued diffusion coefficient, but the factor at the imaginary correction is 2/5 instead of 2:

$$D_f(I) = D_f\left(1 - \mathrm{i}\,\frac{2}{5}\,I\tau_f\right) \tag{9}$$

(the imaginary correction must be written in the numerator rather than in the denominator, since here there is an expansion in powers of a small parameter). Keeping the complex-valued correction here is justified, since it corresponds to the accuracy of the derivation. The result (9) was first obtained by Tagirov and Buzdin [15] in their discussion of the problem of a complex-valued diffusion coefficient with the authors of Ref. [14]. The derivation of this result can be found in a recent paper by Buzdin and Baladié [16].

Thus, the complex-valued diffusion coefficient in the Usadel equation for a ferromagnet must have the form of (9) instead of (4), i.e., the magnitude of the complex-valued correction must be five times as small as that in Ref. [1]. In view of this, the results of the review [1] for the case (3), where this correction provides a sizable contribution, are incorrect.

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