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On existence of a paramagnetic contribution to the susceptibility of a mesoscopic cylindrical normal metal-superconductor structure

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The density of states $N(\varepsilon)$ of a mesoscopic cylindrical structure consisting of a normal pure metal and a superconductor has been calculated using the Gorkov–Green functions. It is shown that magnetic fluxes of certain values cause resonance spikes of $N(\varepsilon)$ suggesting a large-amplitude paramagnetic contribution which accounts for the reentrant effect detected (P. Visani, A.C. Mota, and A. Pollini, *Phys. Rev. Lett*. **65**, 1514 (1990)).

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1. Introduction

The technological advance in preparation of pure samples has enabled investigations of the coherent properties of mesoscopic structures taking into account the proximity effect [1]. The samples were superconducting (S) Nb wires (cylinders with a radius of tens of microns) coated with a thin layer of a normal (N) pure metal (Cu or Ag). The structure was placed in a weak magnetic field. A.C. Mota et al. [2,3] who measured the magnetic susceptibility χ of such structures could observe its rather surprising reentrant-type behavior: at lowering temperatures the (diamagnetic) susceptibility (in a constant field) changed in accordance with theory but it unexpectedly started growing at $T < 100$ mK. A similar behavior was observed with the isothermal reentrant effect in a decreasing magnetic field: the susceptibility started to grow sharply in a held decreasing below a certain value. The effect was observed only on mesoscopic NS structures. The authors [2] interpreted the discovered phenomenon as a new coherent quantum effect in pure NS structures. They assumed that a paramagnetic contribution could appear for some reason in the NS structure in addition to the diamagnetic current.

The origin of the paramagnetic currents in NS structures has been discussed in several theoretical publications. Bruder and Imry [4] analyze the paramagnetic contribution to susceptibility made by quasiclassical («glancing») trajectories of quasiparticles that do not collide with the superconducting boundary. The authors [4] point to a large paramagnetic effect within their physical model. However, their ratio between the paramagnetic and diamagnetic contributions is rather low and cannot account for the experimental results [2,3].

Fauchere, Belzig, and Blatter [5] explain the large paramagnetic effect assuming a pure repulsive electron-electron interaction in noble metals. The proximity effect in the N metal induces an order parameter whose phase is shifted by π from the order parameter Δ of the superconductor. This generates the paramagnetic instability of the Andreev states, and the density of states of the NS structure exhibits a single peak near the zero energy. The theory in Ref. 5 essentially rests on the assumption of the repulsive electron interaction in the N metal. Is the reentrant effect a result of specific properties of the noble metals, or does it display the behavior of any normal metal experiencing the proximity effect from the neighboring superconductor? Only experiment can provide answers to these questions. We just note that the theories of Refs. 4 and 5 do not account for the temperature and field dependences of the paramagnetic susceptibility and the nonlinear behavior χ of the NS structure. The current theories cannot explain the origin of the anomalously large paramagnetic reentrant susceptibility in the region of very low temperatures and weak magnetic fields.

It is worth mentioning the assumption made by Maki and Haas [6] that below the transition temperature (~ 10 mK) some noble metals (Cu, Ag, Au) can exhibit p-ware superconducting ordering, which may be responsible for the reentrant effect. This theory does not explain the high paramagnetic reentrant effect either.

Note that the NS structure generally has three contributions to its magnetic susceptibility. First, this is a diamagnetic response induced by the electron excitation specularly reflected from the dielectric boundary and scattered (Andreev scattering) at the NS boundary. The Andreev levels form in the film [7] when the normal layer thickness is small. The peculiar feature of the quantum proximity effect is the magnetic susceptibility diamagnetism (Meissner effect) modified by the Andreev levels.

Another contribution is related to the electron trajectories that do not collide with the NS boundary («whispering mode»). They generate persistent current in the normal layer and make a weak paramagnetic contribution to the magnetic moment of the system.

Finally, a large-amplitude persistent paramagnetic current is induced in the normal layer due to the Aharonov–Bohm effect [8] caused by strong degeneracy of the system when the Andreev level superimposes on the Fermi level of the metal. In this case resonance spikes are observed in the density of states. The spectrum of quasiparticles (sec Eq. (4)) includes an angle α at which they hit the dielectric boundary. For a pre-assigned α-value, resonance occurs at a certain magnetic flux through the quantized area enclosed by the trajectory. For other angles α the resonance-inducing fluxes are slightly different. The total contribution to the density of the states of a NS structure is a sum of contributions from all trajectories. It is found [9,10] that the high paramagnetic response can occur in a certain range of weak magnetic fields and at temperatures no higher than 100 mK.

We obtained a large paramagnetic contribution χ^{p} to the susceptibility of a NS structure within the model of free electrons. When χ^{p} is added to the diamagnetic contribution χ^{d}, the resulting total susceptibility features the reentrant effect. Theoretically [9,10] the effect is expected in samples in which quasiparticles have large mean free paths comparable with the cylindrical N layer perimeter. This was observed experimentally in Refs. 2,3.

As the magnetic field (or temperature) increases and eliminates a prerequisite to resonance, the large paramagnetic contribution disappears. The paramagnetic contribution from the «whispering mode» persists but it is small due to the smallness of the quasiclassical parameter of the problem $\sim 1/(k_F R)$ ($\hbar k_F$ is the Fermi momentum, R is the radius of the cylinder) and cannot effect the total susceptibility of the system.

When the magnetic field increases considerably (or the temperature approaches T_c of the superconductor) the diamagnetic susceptibility has only the «classical» contribution from the motion of the Cooper pains inside the superconducting layer near the NS boundary. This contribution persists in the whole range of temperatures (and magnetic fields) where the superconducting state exists.

The Meissner effect in pure NS structure has been investigated recently by Galaktionov and Zaikin [11] who used the Gorkov microscopic equations of superconductivity [12]. They calculated the diamagnetic current of a NS structure taking into account the proximity effect. The result obtained is essentially similar to that in Zaikin's first publication on the subject [13]. At the same time it was stated [11] that a paramagnetic contribution to the susceptibility of a NS structure is absent if there is no strong electron–electron repulsion in the N layer.

The goal of this study is to show the existence of a paramagnetic contribution to the susceptibility of a NS structure using the Green function approach. The density of states $N(\varepsilon)$ of such a structure has been calculated. It is shown that magnetic flux of certain values induce resonance spikes of $N(\varepsilon)$. The model of free electrons was applied.

Note that a large paramagnetic contribution is unobtainable with the Eilenberger–Green functions [13]. The Eilenberger equations [14] were derived by integrating the Gorkov microscopic equations of superconductivity with respect to the quasipartieles energies. The remarkable effect of quasiparticle state degeneracy occurs in a narrow interval of energies approximately equal to the energy gap of the superconductor (see above). This is the interval in which the density of states of a NS structure exhibits resonance features. The integration of the Gorkov Green functions over energies reduces the large contribution from the quantized Andreev levels to its average value. As a result the paramagnetic contribution to the thermodynamics of the NS structure disappears. To detect this contribution one should proceed from the exact Gorkov equations for the Green functions of a contact in a magnetic field.

2. The density of states of a NS structure

A mesoscopic cylindrical NS contact consisting of a thin pure normal-metal layer ($0 < x < d$) and a bulk superconductor has been considered (see Fig. l). We neglect the curvature of the cylindrical NS surface, which is permissible if $R >> d$. The assumption of a flat NS boundary largely simplifies the consideration, whereas the value of the screened current is practically similar for cylindrical and flat geometries. The currents are distinctive in parameter $\sqrt{d / R} << 1$ (see [11]). We proceed from the free-electron model and assume a stepwise variation of the order parameter at the NS boundary. The mean free paths of the

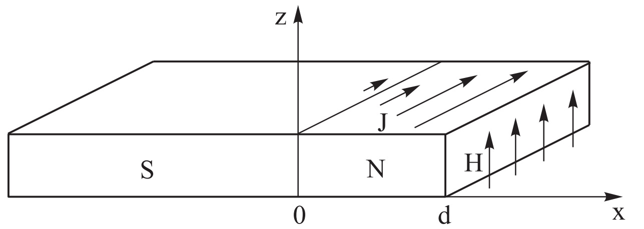

Fig. 1. Normal-metal layer (thickness d) in proximity to a bulk superconductor. A magnetic field H parallel to the surface is applied, driving screening current J along the surface.

quasiparticle exceed the characteristic dimensions of the structure.

The density of states of a NS contact, is found as

$$N(\varepsilon) = -\frac{1}{\pi}\,\mathrm{Im}\,\mathrm{Sp}\,G_{11}^{R}(\varepsilon + i\gamma). \qquad (1)$$

The Spur operation is performed over the variables determining the Green matrix function. G_{11}^{R} is the analytical continuation of the 11-th component of the Green function $\gamma \to 0$. It is assumed that the NS structure is homogeneous along the interface.

The Fourier transformation of the normal (G) and anomalous (F^{+}) Green functions in the coordinates along the NS boundary yields

$$G_{\omega_n}(\mathbf{r},\mathbf{r}') = \int \frac{d^2\mathbf{k}_{\parallel}}{(2\pi)^2} G_{\omega_n}(x, x', \mathbf{k}_{\parallel})\, e^{i\mathbf{k}_{\parallel}(\mathbf{r}_{\parallel}-\mathbf{r}'_{\parallel})}.$$

The Gorkov equations become [12]:

$$\begin{pmatrix} i\omega_n - \hat{H} & \Delta(x) \\ \Delta^*(x) & i\omega_n + \hat{H}_c \end{pmatrix} \begin{pmatrix} G_{\omega_n}(x, x', \mathbf{k}_{\parallel}) \\ F^{+}_{\omega_n}(x, x', \mathbf{k}_{\parallel}) \end{pmatrix} = \begin{pmatrix} \delta(x - x') \\ 0 \end{pmatrix}. \qquad (2)$$

Here $\omega_n = (2n+1)\pi T$ is the Matsubara frequency; $\Delta(x)$ is the superconducting order parameter. The Hamiltonian $\hat{H}$ is determined as $\hat{H} = -\frac{1}{2m^*}\frac{\partial^2}{\partial x^2} + \frac{\tilde{\mathbf{k}}_{\parallel}^2}{2m^*} - \mu + V(x)$, $\mathbf{A}$ is the vector potential field; $\tilde{\mathbf{k}}_{\parallel} = \mathbf{k}_{\parallel} - \frac{e}{c}\mathbf{A}_{\parallel}(x)$; μ — is the Fermi energy and $V(x)$ is the potential of the metal boundary. $\hat{H}_c$ is found from $\hat{H}$ by reversing the sign of the electron charge e. It is assumed that the London penetration depth of the field is small in a bulk superconductor and the field inside the superconductor of our structure is zero.

The Green function of a NS structure was calculated for pure metals in a zero magnetic field by Arnold [15]. The Green function of the composite structure was found from the Green functions of isolated systems $G_N(x, x')$ and $G_S(x, x')$ and was then used to calculate the tunnel density of states at the NS boundary. The Green function of a NS structure in a magnetic field was obtained by Galaktionov and Zaikin [11]:

$$G_{\omega_n}(x, x', \mathbf{k}_{\parallel}) = \frac{-2i}{\pi v_x}\,\mathrm{th}\,\chi\,, \qquad (3)$$

where $\chi = \frac{2\omega_n d}{v_x} - i\tan\alpha\phi$. Here $\phi = \frac{2\pi}{\phi_0}\int_0^d A(x)dx$, v_x is the velocity component along the normal to the NS boundary, α is the angle at which a quasiparticle hits the dielectric boundary of the normal layer, $\phi_0 = hc/2e$ is the superconducting flux quantum.

It is seen that the function $G_{\omega_n}(x, x', \mathbf{k}_{\parallel})$ is constant in x but dependent on the magnetic flux. Such dependence disappears in a bulk metal ($d \to \infty$). If the normal metal has a finite thickness, its electron states are obtainable from the poles of the Green function. For low-lying states the spectrum of the Andreev levels in the N layer is

$$\varepsilon_n(q, \alpha; \phi) = \frac{\pi v(q)\cos\alpha}{2d}\left(n + \frac{1}{2} - \frac{\tan\alpha}{\pi}\phi\right). \qquad (4)$$

Here $v(q) = \sqrt{p_F^2 - q^2}\,/\,m^*$, v_F is the Fermi velocity, q is the momentum component along the symmetry axis of the cylinder, m^* is the effective mass of a quasiparticle, $n = 0, \pm 1, \pm 2, \ldots$

$A(x)$ is found by solving the Maxwell equation for the boundary condition $A(x=0) = 0$, $\frac{\partial A}{\partial x}|_{x=d} = H$ [13]:

$$A(x) = Hx + \frac{4\pi}{3c}\, j\left(d - \frac{x}{2}\right)x, \qquad (5)$$

where j is the current density in the N layer. We assume that this current includes both the diamagnetic and paramagnetic components. Integration of both sides of Eq. (5) over the normal layer thickness gives a sell-consistent equation for $a \equiv \int_0^d A(x)dx$:

$$a = \frac{Hd^2}{2} + \frac{4\pi}{3c}\, j(a) d^3.$$

The parameter a is dependent both on the magnetic field and the temperature.

The expression for the density of states depends from the Green function of the structure. Substituting Eq. (3) in the Green function and using its analytical continuation, we can obtain after summation over the spin

$$N(\varepsilon) = \frac{4m^*}{(2\pi)^2\pi^2}\,\mathrm{Im}\sum_n \int_{-p_F}^{p_F} dq \int_{-\alpha_c}^{\alpha_c} d\alpha \quad \tan\left[\frac{2d\varepsilon}{v_x} + \tan\alpha\phi\right].$$

The angle α is measured from the positive direction of the normal to the boundary. The spectrum of the Andreev levels (Eq. (4)) is formed by the quasiparticle paths in the N layer whose angles vary within $0 \lesssim |\alpha| \lesssim \alpha_c$. α_c is the angle at which the quasiparticle trajectory touches the NS boundary [10]. Those trajectories are responsible for a large paramagnetic contribution to the susceptibility and hence for the reentrant effect. Another group includes the trajectories with $\alpha > \alpha_c$ that collide only with the dielectric boundary. They induce states practically coinciding with the «whispering gallery» type of states occurring in the cross-section of a normal solid cylinder in a weak magnetic field [16]. These trajectories generate paramagnetic contribution of small amplitudes (see the Introduction) and are therefore discarded from this consideration.

The main contribution to the density of states comes from the vicinity of the tangent poles. Expanding the numerator and denominator into a series near the Andreev levels, we obtain ($\gamma \to 0$)

$$N(\varepsilon) = -\frac{2}{\pi^3 d} \operatorname{Im} \sum_n \int_0^{p_F} dq \sqrt{p_F^2 - q^2} \int_0^{\alpha_c} \frac{d\alpha}{\varepsilon - \varepsilon_n + i\gamma} . \quad (6)$$

We then use the relation

$$\frac{1}{x \pm i\gamma} = P\frac{1}{x} \mp i\pi\delta(x),$$

where P is the principal quantity and obtain an expression similar to that in [10]:

$$N(\varepsilon) = +\frac{2}{\pi^2 d} \sum_n \int_0^{p_F} dq \sqrt{p_F^2 - q^2} \int_0^{\alpha_c} d\alpha \;\; \delta[\varepsilon - \varepsilon_n(q,\alpha;\phi)]. \quad (7)$$

The density of states can be written as

$$N(\varepsilon) = \int_0^{\alpha_c} d\alpha \nu(\varepsilon;\alpha),$$

where $\nu(\varepsilon;\alpha)$ is the contribution to the density of states from the pre-assigned trajectory with a fixed α. After integration with respect to q and introduction of the notation $\beta = \pi\hbar/(2dm^*)$, we can pass on to the dimensionless energy $\epsilon = \varepsilon / \beta p_F$. We obtain the expression for $\nu(\epsilon;\alpha)$

$$\nu(\epsilon;\alpha) = \frac{2p_F}{\pi^2 \beta d} \epsilon^2 \sum_n \frac{\sec^2\alpha\theta\,[n + \kappa - \epsilon \sec\alpha]}{(n+\kappa)^2 \sqrt{(n+\kappa)^2 - \epsilon^2 \sec^2\alpha}} , \quad (8)$$

where $\kappa = \frac{1}{2} - \frac{\tan\alpha}{\pi}\phi$ and $\theta(x)$ is the Heaviside step function. Equation (8) suggests two cases depending on the parameter $n + \kappa$.

Nonresonance case

If $n + \kappa \neq 0$ the energy dependence under the radical sign in Eq. (8) can be neglected for small energies. Then, the nonresonance contribution to the density of states is

$$\nu^{(0)} \sim \frac{2p_F}{\pi^2 \beta d} \epsilon^2 \int_0^{\alpha_c} d\alpha \sum_{n=-\infty}^{+\infty} \frac{\sec^2\alpha}{(n+\kappa)^3} . \quad (9)$$

The series in Eq. (9) is calculated by the formula of Ref. 17:

$$\sum_{k=-\infty}^{+\infty} \frac{1}{(k+\kappa)^n} = (-1)^{n-1} \frac{\pi}{(n-1)!} \frac{d^{n-1}}{d\kappa^{n-1}} \cot \pi\kappa,$$

which yields

$$\nu^{(0)} \sim \frac{2p_F \pi}{\beta d} \epsilon^2 \int_0^{\alpha_c} d\alpha \frac{\sin[\tan\alpha\phi]}{\cos^2\alpha \cos^3[\tan\alpha\phi]} . \quad (10)$$

After integration over α we have

$$\nu^{(0)} \sim 4k_F m^* \frac{\epsilon^2 \phi_0}{2\pi a} \tan^2\left[\frac{2\pi a}{\phi_0}\sqrt{\frac{2R}{d}}\right],$$

where $\sqrt{2R/d} \simeq \tan\alpha_c$.

Resonance case

Now we go back to Eq. (8) and find ν^{res} as

$$\nu^{\text{res}} = \frac{4k_F m^*}{\pi^3} \epsilon^2 \times$$

$$\times \int_0^{\alpha_c} d\alpha \sum_n \frac{\sec^2\alpha\theta\,[a_n - b\tan\alpha - \epsilon\sec\alpha]}{(a_n - b\tan\alpha)^2 \sqrt{(a_n - b\tan\alpha)^2 - \epsilon^2 \sec^2\alpha}} , \quad (11)$$

where the notations $a_n = n + 1/2$, $b = 2a/\phi_0$ are introduced. Equation (11) shows that at certain values of the magnetic flux, the radicand in the denominator tends to zero. Our interest is concentrated on the asymptotics of $\nu(\epsilon)$ at low energies ϵ.

Prier to estimation of ν^{res}, we shall consider the contributions of different angles α to the resonance amplitude. It is reasonable to assume that because of the factor $\sec^2\alpha$ in the numerator of Eq. (11), the angles $\alpha \sim \alpha_c$ are the main contributors to the integral. It is convenient to introduce a new variable of integration $x = \tan\alpha$. Then the neighborhood of the upper limit $x_0 = \tan\alpha_c$ is the main contributor to the integral. Introducing the notation $\tilde{a} = a_n - bx_0$ and the small deviation $\xi = x_0 - x \ll 1$, we can write down the equation for the resonance condition as:

$$(b^2 - \epsilon^2)\xi^2 + 2(\tilde{a}b + \epsilon^2 x_0)\xi + \tilde{a}^2 - \epsilon^2(1 + x_0^2) = 0 . \quad (12)$$

The solution of Eq. (12) to the accuracy within the first order terms of $|\epsilon|$ gives:

$$\xi_{1,2} \simeq -\frac{\widetilde{a}}{b(1+x_0^2)} \pm \frac{|\epsilon|}{b\sqrt{1+x_0^2}}. \qquad (13)$$

The expression in front of the radical in the denominator is of second order smallness in $|\epsilon|$, i.e. $|\widetilde{a}|^2 \gtrsim |\epsilon|^2 (1+x_0^2)$, which leads to its cancellation with the similar term in the numerator.

The remaining integral is estimated to be a constant of about unity. Resonance-induced spikes of the density of states appear when the Andreev level coincides with the Fermi energy at a certain flux in the N layer. In the vicinity of the chemical potential there is a strong degeneracy of the quasiparticle states with respect to the quantum number q. As a result a macroscopic number of q states contributes significantly to the amplitude of the effect. Near the resonance, the ratio of the resonance and nonresonance amplitudes of the density of states is

$$\frac{\nu^{\text{res}}}{\nu^{(0)}} \sim \frac{1}{|\epsilon|^2} >> 1\,. \qquad (14)$$

Thus, we have shown that a change in the magnetic flux leads to resonance spikes in the density of states of the NS contact. The flux interval between the spikes is equal to the superconducting flux quantum ϕ_0.

3. Discussion

The flux quantization effect and the paramagnetic contribution to the susceptibility of a thin-wall pure metal cylinder in the vector potential field were predicted by Kulik [18]. In pure normal metals there is a length $\xi_N = \hbar v_F / k_B T$, which has the meaning of a coherence length of a system with disturbed long-range order. When the temperature lowers, this length becomes equal to the characteristic dimensions of the system, which can lead to interference effects in the system. Kulik [18] shows that the magnetic moment of a thin-wall cylinder is an oscillating function of the magnetic flux through the cross section of the cylinder, its oscillation period being equal to the flux quantum of the normal metal hc/e. The effect is generated by quantization of the electron motion along the perimeter of the cylinder and is due to the sensitivity of the states of the system to the vector potential field (Aharonov–Bohm effect [8]). Bogachek and Gogadze [16] investigated the coherent quantum effect in singly connected normal cylindrical conductors in a weak magnetic field. The authors proved the existence of an oscillating component with a flux period hc/e in the magnetic moment The oscillation amplitude is small due to the smallness of the quasiclassical parameter of the problem $1/k_F R$. The amplitude of the effect decreases exponentially as the radius R increases. As a result, the persistent current disappears in macroscopic systems. The effect of flux quantization in pure Bi whiskers was first detected experimentally by Brandt et al. ([19,20]). That was the first observation of the interference effect of the flux quantization in nonsupercondncting condensed matter.

Mota et al. [2,3] investigated a mesoscopic NS structure in a magnetic field. If the electric contact between the N and S elements is good, the electrons penetrate easily from the superconductor to the normal layer and thus significantly affect the properties of the NS system. There are two types of electron collisions in a normal film — a specular reflection from one boundary and the Andreev reflection from the another. Along with the quasiparticle trajectories closed around the circular perimeter of the cylinder, new trajectories appear in a weak field, which screen the normal metal. The new trajectories of the «particles» and «holes» confine the area of a triangle whose base in a part of the NS boundary between the points at which the quasiparticle collides with this boundary. This area is maximum for the trajectories touching the superconductor. At certain values of the magnetic flux through the triangle area, the electron density of states experiences resonance spikes [9,10]. Their existence was proved in the standard calculation of the density of states $N(\varepsilon) = \sum \delta[\varepsilon - \varepsilon_n(q,\alpha,\phi)]$. Here we demonstrate this using the Greens functions of Gorkov.

Thus, the quantum proximity effect transforms the periodic flux-induced oscillations of the thermodynamic value with a period hc/e into periodic resonance spikes with a period equal to a superconducting flux quantum $hc/2e$. The response to a weak magnetic field ($H \sim 10$ Oe) is paramagnetic and the susceptibility amplitude becomes anomalously large. The resonance features disappear when the magnetic flux increases and its value divided by $hc/2e$ starts the exceed the highest Andreev «subband» numbei (see Eq. (4)). Only the quasiparticle trajectories that do not collide with the superconducting boundary contribute to the susceptibility, but their amplitudes are rather small (see above). Under this condition, the experiment only registers a large diamagnetic response (Meissner effect) We can therefore conclude that, the resonance contribution to the paramagnetic susceptibility can only appear in comparatively weak magnetic fields. In this case the experiment shows the reentrant effect [2,3].

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