Orbital Paramagnetism of Electrons in Proximity to a Superconductor

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We consider the magnetic response of a normal layer (N) coating a superconducting cylinder (S). The diamagnetic response of the normal layer (proximity effect) is related to the formation of Andreev levels. At low energies, the density of such states goes linearly to zero which enhances the various scattering mean-free paths. As a consequence, the low-energy glancing states, which can skip along the outer boundary, can have relatively large magnetic moments that lead to a low-temperature paramagnetic correction to the Meissner result.

One of the most intriguing recent experimental results [1] on the superconducting proximity effect has been the observation of a paramagnetic correction to the magnetic response at low temperatures of a normal metal (N) coating a superconducting cylinder (S). In these experiments S was Nb or Ta and N was relatively clean Cu or Ag. The effect was obtained below a well-defined temperature, $T_{\min}$, which depended on the sample geometry and other material properties. $T_{\min}$ ranged from below 10 mK to above 100 mK and at $T_{\min}$ the diamagnetic response showed a minimum. The temperature dependence of this novel effect could be fitted by $\exp(-T/T^*)$, where $T^* \sim \hbar v_F/L_y$ is inversely proportional to the circumference of the cylinder, L_y (whose radius is R). This dependence prompted a tentative description of this phenomenon as a mesoscopic one, although it has to be borne in mind that the values of L_y ranged up to a fraction of a mm. Our purpose here is not to explain the experimental results of Ref. [1], but to provide a physical picture according to which a significant paramagnetic effect may be possible. Previous theoretical calculations [2–5] using the Eilenberger and Usadel formulations of the theory of the proximity effect predicted a saturation of the magnetic response of an NS layer at low temperatures (including the range around $T_{\min}$).

We start by considering the problem of a clean normal layer parallel to the y-z plane and of width d, separating a superconductor ($x \geq d$) and the vacuum ($x \leq 0$). (We shall return later to the effects of the cylindrical geometry). The spectrum of this system can be determined by solving the Andreev equations [6,7] which are a linearized version of the Bogolubov-de Gennes equations. The interface with the vacuum is regarded as a perfect sharp reflector. The interface with the superconductor is described by proper boundary conditions for the various reflections.

One finds for an ideally transparent interface that the linearized spectrum consists of equidistant levels characterized by the x-component of the Fermi velocity v_{Fx}

$$\epsilon_n = \hbar\pi \frac{v_{Fx}}{2d}(1/2 + n) \; . \qquad (1)$$

The density of states (DOS) at low energies, $\epsilon \ll \hbar v_F/d \ll \Delta$, is of relevance to us (the second inequality means that the normal-metal thickness is larger than the coherence length in the superconductor). Since the kinetic energy of the lateral motion (in the y and z directions) does not appear, it follows from Eq. (1) that the only way to get a low excitation energy is to have a small v_{Fx}, i.e., a glancing-incidence state [8]. Thus, the low-energy DOS can be obtained from the density of the values of v_{Fy} and v_{Fz} that yields the required v_{Fx}. A simple calculation yields for the DOS per volume due to the $n = 0$ state:

$$n_0(\epsilon) = \frac{d}{2\pi}[\frac{4m}{\pi\hbar^2}]^2\epsilon \sim N(0)\frac{\epsilon}{\hbar v_F/d} \; . \qquad (2)$$

The linear decrease of the DOS with respect to the normal density of states at the Fermi energy, $N(0)$, due to the proximity effect was obtained already in Ref. [9]. It has serious implications. For example, it implies that the impurity scattering rate of these states in the Born approximation is also energy dependent and greatly reduced at low energies:

$$\frac{1}{\ell(\epsilon)} = \frac{1}{\ell_{\rm el}}\frac{\epsilon d}{\hbar v_F} \; , \qquad (3)$$

where $\ell_{\rm el}$ is the elastic mean-free path in the normal metal. This protection of the low-energy states from impurity scattering will play an important role in our considerations. In fact, for energies below $T^* \sim \hbar v_F/L_y$ and for $\ell_{\rm el} \gtrsim d$, $\ell(\epsilon)$ will exceed L_y. Thus, such low-energy states will behave *ballistically.*

We now introduce a small (since we consider here the linear response) magnetic field, B in the positive z-direction. It is described by a vector potential $\boldsymbol{A} = (0, B(x - x_0), 0)$, where x_0 is a constant. The effect of B is manifested in the B-dependent phases in the Andreev equations. The total relevant, gauge-invariant, phase shift due to B is determined by the flux, $\Phi = Bd^2 v_y/v_x$

through the triangle bounded by the two Andreev reflections mentioned above, and the ordinary reflection at the wall with the vacuum (we neglect any penetration of the field into the superconductor). Besides the usual diamagnetic, $O(B^2)$, upward shift of each energy level, the degenerate pair of levels with $\pm v_y$ splits as follows:

$$\epsilon_{n=0} = \hbar\pi \frac{v_{Fx}}{4d}[1 \pm \frac{4\Phi}{\Phi_0}] \; . \qquad (4)$$

Here Φ_0 is the flux quantum h/e and we displayed the result for the lowest, $n = 0$, levels. The paramagnetic linear splitting $\pm e|v_y|dB/2$ is just the energy of the magnetic moment of the current loop of the state under discussion, in the magnetic field B.

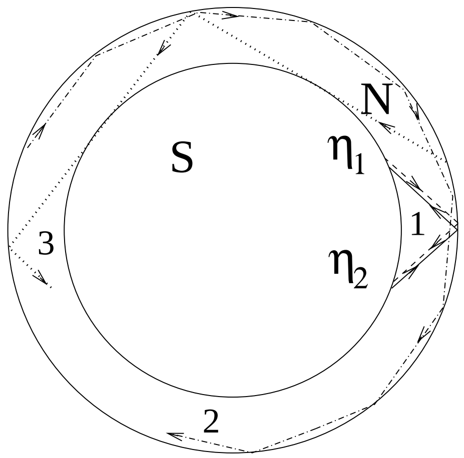

FIG. 1. Cylindrical geometry showing electron and hole orbits. (1) Andreev orbit. (2) “glancing” orbit which does not touch the superconductor. (3) limiting case.

A crucial observation at this stage is the following: For low-energy states with v_{Fx}/v_{Fy} small enough, the quasiparticle will have a classical orbit of a topologically different nature: The orbit will encircle the whole cylinder without hitting the superconducting surface even once, see Fig. 1. This gives a current loop of area πR^2, where $R = L_y/2\pi$, and a magnetic moment much larger than that of the Andreev electrons. These states, to which we shall refer as “glancing states” may give a large contribution to the magnetic response of the system. We shall see later that these states occur at energies below $\epsilon_g \sim \frac{\hbar v_F}{\sqrt{dL_y}}$, where the linear behavior of the density of states, Eq. (2) saturates at a value of the order of $N_s \sim N(0)\sqrt{d/R}$. ϵ_g is the crossover energy between the two types of states: Andreev (bulk) and glancing (surface) ones.

Even at very low temperatures, when inelastic scattering is weak enough, these states will give their full contribution only when their effective elastic mean-free path $\ell(\epsilon)$ is larger than or comparable to the circumference L_y. Otherwise, their contribution is reduced. We shall from now on consider *only* the states in the energy range below T^*, where the glancing states are effectively separated from the Andreev ones.

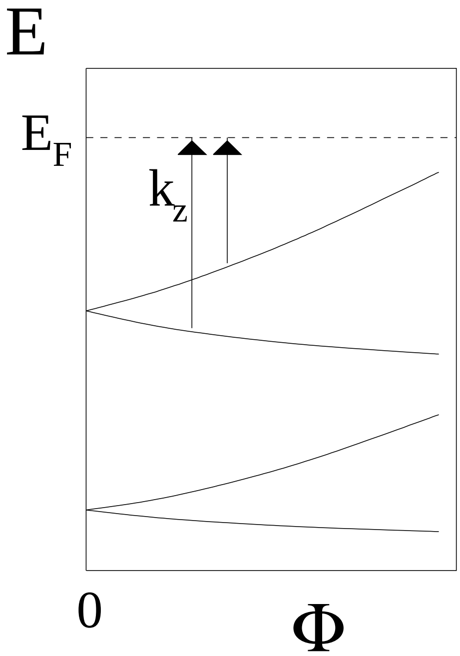

FIG. 2. Quantized levels in y-direction are split by the magnetic flux Φ. This is shown schematically for small values of Φ/Φ_0. By choosing appropriate values of k_z, a split pair is shifted to the Fermi energy. Note the curvatures of the upper and lower members of a split pair (those lead to the diamagnetic part of the response).

So far, we have neglected the possibility of scattering from a glancing state to other glancing states. This was not considered in calculating the effective lifetime, Eq. (3). It can be easily seen that the matrix elements for such a scattering are of the same order of magnitude as for scattering of regular states. The reduced density of glancing states mentioned above (N_s) will lead to an enhancement of the effective mean-free path, ℓ_{eff} for this scattering of glancing into glancing states by a factor of order $\sqrt{L_y/d}$. One may, for simplicity, discuss the case where the nominal mean-free path $\ell_{\rm el} \gtrsim \sqrt{dR}$, corresponding to the cleaner samples of Ref. [1]. Here the total effective mean-free path of the low-energy states under discussion is comparable to or larger than L_y. For the less clean samples, with $\sqrt{dR} \gtrsim \ell_{\rm el} \gtrsim d$, the glancing states are still effectively separated from the bulk, Andreev ones. However their elastic mean-free path for scattering among themselves is smaller than L_y. This will reduce the magnetic response associated with these states significantly but not catastrophically. In the well-known case of normal persistent currents, this reduction factor [10] is of the order of ℓ_{eff}/L. Here, ℓ_{eff} should be the effective mean-free path for scattering from glancing to glancing states estimated above.

It is straightforward to evaluate the paramagnetic response of a pair of levels whose B-dependences are $\epsilon \pm \mu B$. One finds for the magnetization, denoting $\beta = 1/k_BT$, and including both “electron”- and “hole”-type excitations, $M = \mu^2\beta\cosh^{-2}(\beta\epsilon_n/2)B$. The Pauli-type susceptibility is obtained from the above, by noting that for a system with a DOS $N(0)$ at the Fermi level, the number of states with $\beta\epsilon_n \lesssim 1$ contributing to M is $N(0)k_BT$, this susceptibility is independent of T in the limit $T \to 0$. This type of paramagnetism is due to a band of states of width k_BT around E_F. In the usual normal-metal persistent current case in a long cylinder, the continuum DOS at the Fermi level is due to the k_z states, for each k_x, k_y state. In fact, for each pair of k_x, k_y levels below E_F having a magnetic-field dependence $\epsilon \pm \mu B$ (which lead to hole-type excitations), there is a pair of filled k_z continua, starting from each of these levels and filled up to E_F, see Fig. 2. Each of those has a 1D-type DOS. Their population difference at $T \to 0$ leads to the paramagnetic part of the response, reviewed below.

An important observation is that within the Andreev approximation the energies of the Andreev states do not have a direct dependence on k_y and k_z, see Eq. (1). Thus, they do *not* have the continuum going up to the Fermi energy and hence do *not* contribute to the paramagnetic response at $T = 0$. This is a physical explanation for the diamagnetic screening in the proximity layer.

We shall from now on concentrate on the glancing states, since the magnetic moments they create are relatively large and survive the proximity effect. The Andreev states do not contain k_z, therefore, as discussed above, *they do not contribute to the paramagnetic response.*

Let us now consider the contribution of the glancing states. These states do not experience an Andreev reflection. In quantum-mechanical language, the glancing states are bound to the outer wall by the centrifugal potential $\sim\ m^*(Rv_y)^2/2r^2$. (Here, m^*Rv_y is the angular momentum). Going away from the outer wall, these states will decay exponentially for large enough v_y and have a negligibly small probability to touch the superconductor. It is straightforward to see that the full quantum-mechanical treatment of these states (see for example Ref. [11]) reproduces the semiclassical estimates of ϵ_g and N_s.

The role of the Andreev reflections is, as discussed above, to modify the magnetic response of the Andreev electrons, and to reduce the DOS at these low energies and thus decrease the impurity scattering of the glancing states. The energies and flux-dependences of the latter are as in the purely normal case. A very instructive way to calculate the magnetic response of these states is to go back to older calculations [12,10] of normal persistent currents in a long cylinder, and use the appropriate portion of those calculations. Defining: $\xi_j^2 = (E_F - \epsilon_j)/(h^2/2m^*L_y^2)$ where $\epsilon_j = (\hbar\pi j/d)^2/(2m^*)$ and $\phi = BL_yd/\Phi_0$, one finds for the persistent current linear in ϕ, in the clean case:

$$I = (2L_z/L_y)\tfrac{2c}{\Phi_0}(h^2/2m^*L_y^2)\phi \sum_{j=1}^{k_F d/\pi} \sum_{m\le\xi_j} \left[\sqrt{\xi_j^2 - m^2} - \frac{m^2}{\sqrt{\xi_j^2 - m^2}}\right] . \qquad (5)$$

The first term is a diamagnetic contribution, the second one is of paramagnetic nature (the eigenenergies go like $\pm\mu B$ and their populations differ by $O(B)$). These two contributions almost cancel and their lack of cancellation due to the finite L_y produces the small normal-metal persistent current. The way the cancellation occurs is rather nontrivial: It is seen from Eq. (5) that the diamagnetic term is dominant at low m while the paramagnetic one dominates at higher values of m. In the purely normal case, the sum for each j cancels except for single-level contributions. In our proximity problem, only the glancing, large m, states behave as in the normal metal, they also survive better elastic scattering. Thus, their paramagnetic contribution to the normal-type persistent current is not cancelled by the diamagnetic terms for each j separately.

To make an estimate of the number, N_g, of v_x values which lead to glancing states, we note from Fig. 1 that the last glancing state has $k_x/k_y = \sqrt{\pi d/2L_y}$ (for $d \ll L_y$). Thus, taking for glancing states $k_y \approx k_F$, $N_g(d) = k_F d\sqrt{d/\pi L_y}$. Its order of magnitude is around 1000 for typical samples of Ref. [1], i.e., $N_g \gg 1$. We shall now estimate the paramagnetic contribution of these glancing states. To evaluate the second term in Eq. (5), we replace the sum over the glancing states by an integral. This is a good approximation since the summand varies slowly with j and m and all the terms are of the same sign. We approximate the centrifugal potential near the outer wall of the cylinder by a triangular well and use for its eigenvalues a quasiclassical approximation (which is known to give reasonable results for even the low-lying states [11]). This gives $(4/9\pi)\xi^3(d/R)^{3/2}$ for the double sum over the glancing states (without the prefactor), where $\xi = k_F R$. We assumed that $k_F^2 d^3/R \gg 1$. We now evaluate the full diamagnetic current, i.e., the first term in Eq. (5), I_D. Replacing the sum by an integral, we find

$$I_D = \frac{\pi L_z e^2}{c}\frac{dB}{4\pi^2}\sum_j \epsilon_j \sim k_F^3 d^2 L_z B \frac{e^2}{m^* c} . \qquad (6)$$

The ratio of the paramagnetic contribution of the glancing states, I_P to the total diamagnetic current is:

$$I_P/I_D \cong \frac{1}{3\pi}\sqrt{\frac{d}{L_y}} . \qquad (7)$$

Neglecting the diamagnetic screening, this means that I_P is of the order of percents of I_D.

In a realistic situation where strong screening of the external field takes place in the proximity layer (as in the experiment of Ref. [1]), the total Meissner current is given by $I_M \sim (c/4\pi)L_z B \sim (\lambda_L/d)^2 I_D \ll I_D$, where $\lambda_L^{-2} = 4\pi ne^2/m^*c^2$. On the other hand, I_P is decreased by screening only like λ_{eff}/d, where λ_{eff} is the actual screening length in the proximity layer. For simplicity we have used here the local picture: it turns out that already for a large mean free path of order $10d$, the magnetic field is exponentially screened. *i.e.* the clean limit in which the magnetic field follows a power law [2] is practically never realized [13]. The surprisingly large ratio of the paramagnetic current and the Meissner current screening the *external* field is thus $(1/3\pi)(\lambda_{eff}d/\lambda_L^2)\sqrt{d/R}$. In addition, the fields produced by the paramagnetic currents will themselves be diamagnetically screened over the length λ_{eff}. This should further reduce the measured paramagnetic signal by (λ_{eff}/d). Of course we did not provide a self-consistent evaluation of the magnetic response, our aim was to point out a significant correction to the Meissner effect of the proximity layer.

An upper limit (see for example Ref. [14]) for the magnetic field scale above which nonlinearity will start and the effect will be suppressed is given (in the unscreened case) by $BL_yd \sim \Phi_0$, i.e., $B \sim 0.1$G. This is in agreement with experimental observations, see Ref. [1].

So far we considered the limit $T = 0$ and a dephasing length $L_\phi \gtrsim L_y$. A first source of temperature dependence is contained in the effective mean free path defined in Eq. (3). Denoting by T_0 the low-energy scale for which the effective elastic mean free path becomes of order L ($k_BT_0 \sim \hbar v_F/\sqrt{Ld}$ for the case considered above) one finds, for $T >> T_0$, a T_0/T reduction of the paramagnetic response.

At high enough temperatures, coherence is lost and I_P, being a normal mesoscopic persistent current, should decrease exponentially, like $\exp(-L/L_\phi)$. We also note that some of the characteristic energies involved in the process are of the order of $\hbar v_F/L_y \sim T^*$. However, further calculations are needed for the temperature dependence.

The effects of disorder deserve a more complete treatment as well. In the usual persistent current case, disorder decreases the amplitude of the orbital effects by a factor of $\ell_{\rm el}/L_y$, as mentioned above.

Finally, we emphasize that in our rough initial treatment we did not deal with the self-consistency of the pair potential at the NS boundary. Likewise, the magnetic field effects were also discussed without full self-consistency. An experimental test for the ideas presented in this note would be to cut the normal metal cylinder along the z-axis and check whether the effect would disappear. Another check would be to examine whether a short cylinder or a ring would not exhibit the effect.

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