Tunneling Anomaly in Superconductor above Paramagnetic Limit

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We study the tunneling density of states (DoS) in the superconducting systems driven by Zeeman splitting $E_Z$ into the paramagnetic phase. We show that, even though the BCS gap disappears, superconducting fluctuations cause a strong DoS singularity in the vicinity of energies $-E^*$ for electrons polarized along the magnetic field and $E^*$ for the opposite polarization. The position of the singularity $E^* = \frac{1}{2} \left( E_Z + \sqrt{E_Z^2 - \Delta^2} \right)$ (where $\Delta$ is BCS gap at $E_Z = 0$) is universal. We found analytically the shape of the DoS for different dimensionality of the system. For ultrasmall grains the singularity has the form of the hard gap, while in higher dimensions it appears as a significant though finite dip. Our results are consistent with recent experiments in superconducting films.

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It is known that magnetic field, $H$, suppresses superconductivity because it violates the time reversal symmetry \cite{1}. Typically, this symmetry breaking is associated with effect of the magnetic field on the orbital motion of electrons. However, in some physical situations, the main mechanism of the destruction of the Cooper pairing is the Zeeman splitting of states with the same spatial wave-functions but opposite spin directions. One can consider, e.g., a thin superconducting film placed in magnetic field parallel to the plane of the film \cite{2}. Recently, another possibility was exploited experimentally \cite{3} – ultrasmall superconducting grains. In both cases, the size of a Cooper pair is restricted geometrically and a flux through this pair reaches flux quantum at fields higher that the bulk critical field $H_c2$. We assume that superconductivity is already destroyed by the Zeeman splitting $E_Z = g_l \mu_B H$ when it happens \cite{4}, (here $g_L$ is the $g$-factor Lande, and $\mu_B$ is the Bohr magneton).

Strictly speaking, the spin splitting destroys superconductivity as soon as $E_Z \geq \sqrt{2} \Delta$, with $\Delta$ being the superconducting gap. This transition from superconducting to paramagnetic normal state is of the first order, and these phases coexist in the interval $\Delta \leq E_Z \leq 2 \Delta$. From now on, we assume the condition $E_Z > \sqrt{2} \Delta$.

One might expect that the Cooper pairing is irrelevant for the properties of the normal paramagnetic phase. In this Letter we show that, on the contrary, there are clear and observable effects of the pairing in paramagnetic state \textit{even far from the transition region}.

One of the most fundamental manifestation of the superconductivity is the gap in the tunneling density of states (DoS) around the zero energy \cite{2}. This gap apparently disappears when the system becomes paramagnetic. We will show that at the same time there appears a dip in the DoS. The shape of this dip depends on the dimensionality of the system. However, its position $E^*$ is remarkably universal:

$$E^* = \frac{1}{2} \left( E_Z + \sqrt{E_Z^2 - \Delta^2} \right)$$

for OD (grain), 1D (strip) and 2D (film) cases.

This result should be compared with the singularity in the DoS due to the usual superconducting fluctuations in the normal metal. According to Ref. \cite{5}, in the magnetic field another anomaly appears in addition to the zero bias anomaly when bias $V$ corresponding to Zeeman splitting $eV_s = E_Z$. The singularity we consider here is positioned at substantially lower energy and, as we will see, is stronger than those considered in Ref. \cite{5}.

Recently, singularity of this type was observed in granular Al film in a parallel magnetic field \cite{6}. We believe that the deviation from the law $eV_s = E_Z$ observed in Ref. \cite{6} is coherently explained by our Eq. (1).

We begin with the simplest but instructive case of 0D system (ultrasmall grain). The Hamiltonian $\hat{H}$ of this system can be written in the basis of the exact single-electron states for non-interacting system as

$$\hat{H} = \sum_{i; \sigma = \uparrow, \downarrow} E_{i,\sigma} a_{i,\sigma}^\dagger a_{i,\sigma} - \lambda \delta \sum_{i,j} a_{i,\uparrow}^\dagger a_{j,\downarrow}^\dagger a_{j,\downarrow} a_{i,\uparrow}.$$ (2)

Indices $i,j$ and $\sigma = \uparrow, \downarrow$ label the orbital and spin state of an electron: its total energy $E_{i,\sigma}$ is the sum of orbital $e_i$ and spin parts, $E_{i,\uparrow(\downarrow)} = e_i \mp E_Z/2$; and $a_{i,\sigma}^\dagger a_{i,\sigma}$ are the corresponding fermionic creation-annihilation operators. Finally, $\delta = \langle \epsilon_{i+1} - \epsilon_i \rangle$ is the mean level spacing and $\lambda$ is the dimensionless interaction constant. In Eq. (2), we omitted some diagonal terms (those with orbital indices equal pairwice), which with the help of Hartree-Fock approximation can be included into the definition of the eigenergies $\epsilon_i$. We also omitted off-diagonal terms (involving the fermionic operators with non-paired indices): the corresponding matrix elements are known \cite{7} to be smaller than the diagonal ones by a large factor $1/g$, where $g \gg 1$ is the dimensionless conductance of the grain. Hamiltonian \cite{6} is nothing but the usual BCS Hamiltonian, and it was used in numerous publications \cite{8} on the properties of ultrasmall superconducting grains.
BCS instability which system \( \frac{\Omega}{2} \) has at temperature \( T = 0 \) and \( H = 0 \), disappears as soon as \( E_Z \) exceeds \( \sqrt{2}\Delta \). In the absence of the superconducting gap, the structure of the ground state is similar to that without interaction: orbitals with energies \( |\epsilon_i| > E_Z/2 \) are spinless (orbitals with \( \epsilon_i < -E_Z/2 \) are double occupied while those with \( \epsilon_i > E_Z/2 \) are empty), while orbitals in the energy strip \( |\epsilon| < E_Z/2 \) are spin polarized with spin up (\( \uparrow \)) (we measure all the energies from the Fermi level). The interaction term in the Hamiltonian \( \frac{\Omega}{2} \) does not affect the spin polarized states, but mixes the double-occupied and empty states. Those states are separated from each other by large gap \( E_Z \). Therefore, this mixing is perturbative, and it does not change ground state qualitatively.

Contrarily, the spectrum of the excitations, e.g. the tunneling DoS changes drastically due to the interaction. Consider a spin-down electron with energy \( 0 < E < E_Z/2 \) entering the grain. The orbital energy of this electron \( \epsilon_0 \) is close to \( \epsilon_0 = E - E_Z/2 \), and this orbital is occupied by a spin-up electron. Therefore, the tunneling event creates the spin-singlet state with the energy \( 2\epsilon_0 \) and this state can mix with all the empty states \( \epsilon_i \). This mixing turns out to be resonant at some energy \( E = E^* \); for \( E \) close to \( E^* \) it requires nonperturbative treatment.

Before turning to the rigorous calculations, let us discuss this effect qualitatively using the following simplification. Instead of the whole many-body system, we consider two-electrons only, however, the single-electron orbitals for this pair are restricted by the orbitals with \( \epsilon_i > E_Z \) and by one orbital \( \epsilon_0 \). The role of the rest of the electrons is to restrict the Hilbert space for a given electron pair. This simplification is similar to the Cooper pair. The interaction in Eq. \( \frac{\Omega}{2} \) involves spin singlet orbitals only. Thus, the wave function of the electron pair \( \psi \) can be labeled by one orbital index and it obeys the Schrödinger equation \( \epsilon_j \psi_j = 2\epsilon_j |\psi_j| - \lambda \delta \sum_j |\psi_j| \). The eigenenergies \( \epsilon \) of this equation can be found from

\[
\frac{\delta}{2\epsilon_0 - \epsilon} + \sum_{\epsilon_i > E_Z/2} \frac{\delta}{2\epsilon_i - \epsilon} = \frac{1}{\lambda}. \tag{3}
\]

For low-lying eignestates \( \epsilon < E_Z \) one can substitute the summation in Eq. \( \frac{\Omega}{2} \) by integration. Given the high-energy cut-off \( \omega \), it yields

\[
\frac{2\delta}{2\epsilon_0 - \epsilon} + \ln \left( \frac{\Delta_b}{E_Z - \epsilon} \right) = 0, \quad \Delta_b = \omega e^{-2/\lambda}. \tag{4}
\]

For small level spacing \( \delta \ll E_Z, \Delta_b \), the pair energy \( \epsilon \) is

\[
\epsilon = \epsilon_0 + \frac{\Omega^b}{2} \sqrt{\left( \frac{\Omega^b}{2} - \epsilon_0 \right)^2 + 2\delta \Delta_b}, \quad \Omega^b = E_Z - \Delta_b. \tag{5}
\]

Prior the spin down electron tunnels in, the energy of the spin-up electron on the orbital \( \epsilon_0 \) was equal to \( E = \epsilon_0 - E_Z/2 \), and thus the energy of one electron excitation \( E = \epsilon - \epsilon_0 \) is given by

\[
E = \epsilon^*_b \pm \left[ \left( \frac{\Omega^b}{2} - \epsilon_0 \right)^2 + 2\delta \Delta_b \right]^{1/2}, \quad \epsilon^*_b = \frac{E_Z + \Omega^b}{2}. \tag{6}
\]

The origin of tunneling anomaly is now transparent from Eq. \( \frac{\Omega}{2} \): due to the repulsion between the state formed immediately after an electron tunnels into the system and the bound states of the Cooper pair, there is no spin-down one electron excitations in the energy strip \( |E - \epsilon^*_b| < (2\delta \Delta_b)^{1/2} \) – hard gap in the DoS is formed. It is important to emphasize that (i) the width of this gap \( \sqrt{8\delta \Delta_b} \) significantly exceeds the single electron level spacing and (ii) this singularity persists even when the system is deep in the paramagnetic phase \( E_Z \gg \sqrt{2}\Delta \).

It is also noteworthy that if a spin-up electron tunnels into the grain, it never finds the pair for itself, and, therefore, no tunneling anomaly happens in this case. It means that the overall DoS does not vanish but rather shows the suppression by a factor of two. However, for the spin-polarized electrons tunneling into the grain, we predict the complete suppression of the tunneling DoS.

The same arguments allow to justify the similar singularity, when spin-up electron with energy \( -E_Z/2 < E < 0 \) tunnels out from the system, while the spin down electrons tunneling from the system are not affected.

The qualitative consideration above grasps the correct physics, however it fails to describe the effect quantitatively, it predicts correctly neither the position \( \epsilon^*_b \) nor the width of the gap. This is similar to the discrepancy between the binding energy \( \Delta_b \) in the original Cooper procedure and the correct BCS gap \( \Delta \): all the electrons below the Fermi energy were frozen. To remedy this drawback, we employ a parametrically exact procedure described below.

We start with the propagator of the superconducting fluctuations. The diagrammatic equation, Fig. \( \frac{\Omega}{2} \), yields

\[
\Lambda^\sigma(\omega) = 2\delta \left[ \ln \left( \frac{E_Z^2 - \omega^2}{2\Delta^2} \right) \right]^{-1}. \tag{7}
\]

Here \( \Delta = \bar{\omega}e^{-1/\lambda} \) is the BCS gap, and \( \omega_+ = \omega + i\omega \). Propagator \( \frac{\Omega}{2} \) has the pole at \( \omega = \pm \Omega \):

\[
\Omega = \sqrt{E_Z^2 - \Delta^2}. \tag{8}
\]

\( \Omega \) has the meaning of the bound state energy of two quasi-particles. Using Eq. \( \frac{\Omega}{2} \) instead of oversimplified Eq. \( \frac{\Omega}{2} \) in the expression for \( \epsilon^*_b \) in Eq. \( \frac{\Omega}{2} \), we obtain Eq. \( \frac{\Omega}{2} \) for the position of the singularity \( \epsilon^*_b \).

Consider now the shape of the tunneling DoS \( \nu(\omega) \) near the singularity. DoS for spin polarizations \( \sigma = \uparrow, \downarrow \) can be expressed through the one particle Green function (GF) \( iG_{1,\sigma}(\omega) \) at zero temperature: \( \nu_{\sigma}(\omega) = \frac{1}{\pi} Im \ln \left( 1 - \frac{\omega}{\omega_+} \right) \) at \( \omega_+ = \omega + i\omega \). See Fig. \( \frac{\Omega}{2} \) for further details.
\[-\frac{1}{2} \Im \sum \frac{G_{\delta, \sigma}}{1 - G_{\delta, \sigma}^0 - \Sigma_{\delta, \sigma}}.\] In its turn, the GF is given by \(1/G_{i, \sigma} = 1/G_{i, \sigma}^0 - \Sigma_{i, \sigma},\) where \(G^0\) is the GF for noninteracting system, \(G^0_{i, \tau, \delta} = (\omega_\delta - \epsilon_i \pm iE_\delta/2)^{-1}\), and \(\Sigma_{i, \sigma}\) is the one-particle self-energy \([11]\).

The leading part of \(\Sigma_{i, \delta}\), diagram Fig. 1b, is given by

\[-i\Sigma_{i, \delta} \propto \int \frac{d\omega_1}{2\pi} \Lambda_\delta(\omega + \omega_1) G^0_{i, \tau, \delta}(\omega_1).\] (9)

The singular contribution to the integral in Eq. (9) comes from the positive pole of \(\Lambda_\delta\), which corresponds exactly to the repulsion between the bound state of the Cooper pair with the state formed after tunneling which we discussed earlier. From Eq. (9) we obtain for the GF

\[G_{i, \delta} = \frac{\omega + \epsilon_i - E_\delta/2 - \Omega}{(\omega + \epsilon_i - E_\delta/2 - \Omega) - \delta/\Delta} G^0_{i, \tau, \delta}(\omega_\delta).\] (10)

Summing up over all the orbitals \(\epsilon_i\), and neglecting the fine structure of the DoS on the scale of \(\delta\), we find

\[\nu_{i, \delta} = \nu_0 F_0(\frac{\omega \pm E^\ast}{W_0}), \quad F_0(x) = \frac{\theta(x^2 - 1)|x|}{\sqrt{x^2 - 1}},\] (11)

where \(\nu_0\) is the bare density of states per one spin. Equation (11) describes the hard gap of the width

\[W_0 = \left(\frac{\delta \Delta^2}{\Omega}\right)^{1/2},\] (12)

positioned at \(\omega = E^\ast\) given by Eq. (11).

![Diagram](image)

**FIG. 1.** Diagrams describing (a) the fluctuation propagator \(\Lambda_\delta(\omega)\) and (b) first order contribution to the self energy \(\Sigma_{\delta, \tau}(\omega)\). Diagrams (c) and (d) show second order contributions to \(\Sigma\), which were neglected in comparison with reducible diagram (e).

Higher order corrections to the self-energy (e.g. shown in Fig. 1c,d) are negligible. Indeed, let us compare the contributions of those diagrams with the reducible diagram Fig. 1b, which is included in Eqs. (9) and (10). At \(\omega = E^\ast, 2\epsilon_i = \Omega\), all three GF in diagram (e) diverge after integration over intermediate frequencies, whereas only two GF diverge (non-resonant GF are indicated by •) in Fig. 1c,d. It means that diagrams (c-d) are smaller than the main contribution by a factor \(\delta/\Delta \ll 1\).

According to Eq. (11), the width \(W_0\) of the hard gap is much larger than the mean level spacing \(\delta\). However, it apparently vanishes as \(\delta/\Delta \ll 1\) for infinite systems. To be more precise, our 0D consideration was valid only if the width of the gap \(W_0 \sim \sqrt{\delta \Delta}\) does not exceed the Thouless energy \(E_T = hD/L^2\) (\(D\) is the diffusion coefficient). Since \(\delta \sim 1/L^d, (d\) is the dimensionality of the sample) the condition \(W_0 \lesssim E_T\) breaks down for bulk samples, \(L \to \infty\), in all physical dimensions \(d < 4\) and our zero-dimensional description (2) becomes invalid. Nevertheless, the strong singularity in the vicinity of \(E^\ast\) persists in the tunneling DoS for \(d = 1, 2\) systems \([12]\).

We describe the interaction by usual Hamiltonian \(\hat{H}_{\text{int}} = -\nu_0^{-1} \lambda \int \frac{d^d \alpha}{(2\pi)^{d/2}} a_\uparrow a_\downarrow \). In the bulk system, the superconducting fluctuations can be inhomogeneous. Thus, the propagator similar to Eq. (3) depends on the wavevector \(Q\). Diagram for such propagator averaged over disorder is similar to Fig. 1a and standard calculation yields:

\[\Lambda_\delta(\omega, Q) = 2 \bigg[ \nu \ln \left(\frac{E_Z^2 - (|\omega| + iDQ^2)^2}{\Delta^2}\right) \bigg]^{-1}.\] (13)

\(\Lambda_\delta\) at small \(Q, (DQ^2 \ll \Omega)\) has the singularity at \(\omega\) close to the energy \(\Omega\) from Eq. (8). Therefore, one may expect the singularity in DoS at the same energy \(E^\ast\).

Evaluation of the first order correction \(\delta\sigma_{\tau}(\omega)\) to the DoS, similar to Fig. 1b, confirms this expectation. Integrating over intermediate frequency, averaging over disorder, and assuming \(DQ^2 \ll \Omega\), we find the singular part of the correction \([13]\)

\[\frac{\delta\sigma_{\tau}(\omega)}{\nu_0} = \frac{\Delta^2}{2\nu_0 \Delta} \text{Re} \int \frac{d^d Q}{(2\pi)^{d/2}} |C_{\tau}(\omega, Q)|^2,\] (14)

where \(d = 1, 2\) is the dimensionality of the sample, and the Cooperon \(C_{\tau}\) is given by \(C_{\tau} = [\omega \pm E^\ast + iDQ^2]\). For 1D case (strip), integration in Eq. (14) gives:

\[\frac{\delta\sigma_{\tau}(\omega)}{\nu_0} = \frac{\Delta^{1/2}}{2\nu_0 \Omega D^{1/2}} \left(\frac{\Delta}{2|\omega| E^\ast}\right)^{3/2}.\] (15)

In two dimensions the correction to DoS vanishes in the lowest order. Second order correction similar to that in Fig. 1c are averaged over disorder \([13]\)

\[\frac{\delta\sigma_{\tau}(\omega)}{\nu_0} = -2 \bigg(\frac{\Delta^2}{4\nu_0 \Omega}\bigg) \frac{\partial}{\partial \omega} \text{Re} \int \frac{d^d Q}{(2\pi)^{d/2}} C_\sigma(\omega, Q) C^2_\sigma(\omega, Q_2).\]

For two dimensional film integration yields

\[\frac{\delta\sigma_{\tau}(\omega)}{\nu_0} = - \left[\frac{\Delta^2}{4g\Omega (\omega \pm E^\ast)}\right] \ln \left(\frac{\Omega}{\omega \pm E^\ast}\right)^2,\] (16)
where \( g = 4\pi \hbar D v_0 \gg 1 \) is the dimensionless sheet conductance of the film in the normal state. It is worth noticing, that the singularity given by Eq. (16) is much more pronounced than that due to the superconducting fluctuations in the normal metal \([7,8]\), \( g^{-1} \ln[\ln(\omega \pm E_Z)] \).

Both corrections (15) and (16) diverge with approaching the singularity point \( \omega \to \pm E^* \). It means that the higher order terms have to be summed up. Here we present only the results of this nonperturbative treatment [13]. The resulting DoS can be written as

\[
\frac{n_{1,2}^r}{n_0} = F_d \left( \frac{\omega \pm E^*}{W_d} \right), \quad d = 1, 2, \quad (17)
\]

where the characteristic widths of the singularity \( W_{1,2} \) in one and two dimensions are given by

\[
W_1 = \frac{\Delta}{3} \left( \frac{\Delta^{1/2}}{16\hbar_0 \Omega D^{1/2} h^{1/2}} \right)^{2/3}, \quad W_2 = \frac{\Delta^2}{4g\hbar}. \quad (18)
\]

In one dimension DoS acquires a universal shape:

\[
F_1(x) = 1 - \frac{2}{3} \text{Re} \left[ 1 - i x \left( \sqrt{1 + y(x)} + \sqrt{1 - y(x)} \right) \right], \quad (19a)
\]

where \( y(x) = \sqrt{1 + i x^2} \). In two dimensions shape of the DoS weakly depends on the conductance \( g \)

\[
F_2(x) = \text{Re} \left[ \frac{1 - z(x)}{1 + z(x)} \right], \quad z = \frac{1}{-i x + \ln(4g z)} \quad (19b)
\]

At \( x \gg 1 \), Eqs. (19) reproduce perturbative results (15) and (16). DoS near the energy \( E^* \) is shown in Fig. 2.

Finally, let us mention that the DoS singularity is suppressed by a finite temperature and by the spin-orbit scattering: the singularity remains observable only if \( W > T, h/\tau_{so} \). Corresponding expressions will be presented elsewhere [13].

In conclusion, we have shown that the tunneling DoS of superconductors above the paramagnetic limit has a singularity at energy determined by Zeeman splitting and superconducting gap, Eq. (1). This phenomenon is not take into account by usual BCS mean field and can not be obtained within Gorkov-Nambu formalism. The shape of the singularity, obtained nonperturbatively, depends on the dimensionality of the system, Eqs. (17) and (19). We believe that our theory is adequate for existing experiments [2], and suggests new effect in ultrasmall grains.

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