Quantum fluctuations and dynamic clustering of fluctuating Cooper pairs

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Abstract – We derive the complete expression for the fluctuation conductivity in two-dimensional superconductors as a function of the temperature and the magnetic field in the whole fluctuation region above the upper critical field \(H_{c2}(T)\). Focusing on the vicinity of the quantum phase transition near zero temperature, we propose that as the magnetic field approaches the line near \(H_{c2}(0)\) from above, a peculiar dynamic state consisting of clusters of coherently rotating fluctuation Cooper pairs forms and estimate the characteristic size and lifetime of such clusters. We find the zero-temperature magnetic-field dependence of the transverse magnetoconductivity above \(H_{c2}(0)\) in layered superconductors.

Understanding the mechanisms of superconducting fluctuations achieved during the past decades [1] gave a unique tool providing the information about the microscopic parameters of superconductors. The fluctuations are customarily described in terms of the so-called quantum corrections to conductivity, i.e. Aslamazov-Larkin (AL) [2] and Maki-Thompson (MT) [3,4] corrections, and/or the fluctuation corrections to the density of states (DOS) of the normal excitations [5,6]. The classic results obtained first in the vicinity of the superconducting critical temperature \(T_c\) were generalized to the far-from-\(T_c\) [7–9] and relatively high-fields [10] regions. More recently quantum fluctuations (QF) became the focus of the research. Effects of QFs on magnetoconductivity and the magnetization of 2D SCs have been studied at low temperatures and fields, \(H\), close to \(H_{c2}(0)\) in ref. [11]. It was found [12] that in granular SCs at very low temperatures and close to \(H_{c2}(0)\) the AL contribution is proportional to \(T^2\), and that the magnetoresistance grows due to the suppression of the DOS. The study of different fluctuation contributions to the Nernst-Ettingshausen effect in 2D SCs in a wide region above the transition line \(H_{c2}(T)\) demonstrated the importance of renormalization of the diffusion coefficient (DCR) due to QF [13]. Yet, the region near \(T = 0\) and near \(H = H_{c2}(0)\) remains poorly understood and a universal picture combining QF at high magnetic fields and finite temperature quantum corrections is still lacking.

In this Letter we offer a general unifying description of fluctuation-induced conductivity of a disordered 2D SC in a perpendicular magnetic field that holds everywhere above the transition line \(H_{c2}(T)\). Considering the vicinity of \(T = 0\), we find that on the approach to \(H_{c2}(0)\) from above, a peculiar dynamic state forms comprised by the clusters of the coherently rotating fluctuation Cooper pairs and estimate the characteristic size \(\xi_{QF}(H)\) and lifetime \(\tau_{QF}(H)\) of such clusters. Using the derived values of \(\xi_{QF}\) and \(\tau_{QF}\) we cross-check our conclusions by reproducing the results of [11–13].

We start with the derivation of universal expressions for the fluctuation contribution to conductivity and the DOS. The electromagnetic response operator in the leading order of the fluctuation propagator is defined by ten diagrams [1] shown in fig. 1. The corresponding contributions to conductivity at arbitrary magnetic fields and temperatures \(T_c(H) < T \ll \tau^{-1}\) (\(\tau\) is the electron elastic scattering time) are

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see eqs. (1), (2) and (3) on the next page
Here \( t = T/T_{c0}, \ h = \frac{\sigma^2}{\lambda_{\phi} T_{c0}} = 0.69 \frac{H}{M_{c2}(0)}, \ \gamma_E = 1.781 \ldots, \ M = (tT_{c0})^{-1}, \ \gamma_\phi = \pi/(8T_{c0}T_0), \ \tau_\phi \) is the phase-breaking time, \( E_m \equiv E_m(t, h, i \chi) \) with \( E_m(t, h, z) = \ln t + \psi \left( \frac{1 + z}{2} \right) + \right \frac{2}{\pi^2} + \frac{2m+1}{\pi^2} \right) - \psi \left( \frac{1}{2} \right) \) and its derivatives \( \frac{d^{(m)}E_m(t, h, z)}{d \chi} \). The sum of eqs. (1)–(3) presents the general expression for the total fluctuation correction to conductivity \( \delta \sigma_{xx}^{\text{tot}}(T, H) \) that hold in the complete \( T-H \) phase diagram above the line \( H_{c2}(T) \). We analyzed these expressions both analytically (by finding the asymptotic expressions for \( \delta \sigma_{xx}^{\text{tot}} \)) and numerically (by developing a program which calculates the complete surface \( \delta \sigma_{xx}^{\text{tot}}(T, H) \) for given parameters \( \tau \) and \( \tau_0 \) [14]. We show that the singular growth of conductivity near \( T_{c0} \) transforms into a relatively weak (logarithmic) growth of fluctuation correction to resistivity as one moves along the \( H_{c2}(T) \) line towards the low-temperatures region (i.e. close to \( H_{c2}(0) \)). The total fluctuation correction to the conductivity \( \delta \sigma_{xx}^{\text{tot}} \) remains negative in a wide domain relatively far from the \( H_{c2}(T) \) line (see fig. 2 where the regions with the dominating fluctuation contributions to magneto-conductivity are shown).

The analysis of fluctuation corrections enables us to develop a qualitative picture of the quantum phase transition in the vicinity of \( H_{c2}(0) \). To gain the physical insight, we first exercise our qualitative approach for the description of the vicinity of \( T_{c0} \). Here the lifetime of fluctuation-induced Cooper pairs (FCP), \( \tau_{GL} \), is obtained using the uncertainty principle: \( \tau_{GL} \sim \hbar/\Delta E \), where \( \Delta E \) is the difference \( k_B(T - T_{c0}) \) ensuring that \( \tau_{GL} \) should become infinite below \( T_{c0} \). This yields the standard Ginzburg-Landau time \( \tau_{GL} \sim k_B(T - T_{c0}) \sim \hbar/(\kappa_B T_0 e) \), where \( \epsilon = (T - T_{c0})/T_{c0} \ll 1 \) is the reduced temperature. The coherence length \( \xi_{GL}(T) \) is estimated as the distance which two electrons move apart during the FL time: \( \xi_{GL}(\epsilon) = (D\tau_{GL})^{1/2} \sim \xi_{BCS}/\sqrt{\epsilon} \) (\( \xi_{BCS} \) is the BCS coherence length, \( D \) is the diffusion coefficient). The order parameter \( \Delta^{(2)}(r, t) \) varies on the larger scale \( \xi_{GL}(T) \gg \xi_{BCS} \). The ratio of the FCP concentration to the corresponding effective mass \( n_{e_F}/m_{e_F} \sim e^2_D/\epsilon \) (with the logarithmic accuracy) in 2D is constant [1].

The major fluctuation contributions to conductivity stem from the direct FCP charge transfer (AL contribution) \( \delta \sigma_{xx}^{\text{AL}} \sim (n_{e_F}/m_{e_F}) e^2 \tau_{GL} \sim e^2/\epsilon \), the specific
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Fig. 2: (Color online) Top: fluctuation correction to conductivity (FC) \( \delta \sigma = \delta \sigma^{(tot)}(t, h) \) as a function of the reduced temperature \( t = T/T_{c0} \) and magnetic field \( h = 0.69H/H_c(0) \) plotted as surface. The FC changes its sign along the thick red line \( (\delta \sigma = 0) \). The boundary of the superconducting region is shown by a dashed line. Here \( \delta \sigma \) is plotted for constant \( \tau T_{c0} = 10^{-2} \) and \( \tau \tau_{c0} = 10 \). Bottom: contours of constant fluctuation conductivity \( (\delta \sigma = \delta \sigma^{(t)}(t, h)) \) shown in units of \( e^2/\hbar \). The dominant FC contributions are indicated in bold italic labels. The dashed line separates the domain of quantum fluctuations (QF) (dark area of \( \delta \sigma > 0 \)) and thermal fluctuations (TF). The contour lines are obtained from eqs. (1)–(3) with \( T_{c0} \tau = 0.01 \) and \( T_{c0} \tau_{\phi} = 10 \).

The quantum process of the one-electron charge transfer related to the coherent scattering of electrons on elastic impurities forming FCPs (anomalous MT contribution) \( \delta \sigma^{MT(AN)}_{xx} \sim \frac{e^2}{\hbar} \ln(\gamma_p) \) \citep{[1]} (fig. 1a, b)), and from the suppression of the quasi-particle density of states near the Fermi level. Indeed, the involvement of quasi-particles in the fluctuation pairing results in the opening of the pseudo-gap in the one-electron spectrum. The lack of corresponding electrons at the Fermi level leads to a decrease of the one-electron Drude-like conductivity. Such an indirect fluctuation effect formally is described by the third group of the four diagrams in fig. 1c), which are usually called DOS diagrams. The corresponding contribution has a negative sign at variance to the positive contributions from the AL and MT corrections, but close to \( T_{c0} \) it turns out to be less singular as a function of temperature \citep{[1]}:

\[
\delta \sigma^{DOS}_{xx} \sim -2n_e R \frac{e^2}{\hbar m_e} \sim -e^2 \int c^2 \frac{d\omega}{\hbar + \Delta_{BCS}(e)} \sim -e^2 \ln \frac{1}{\epsilon}.
\]  

The DOS contribution formally appears due to the summation of \( \langle [\Delta^{(0)}(q, \omega)]^2 \rangle \) over all long-wavelength fluctuation modes \( q < \xi_{BCS}(\sqrt{\epsilon}) \) in the static approximation \( (\omega \to 0) \). Relatively far from \( T_{c0} \) in zero field, \( \delta \sigma^{DOS}_{xx} \) can change the sign of \( \delta \sigma^{(t)}_{xx} \), but this happens only in the case of strong pair breaking, when the phase-sensitive anomalous MT contribution is suppressed. Finally, the last four diagrams in fig. 1d) together with the regular part of the MT diagram describe the renormalization of the diffusion coefficient (DCR) in the presence of fluctuation pairing. Close to \( T_{c0} \) their contribution is not singular, but becomes of primary importance in the periphery of the phase diagram and in particular at very low temperatures.

At zero temperature and the field above \( H_c(0) \), the systematics of the fluctuation contributions to the conductivity considerably changes with respect to that close to \( T_{c0} \). The collisionless rotation of FCPs (they do not “feel” the presence of elastic impurities) results in the lack of their direct contribution to the longitudinal (along the applied electric field) electric transport (analogously to the suppression of one-electron conductivity \( \delta \sigma^{e}_xx \) in strong magnetic fields \( \omega_\tau \sim 1 \), see ref. \citep{[15]}) and the AL contribution to \( \delta \sigma^{(e)}_{xx} \) becomes zero. The anomalous MT and DOS contributions turn zero as well but due to different reasons: Namely, the former vanishes since the magnetic fields as large as \( H_c(0) \) completely destroy the phase coherence, whereas the latter disappears since the magnetic field suppresses the fluctuation gap in the one-electron spectrum. Therefore the effect of fluctuations on the conductivity at zero temperature is reduced to the renormalization of the one-electron diffusion coefficient. FCPs here occupy the lowest Landau level, but all the dynamic fluctuations in the interval of frequencies from 0 to \( \Delta_{BCS} \) should be taken into account. The corresponding fluctuation propagator at zero temperature close to \( H_c(0) \) has the form \( \Delta_{0}(\omega) = -N_0^{-1} \left( \hbar + \omega/\Delta_{BCS} \right)^{-1} \) and

\[
\delta \sigma^{DCR}_{xx} + \delta \sigma^{MT(e)}_{xx} \sim - \frac{e^2}{\Delta_{BCS}} \int \frac{d\omega}{\hbar + \Delta_{BCS}} \sim -e^2 \ln \frac{1}{\hbar},
\]  

where \( \hbar = [H - H_{c2}(0)]/H_{c2}(0) \) is the parameter playing the role of the reduced temperature \( \epsilon \) in the case of the classical transition; \( \Delta_{BCS} \) is the BCS value of the gap at zero temperature in zero field, and \( N_0 \) is the one-electron density of states.

The microscopic theory provides (unlike the GL approach) a correct description of the short wavelength and high-frequency dynamic fluctuations for the complete magnetic field and temperature range. For instance, in
the region of QFs, but for non-zero temperatures $t \ll \hbar$ (see fig. 2) the AL and the anomalous MT contributions are equal to each other and grow as the square of temperature; moreover, one of them is exactly cancelled by the appearing negative contribution of the four DOS-like diagrams:

$$\delta \sigma_{xx}^{\text{AL}} = \delta \sigma_{xx}^{\text{MT(an)}} = -\delta \sigma_{xx}^{\text{DOS}} = \frac{4e^2 \gamma_0^2 t^2}{3\pi^2 \hbar^2}. \quad (6)$$

While eq. (4) defines the characteristic wavelength $\xi_{\text{GL}}(T)$ of the fluctuation modes close to $T_{c0}$, eq. (5) defines the characteristic coherence time $\tau_{\text{QF}}(\hbar)$ of QFs near $H_{c2}(0)$ (where $t \ll \hbar$). The integral there is determined by its lower cut-off $\omega_{\text{QF}} \sim \Delta_{\text{BCS}} \hbar$, and the corresponding time scale is $\tau_{\text{QF}} \sim \hbar(\Delta_{\text{BCS}} \hbar)^{-1}$. One sees that the functional form of $\tau_{\text{QF}}$ is analogous to that of $\tau_{\text{GL}}$, this can be obtained also from the uncertainty principle. The energy, characterizing the proximity to $H_{c2}(0)$, is $\Delta E = \hbar \omega_{\text{QF}}(H) - \hbar \omega_{\text{QF}}(H_{c2}(0)) \sim \Delta_{\text{BCS}} \hbar$ in this case. However, the spatial scale of QFs close to $H_{c2}(0)$ cannot be found from the propagator describing QFs, since the latter in the Landau representation does not contain space variables. Nevertheless, the spatial coherence scale can be estimated from the value of $\tau_{\text{QF}}$ analogously to consideration near $T_{c0}$. Namely, two electrons with the coherent phase starting from the same point get separated by the distance $\xi_{\text{QF}} \sim (D_{\text{QF}})^{1/2} \sim L_{\text{BCS}} \sqrt{\hbar}$ after the time $\tau_{\text{QF}}$. During this time they participate in multiple fluctuating Cooper pairings maintaining their coherence.

To clarify the physical meaning of $\tau_{\text{QF}}$ and $\xi_{\text{QF}}$, note that near the quantum phase transition at zero temperature, where $H \rightarrow H_{c2}(0)$, the fluctuations of the order parameter $\Delta^{(0)}(r, t)$ become highly inhomogeneous, contrary to the situation near $T_{c0}$. Indeed, below $H_{c2}(0)$, the spatial distribution of the order parameter at finite magnetic field reflects the existence of Abrikosov vortices with average spacing (close to $H_{c2}(0)$ but in the region where the notion of vortices is still adequate) equal to $\xi_{\text{BCS}} \sqrt{H/H_{c2}(0)}$. Therefore, one expects that close to and above $H_{c2}(0)$ the fluctuation order parameter also varies over the scale of $\xi_{\text{BCS}}$. At fields higher than but not too far from $H_{c2}(0)$ one can expect that these “vortex-like” spatial inhomogeneities are preserved over the time scale $\tau_{\text{QF}}$, whereas $\Delta^{(d)}(r, t)$ varies over the spatial scale $\xi_{\text{BCS}}$. The coherence length $\xi_{\text{QF}}$ is thus a characteristic size of a cluster of coherently rotating (due to the strong magnetic field) FCPs, and $\tau_{\text{QF}}$ estimates the lifetime of such flickering clusters (see fig. 3).

In terms of these introduced QF characteristics $\tau_{\text{QF}}$ and $\xi_{\text{QF}}$ one can understand the meaning of already found microscopic QF contributions and derive others which are required. For example, the physical meaning of eq. (6) can be understood as follows: one can estimate the FCP conductivity by merely replacing $\tau_{\text{GL}} \rightarrow \tau_{\text{QF}}$ in the classical AL formula, which would give $\delta \sigma_{xx}^{\text{AL}} \sim \epsilon^2 \tau_{\text{QF}}$. Nevertheless, as we already noticed, the FCPs at zero temperature cannot move in the electric field but only rotate. As temperature deviates from zero, the FCPs can change their state due to the interaction with the thermal bath, i.e. their hopping to an adjacent rotation trajectory along the applied electric field becomes possible. This means that they can participate in longitudinal charge transfer. This process can be mapped onto the paraconductivity of a granular superconductor [16] at temperatures above $T_{c0}$, where the FCP tunnelling between grains occurs in two steps: first one electron jumps, then the second follows. The probability of each hopping event is proportional to the inter-grain tunneling rate $\Gamma$. To conserve the superconductive coherence between both events, the latter should occur during the FCP lifetime $\tau_{\text{GL}}$. The probability of FCP tunnelling between two grains is determined as the conditional probability of two one-electron hopping events and is proportional to $W_T = \Gamma^2 \tau_{\text{GL}}$. Coming back to the situation of FCPs above $H_{c2}(0)$, one can identify the tunnelling rate with temperature $T$ while $\tau_{\text{GL}}$ corresponds to $\tau_{\text{QF}}$. Therefore, in order to get a final expression, $\delta \sigma_{xx}^{\text{AL}}$ should be multiplied by the probability factor $W_{\text{QF}} = \tau_{\text{QF}}^2 \tau_{\text{QF}}$ of the FCP hopping to the neighboring trajectory: $\delta \sigma_{xx}^{\text{AL}} \sim \delta \sigma_{xx}^{\text{AL}} W_{\text{QF}} \sim \epsilon^2 t^2 / \hbar^2$, which corresponds to the low-temperature asymptotic eq. (6).

The total fluctuation contribution to conductivity $\delta \sigma_{xx}^{\text{(tot)}}$ at $t \ll \hbar$ turns out to be negative and diverges logarithmically when the magnetic field approaches $H_{c2}(0)$:

$$\delta \sigma_{xx}^{\text{(tot)}} = -\frac{2e^2}{3\pi^2} \ln \frac{1}{\hbar} - \frac{2\gamma E \epsilon^2 t}{3\pi^2 \hbar} + O\left(\left(\frac{t}{\hbar}\right)^2\right). \quad (7)$$

The numerical analysis of eqs. (1)–(3) shows that the asymptotic eq. (7) is valid only within the narrow field range $\hbar \lesssim 10^{-6}$. The nontrivial fact following from eqs. (1)–(3) is that an increase of temperature at a fixed value of the magnetic field in this domain results in a non-monotonic behavior of the fluctuation part of the conductivity [11] (see fig. 4), observed in experiments [17–19].

Now we estimate the contributions of QFs to different characteristics of the SC in the vicinity of $H_{c2}(0)$. Applying the Langevin formula $\chi = \epsilon^2 (n/mc)(R^2)$ to a coherent cluster and taking $n_{c.p.} / m_{c.p.} \sim 1$, $R^2 \sim \xi_{\text{QF}}^2(\hbar)$,
one finds the fluctuation magnetic susceptibility \( \chi_{\text{FL}} \sim \frac{\sigma^2_{\text{BCS}}}{c_0 h} \) in agreement with ref. [11]. One further reproduces the contribution of QF to the Nernst coefficient [13]. Using \( \nu \sim \frac{\sigma}{c_0 h} \) and identifying the chemical potential of FCP, \( \mu_{\text{FCP}} \), as \( h \omega_c(H_{\text{c2}}(0)) - h \omega_c(H) \) (as in ref. [13], close to \( T_{\text{c0}} \), \( \mu_{\text{FCP}} = T_{\text{c0}} - T \), one finds that \( \delta \omega_{\text{FCP}}/dT \sim \delta H = \frac{\Delta}{2} + T/\Delta_{\text{BCS}} \) and \( \nu^{\text{AL}} \sim \frac{\sigma^2_{\text{BCS}}}{c_0 h} \). One can also predict the non-monotonic dependence of the transverse fluctuation conductivity \( \delta \sigma_{zz}^{(\text{tot})}(h,0) \) above \( H_{\text{c2}}(0) \) for layered SCs in a field perpendicular to the layers. The motion of FCPs along the z-axis has hopping character, and applying the above mapping procedure one finds: \( \delta \sigma_{zz}^{(h,0)} \sim \delta \sigma_{zz}^{(h,0)} - (1/\Gamma/E_F) c^2 \ln(1/h) \). As a result \( \delta \sigma_{zz}^{(\text{tot})} \sim c^2 T^2 / h^2 - (1/\Gamma/E_F) c^2 \ln(1/h) \) explaining the strong growth of the transversal magneto-resistance in organic superconductors at the edge of the transition at very low temperatures [20].

Finally, as an example of the practical use for our results, we fitted our general eqs. (1)–(3) to resistivity measurements in thin disordered indium oxide films, presented in ref. [17]. Figure 4 shows the low-temperature data for one sample (referred to as “Weak” in ref. [17]) of a film with thickness 30 nm, transition temperature \( T_{\text{c0}} = 3.35 \) K and critical magnetic field \( B_{\text{c2}}(0) = 13 \) T. The resistivity was measured, depending on magnetic field, for low temperature values \( T = 200, 300, 400, 500 \) mK. We plotted the theoretical expression for \( \delta \sigma_{zz}^{(\text{tot})} \) using the fitting parameter values \( B_{\text{c2}}(0) = 13.7 \) T, \( T_{\text{c0}} \sigma_0 = 5 \pm 1 \), and the experimentally found value of \( T_{\text{c0}} = 3.35 \) K. Overall the fitted FC curves show good agreement with the results of the measurements. In a forthcoming work we will show how our results can be used for the analysis of novel studies of superconductors in ultra-high magnetic fields [18], for the separation of quantum corrections in the vicinity of the superconductor–insulator transition [19], and the precise definition of the critical temperature and extraction of the temperature dependence of \( \sigma_0(T, H) \).

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