Superconductivity in Disordered Thin Films: Giant Mesoscopic Fluctuations

M. A. Skvortsov* and M. V. Feigel'man

L. D. Landau Institute for Theoretical Physics, Moscow 119334, Russia

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We discuss the intrinsic inhomogeneities of superconductive properties of uniformly disordered thin films with a large dimensionless conductance g. It is shown that mesoscopic fluctuations, which usually contain a small factor $1/g$, are crucially enhanced near the critical conductance $g_{cF} \gg 1$ where superconductivity is destroyed at $T = 0$ due to Coulomb suppression of the Cooper attraction. This leads to strong spatial fluctuations of the local transition temperature and thus to the percolative nature of the thermal superconductive transition.

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Since the very early stages of superconductivity theory it has been known [1,2] that the superconducting transition temperature, T_c, is insensitive to the rate τ^{-1} of elastic impurity scattering; i.e., it does not depend on the parameter $\tau T_c/\hbar$. This statement, known as the "Anderson theorem," is valid provided that both (i) Coulomb interaction effects and (ii) mesoscopic fluctuations are negligible. However, in sufficiently disordered metals, close to the Anderson localization transition, these effects become important and the Anderson theorem is violated.

In disordered samples, Coulomb repulsion enhanced because of the diffusive character of electron motion [3] leads to the suppression of T_c with the increase of disorder (cf. Ref. [4] for a review). For two-dimensional (2D) thin films with the dimensionless conductance $g = 2\pi\hbar/e^2 R_\square \gg 1$, the first-order perturbative correction had been calculated in Ref. [5], and the general expression for $T_c(g)$ was obtained by Finkel'stein [6]. In the leading order over $g^{-1/2}$ his result reads

$$\frac{T_c \tau_*}{\hbar} = \left[\frac{\sqrt{2\pi g} - \ln(\hbar/T_{c0}\tau_*)}{\sqrt{2\pi g} + \ln(\hbar/T_{c0}\tau_*)}\right]^{\sqrt{\pi g/2}}, \tag{1}$$

where T_{c0} is the transition temperature in the clean ($g \to \infty$) system, and $\tau_* = \max\{\tau, \tau(d/l)^2\}$, with d being the film thickness and $l = v_F \tau$ being the mean free path. According to Eq. (1), T_c vanishes at the critical conductance $g_{cF} = \ln^2(\hbar/T_{c0}\tau_*)/(2\pi)$ (which is supposed to be large enough for the theory to be self-consistent).

Finkel'stein's theory is an extended version of the mean-field theory of superconductivity which takes into account that the effective Cooper attraction acquires (due to Coulomb interaction and slow diffusion) a negative energy-dependent contribution. Within this ("fermionic") mechanism, the vanishing of T_c is accompanied by the vanishing of the *amplitude* of the superconductive order parameter Δ. Another ("bosonic") mechanism of T_c suppression [7] is due to *phase fluctuations* of the order parameter. This mechanism seems to be adequate mainly for structurally inhomogeneous superconductors (granular films of artificial arrays) with well-defined superconductive grains interconnected by weak links. Below we see, however, that phase fluctuations inevitably become relevant for homogeneously disordered films with the conductance g close to its critical value g_{cF}.

Another phenomenon important for 2D conductors is known as *mesoscopic fluctuations* [8] and is due to the nonlocal interference of electron waves scattering on impurities. It was recognized by Spivak and Zhou [9,10] that similar fluctuations are pertinent also for the Cooper pairing susceptibility, $K(\mathbf{r}, \mathbf{r}') = \langle K(\mathbf{r} - \mathbf{r}')\rangle + \delta K(\mathbf{r}, \mathbf{r}')$, which enters the BCS self-consistent equation

$$\Delta(\mathbf{r}) = \frac{\lambda}{\nu} \int K(\mathbf{r}, \mathbf{r}')\Delta(\mathbf{r}')d\mathbf{r}', \tag{2}$$

where λ is the dimensionless Cooper coupling constant and ν is the single-particle density of states per spin. Equation (2) with the *exact* disorder-dependent kernel $K(\mathbf{r}, \mathbf{r}')$ possesses localized solutions for $\Delta(\mathbf{r})$ *above* the mean-field transition line. They describe droplets of the superconducting phase that nucleate prior to the transition of the whole system. Since the relative magnitude of mesoscopic fluctuations of $\delta K(\mathbf{r}, \mathbf{r}')$ is of the order of $1/g$ and is small for a good metal, the effect of localized droplets on the *zero-field* superconductive transition is negligible and the transition width is determined by thermal fluctuations. On the contrary, at low temperatures near the upper critical field $H_{c2}(0)$, thermal degrees of freedom are frozen out and mesoscopic fluctuations are fully responsible for the width of the *field-driven* superconductor–normal-metal (SN) transition [10,11]. Still, the relative magnitude of the $H_{c2}(0)$ shift and of the transition width due to mesoscopic fluctuations are of the order of $1/g \ll 1$ as long as Coulomb effects are neglected.

The goal of the present Letter is to develop a combined theory of the superconductive transition in 2D disordered films, which takes into account both Coulomb effects and mesoscopic fluctuations. Our main result is the expression for the relative smearing $\delta_d = \delta T_c/T_c$ of the zero-field transition due to the formation of localized islands:

$$\delta_d = \frac{a_d}{g(g - g_{cF})}, \tag{3}$$

where $a_d \approx 0.4$. In the vicinity of Finkel'stein's critical point, at $g - g_{cF} \lesssim 1$, the formation of islands dominates over thermal fluctuations characterized by the Ginzburg number $\mathrm{Gi} = (\pi/8g)$ [12]. Moreover, in the very close vicinity of the quantum critical point, at $g - g_{cF} \lesssim 1/g_{cF}$, fluctuations of the "local transition temperature" become large on the absolute scale, $\delta T_c \sim T_c$, and the superconductive state becomes strongly inhomogeneous *in the absence of any predetermined structural granularity* [9].

We emphasize that mesoscopic fluctuations are minimal fluctuations that are inevitably present in any disordered system. In real samples, their effect may be enhanced by various types of structural inhomogeneities.

Ginzburg-Landau expansion.—We begin with deriving the Ginzburg-Landau (GL) expansion in the vicinity of the Finkel'stein transition temperature (1). The GL free energy for the static order parameter has the form

$$\mathcal{F}[\Delta] = \int\left(\alpha|\Delta|^2 + \frac{\beta}{2}|\Delta|^4 + \gamma|\nabla\Delta|^2\right)d\mathbf{r} + \tilde{\mathcal{F}}[\Delta], \quad (4)$$

where the first term is the disorder-averaged contribution, and the last term accounts for mesoscopic fluctuations; its form will be found later. The diagrams for the disorder-averaged free energy [13] are shown in Fig. 1. Apart from the standard impurity averaging, Cooperons should be averaged over fluctuations of the electric field (this is shown by the gray rectangle). The order parameter always enters in the combination $\langle K_\varepsilon(\mathbf{r} - \mathbf{r}')\rangle\Delta(\mathbf{r}')$, where we introduced the reduced Cooper kernel:

$$\langle K_{\varepsilon_k}(\mathbf{r} - \mathbf{r}')\rangle = T\sum_m \langle G_{\varepsilon_k,\varepsilon_m}(\mathbf{r}, \mathbf{r}')G_{-\varepsilon_k,-\varepsilon_m}(\mathbf{r}, \mathbf{r}')\rangle, \quad (5)$$

with $G_{\varepsilon_k,\varepsilon_m}(\mathbf{r}, \mathbf{r}')$ being the Matsubara exact Green function which, in the presence of the fluctuating electric field, depends on two energy arguments. After summation over energy Eq. (5) gives the (averaged) pairing susceptibility: $\langle K(\mathbf{r} - \mathbf{r}')\rangle = T\Sigma_\varepsilon\langle K_\varepsilon(\mathbf{r} - \mathbf{r}')\rangle$.

The kernel $\langle K_\varepsilon(\mathbf{r} - \mathbf{r}')\rangle$ obeys the linear equation shown diagrammatically in Fig. 2. To write it in a compact form we define the *Cooperon screening factor* $w_{\mathbf{q}}(\varepsilon)$:

$$\langle K_\varepsilon(\mathbf{q})\rangle = \frac{2\pi\nu}{Dq^2 + 2|\varepsilon|}w_{\mathbf{q}}(\varepsilon), \quad (6)$$

which shows how the free metallic Cooperon gets modified by the fluctuating electric field (D is the diffusion coefficient). The screening factor satisfies the equation

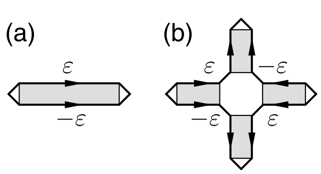

FIG. 1. Diagrams for the GL free energy: (a) $|\Delta|^2$ term; (b) $|\Delta|^4$ term; its central part is the Hikami box.

$$w_{\mathbf{q}}(\varepsilon_k) = 1 - T\sum_n \frac{\mathcal{L}_{\varepsilon_k\varepsilon_n}w_{\mathbf{q}}(\varepsilon_n)}{Dq^2 + 2|\varepsilon_n|}, \quad (7)$$

where (for thick films with $d > l$ replace τ by τ_*)

$$\mathcal{L}_{\varepsilon_k\varepsilon_n} = \frac{2}{g}\theta(\varepsilon_k\varepsilon_n)\ln\frac{1}{|\varepsilon_k + \varepsilon_n|\tau} \quad (8)$$

is the disorder-enhanced Coulomb interaction vertex [14] proportional to the return probability, with $\theta(x)$ being a unit step function.

Equation (7) looks pretty similar to the equation for the energy-dependent Cooper vertex $\Gamma_{\varepsilon,\varepsilon'}$ considered in Ref. [14]. It can easily be solved with logarithmic accuracy [14]. Introducing a new variable $\zeta = \ln(1/\varepsilon\tau)$, one finds for the zero-momentum limit of the screening factor [15]

$$w(\zeta) \equiv w_0(\varepsilon) = \cosh(\lambda_g\zeta) - \tanh(\lambda_g\zeta_T)\sinh(\lambda_g\zeta), \quad (9)$$

where $\zeta_T \equiv \ln(1/T\tau)$, and $\lambda_g = 1/\sqrt{2\pi g}$ is Finkel'stein's fixed point. The function $w_0(T)$ decreases from 1 at $\varepsilon \sim 1/\tau$ down to $w(\zeta_T) = 1/\cosh(\lambda_g\zeta_T)$ at $\varepsilon \sim T$.

The coefficients in the GL free energy (4) are given by

$$\frac{\alpha}{\nu} = \frac{1}{\lambda} = \pi T\sum_\varepsilon \frac{w_0(\varepsilon)}{|\varepsilon|} = \frac{1}{\lambda_*} - \int_0^{\zeta_T} d\zeta w(\zeta), \quad (10a)$$

$$\gamma = \frac{\pi\nu DT}{2}\sum_\varepsilon \frac{w_0^2}{\varepsilon^2} = \gamma_0 w^2(\zeta_T), \quad (10b)$$

$$\beta = \frac{\pi\nu}{2}\sum_\varepsilon \frac{w_0^4(\varepsilon)}{|\varepsilon|^3} = \beta_0 w^4(\zeta_T), \quad (10c)$$

where $\beta_0 = 7\zeta(3)\nu/(8\pi^2T_c^2)$ and $\gamma_0 = \pi\nu D/(8T_c)$ are the standard coefficients for dirty superconductors [13], and λ_* is the running Cooper coupling constant at the energy scale τ^{-1}. The Matsubara sums in Eqs. (10b) and (10c) converge at the thermal scale. Therefore the coefficients β and γ contain the screening factors evaluated at ζ_T. On the contrary, the coefficient α is determined by all energies $\varepsilon < 1/\tau$. The integral in Eq. (10a) is given by $\lambda_g^{-1}\tanh(\lambda_g\zeta_T)$, and when solving $\alpha(T_c) = 0$ one immediately recovers the Finkel'stein expression (1) for T_c. Taking the derivative near T_c, we find $\alpha = \nu(T/T_c - 1)w^2(\zeta_T)$.

Thus, disregarding mesoscopic fluctuations, we see that the Δ field always enters the GL expansion in the combination with the screening factor $w(\zeta_T) = 1/\cosh(\lambda_g\zeta_T)$. If we define $\tilde{\Delta} = \Delta w(\zeta_{T_c})$, then the GL expansion for $\tilde{\Delta}$ will

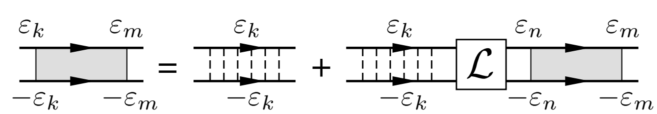

FIG. 2. Equation for the Cooperon in the fluctuating electric field. The Coulomb vertex $\mathcal{L}$ is given by Eq. (8). The kernel $\langle K_{\varepsilon_k}(\mathbf{r} - \mathbf{r}')\rangle$ is obtained after summation over ε_m.

acquire the standard form with the coefficients $\alpha_0 = \nu(T/T_c - 1)$, β_0, and γ_0. As a corollary, the Ginzburg number appears to be unaffected by the Coulomb repulsion: $\mathrm{Gi} = \pi/(8g)$.

Quasiparticle spectrum.—Incorporating the Coulomb screening factors $w_{\mathbf{q}}(\varepsilon)$ into the standard Green function formalism, one finds that a quasiparticle propagating with the energy ε feels an effective pairing potential $\Delta_{\mathrm{eff}}(\varepsilon) = w(\zeta)\Delta$. The gap in the spectrum, E_{gap}, should be found as a solution to $E_{\mathrm{gap}} = w[\ln(1/E_{\mathrm{gap}}\tau)]\Delta$. Deep in the superconducting phase, at $T_c - T \sim T_c$, it coincides with $\tilde{\Delta}$. On the other hand, excited particles with $\varepsilon > E_{\mathrm{gap}}$ see a larger value of $\Delta_{\mathrm{eff}}(\varepsilon)$, and high-energy particles with $\varepsilon \gtrsim 1/\tau$ feel the bare value of Δ.

In the regime of the strong Coulomb suppression of superconductivity, when $T_c \ll T_{c0}$ and $w(\zeta_{T_c}) \ll 1$, the bare Δ significantly exceeds the screened $\tilde{\Delta}$. This enhancement of Δ was irrelevant for the GL expansion since the sums over Matsubara energies that determined the GL coefficients converged at the thermal scale. We see below that this is not the case for mesoscopic fluctuations where the bare value of Δ comes into play.

Mesoscopic fluctuations of the pairing susceptibility.—In order to calculate the correlation function of the pairing susceptibility, one has to draw two diagrams for $K(\mathbf{r}, \mathbf{r}')$ [see Fig. 1(a)] and connect their diffusive modes by impurity lines. In general, the variance $\langle \delta K(\mathbf{r}_1, \mathbf{r}_2)\delta K(\mathbf{r}_3, \mathbf{r}_4)\rangle$ is a complicated function of $\mathbf{r}_i - \mathbf{r}_j$, decaying at the scale of the thermal length $L_T = \sqrt{D/(2\pi T)}$. On the other hand, close to T_c the order parameter varies at the scale of the coherence length $\xi(T) = L_T\sqrt{T_c/(T - T_c)} \gg L_T$. Therefore, in the vicinity of the superconducting transition the fluctuations of $K(\mathbf{r}, \mathbf{r}')$ are effectively *short ranged* and characterized by the single number

$$C = \int \langle \delta K(\mathbf{r}_1, \mathbf{r}_2)\delta K(\mathbf{r}_3, \mathbf{r}_4)\rangle d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4. \qquad (11)$$

Taking into account all possible correlations between diffusive modes in $K(\mathbf{r}, \mathbf{r}')$, we find

$$C = T^4 \sum_{\varepsilon_i > 0} \left(\prod_{i=1}^{4} \frac{w_0(\varepsilon_i)}{\varepsilon_i} \right) \hat{\mathcal{R}}_{\mathbf{q}}^{12} \hat{\mathcal{R}}_{\mathbf{q}'}^{34} M_{\varepsilon_i}(\mathbf{q}, \mathbf{q}'), \qquad (12)$$

where $\hat{\mathcal{R}}_{\mathbf{q}}^{ij}$ is an operator acting on an arbitrary function $X(\mathbf{q})$ as

$$\hat{\mathcal{R}}_{\mathbf{q}}^{ij} X(\mathbf{q}) = \frac{\delta_{\varepsilon_i \varepsilon_j}}{2T} X(0) + \frac{1}{\nu} \int \frac{d\mathbf{q}/(2\pi)^2}{[D\mathbf{q}^2 + \varepsilon_{ij}]^2} X(\mathbf{q}), \qquad (13)$$

$\varepsilon_{ij} \equiv \varepsilon_i + \varepsilon_j$, and $M_{\varepsilon_i}(\mathbf{q}, \mathbf{q}')$ is the 4-Cooperon–diffuson collision vertex shown in Fig. 3 (with the proper construction of internal Hikami boxes by drawing additional impurity lines being implied). The internal diffusive modes can be diffusons or Cooperons. In the vicinity of $H_{c2}(0)$, Fig. 3(b) was considered in Ref. [10], and 3(c) was analyzed in Ref. [16]. At zero magnetic field the vertex is calculated elsewhere [15]:

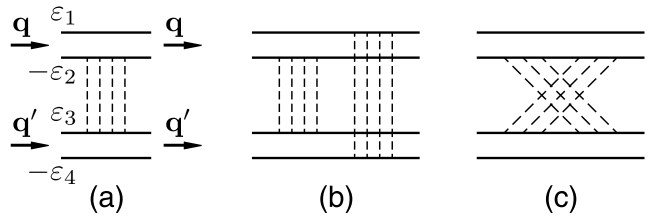

FIG. 3. One-loop diagrams for the 4-Cooperon–diffuson collision vertex $M_{\varepsilon_i}(\mathbf{q}, \mathbf{q}')$. The diagrams (a) and (c) have their symmetric counterparts.

$$M_{\varepsilon_i}(\mathbf{q}, \mathbf{q}') = \frac{[D\mathbf{q}^2 + \varepsilon_{12}][D\mathbf{q}'^2 + \varepsilon_{34}]}{2\pi D} \frac{\varepsilon_{14} + \varepsilon_{23}}{\varepsilon_{14}\varepsilon_{23}}. \qquad (14)$$

The first term in Eq. (13) refers to the Cooperons in the ladder (7) shown in Fig. 2, while its second term refers to the Cooperons or diffusons that are responsible for the return probability in the vertex $\mathcal{L}$ given by Eq. (8). Some summations over energies in Eq. (12) saturate at the thermal scale, whereas summations associated with the return probability are logarithmic, extending up to the high-energy cutoff τ^{-1}:

$$C = \frac{w_0^2(T)}{\pi D} \left(T^2 \sum_{\varepsilon, \varepsilon' > 0} \frac{1}{\varepsilon\varepsilon'(\varepsilon + \varepsilon')} \right) \times \left[w_0(T) + \frac{1}{g} T \sum_{\varepsilon > 0} \frac{w_0(\varepsilon)}{\varepsilon} \ln\frac{1}{\varepsilon\tau} \right]^2. \qquad (15)$$

The second term in the square brackets in Eq. (15) is due to mesoscopic fluctuations of return probability. Calculating the sum as an integral over ζ with the help of Eq. (9), we find that this term is equal to $1 - w_0(T)$, so that the total expression in the square brackets in Eq. (15) is equal to 1. Thus, in the regime of strong Coulomb suppression of superconductivity, the pairing susceptibility fluctuates mainly because of mesoscopic fluctuations of return probability in the vertex (8). Writing $C = C_0 w_0^4(T)$, we get

$$C_0 = \frac{7\zeta(3)}{8\pi^4 DT} \cosh^2(\lambda_g \zeta_T). \qquad (16)$$

Superconductor with fluctuating T_c.—Short-range mesoscopic fluctuations of $\delta K(\mathbf{r}_1, \mathbf{r}_2)$ are equivalent to local fluctuations of the transition temperature $\delta T_c(\mathbf{r})$, which can be described by the following term in the GL free energy:

$$\tilde{\mathcal{F}}[\tilde{\Delta}] = \int \delta\tilde{\alpha}(\mathbf{r}) |\tilde{\Delta}(\mathbf{r})|^2 d\mathbf{r}, \qquad (17)$$

where $\langle \delta\tilde{\alpha}(\mathbf{r})\delta\tilde{\alpha}(\mathbf{r}')\rangle = C_0 \delta(\mathbf{r} - \mathbf{r}')$. Superconductors with local fluctuations of T_c were considered previously within the phenomenological approach by Ioffe and Larkin [17], where the three-dimensional case was mainly discussed. Generalizing their results to the 2D case, we find that the relative smearing of the superconductive transition due to frozen-in mesoscopic fluctuations is given by $\delta_d = C_0/[12T_c\gamma_0(d\alpha_0/dT)]$ [note that in the 2D case the numerical coefficient in the exponent of Eq. (29) in Ref. [17] is equal to 11.8; cf. [18]]. Taking C_0 at $T = T_c$ and using Eq. (1), we obtain a surprisingly simple expression (3) with $a_d = 28\zeta(3)/3\pi^3$.

The increase of δ_d near the critical conductance g_{cF} can be understood in terms of the renormalization of the Cooper attraction constant λ. At low energies, the latter acquires a negative Coulomb contribution proportional to the return probability $\sim g^{-1}\ln(1/\epsilon\tau)$. Mesoscopic fluctuations of g lead then to fluctuations of λ, whose relative effect grows with decreasing λ: $\delta_d \sim \delta T_c/T_c = \delta\lambda/\lambda^2$.

Equation (3) predicts that for $g - g_{cF} \lesssim 1$, the disorder-induced broadening of the transition dominates over the thermal width: $\delta_d > \mathrm{Gi}$. In such a situation, the macroscopic superconductive transition occurs via formation of small superconductive islands of size $L_T = \sqrt{D/(2\pi T_c)}$ surrounded by the normal-metal state. With the temperature decrease, the density of these islands and the proximity-induced coupling between them grows until a percolation-type superconductive transition [17] takes place. At sufficiently low temperatures, $T \lesssim T_c(1 - \delta_d)$, the superconductive state becomes approximately uniform, with weak spatial variations in the amplitude of the order parameter $|\Delta|$.

Another situation occurs in the closest vicinity of the critical conductance, $g - g_{cF} \lesssim 1/g_{cF}$: here local fluctuations of T_c are large on the absolute scale, and strong inhomogeneity of the superconductive order parameter persists down to $T \ll T_c$. As a result, both thermal and quantum fluctuations of phases of superconductive order parameters on different superconductive islands are strongly increased. In other terms, in the close vicinity of the critical conductance g_{cF}, the *bosonic* mechanism of superconductivity suppression becomes relevant.

The inhomogeneous distribution of $|\Delta(\mathbf{r})|$ is known [19,20] to smear the gap in the excitation spectrum of a superconductor. We expect this effect to be very strong for $g \approx g_{cF}$.

The result (3) indicates that strong enhancement (in comparison with the results of Refs. [10,11]) of mesoscopic fluctuation effects in H_{c2} behavior at low temperature should be expected at $g \approx g_{cF}$. This problem needs special treatment since the correlation length of mesoscopic fluctuations diverges at $T \to 0$, and short-range approximation employed for the determination of C in Eq. (11) becomes inappropriate [16]. It is quite clear, however, that long-range features of mesoscopic disorder at $T \to 0$ should increase the effective width of the field-driven $T = 0$ SN transition in comparison with the width δ_d of the zero-field transition driven by temperature. An extension of the present approach to the magnetic-field-induced transition near $H_{c2}(0)$ will be considered separately.

To conclude, we demonstrated that strong inhomogeneities of the superconductive state can be induced by relatively weak ($\sim g^{-1}$) mesoscopic fluctuations, which lead to spatial fluctuations of the effective Cooper attraction constant. As a result, a nominally uniformly disordered film may appear as a granular one in terms of its superconductive properties.

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*Electronic address: skvor@itp.ac.ru

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