Enhancement of the finite-frequency superfluid response in the pseudogap regime of strongly disordered superconducting films

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Supplementary Material

Aslamazov-Larkin and Maki-Thompson expressions for fluctuation conductivity

The correction to the conductivity due to fluctuating Cooper pairs is given by Aslamazov-Larkin (AL) theory. The Maki-Thompson (MT) correction, coming from the coherent scattering of the electrons forming the Cooper pair, is normally suppressed in the presence of strong disorder. The contribution of AL and MT fluctuations to the dc conductivity in 2D and 3D are given by^{1,2}

$$\sigma_{dc}^{2D-AL} = \frac{1}{16} \frac{e^2}{\hbar t} \varepsilon^{-1}$$
(1)

$$\sigma_{dc}^{3D-AL} = \frac{1}{32} \frac{e^2}{\hbar \xi_0} \varepsilon^{-1/2}$$
(2)

$$\sigma_{dc}^{2D-MT} = \frac{1}{8} \frac{e^2}{\hbar t} \frac{1}{\varepsilon - \delta} \ln\left(\frac{\varepsilon}{\delta}\right)$$
(3)

$$\sigma_{dc}^{3D-MT} = \frac{1}{8} \frac{e^2}{\hbar \xi_0} \varepsilon^{-1/2}$$
(4)

where $\varepsilon = \ln(T/T_c)$, *t* is the thickness of the sample and ξ_0 is the BCS coherence length and δ is the Maki-Thompson pair breaking parameter. The two contributions are additive.

The frequency dependence of the fluctuation conductivities are as follows.

Aslamazov-Larkin³:

$$\sigma^{2DAL}(\omega) = \sigma_{DC}^{2DAL} S^{2DAL} \left(\frac{\omega}{\omega_0} \right); \omega_0 = \frac{16k_B T_c}{\pi \hbar} \varepsilon$$

$$S^{2DAL}(x) = \left\{ \frac{2}{x} \tan^{-1} x - \frac{1}{x^2} \ln(1 + x^2) \right\} + i \left\{ \frac{2}{x^2} (\tan^{-1} x - x) + \frac{1}{x} \ln(1 + x^2) \right\}$$
(5)

$$\sigma^{3DAL}(\omega) = \sigma_{DC}^{3DAL} S^{3DAL} \left(\frac{\pi \hbar \omega}{16k_B T_c \varepsilon} \right)$$

$$S^{3DAL}(x) = \left\{ \frac{8}{3x^2} \left(1 - (1 + x^2)^{3/4} \cos(\frac{3}{2} \tan^{-1} x) \right) \right\} + i \left\{ \frac{8}{3x^2} \left(-\frac{3}{2} x + (1 + x^2)^{3/4} \sin(\frac{3}{2} \tan^{-1} x) \right) \right\}$$
(6)

Aslamazov-Larkin and Maki-Thompson⁴:

$$\sigma^{2DAL+MT}(\omega) = \sigma_{DC}^{2DAL} S^{2DAL+MT} \left(\frac{\pi \hbar \omega}{16k_B T_c \varepsilon} \right)$$

$$S^{2DAL+MT}(x) = \left\{ \operatorname{Re} S^{2DAL}(x) + \frac{2\pi x - 2\ln(2x)}{1 + 4x^2} \right\} + i \left\{ \operatorname{Im} S^{2DAL}(x) + \frac{\pi + 4x\ln(2x)}{1 + 4x^2} \right\}$$

$$\sigma^{3DAL+MT}(\omega) = \sigma_{DC}^{3DAL} S^{3DAL+MT} \left(\frac{\pi \hbar \omega}{16k_B T_c \varepsilon} \right)$$
(8)

$$S^{3DAL+MT}(x) = \left\{ \operatorname{Re} S^{3DAL}(x) + \frac{4 - 4x^{1/2} + 8x^{3/2}}{1 + 4x^{1/2}} \right\} + i \left\{ \operatorname{Im} S^{3DAL}(x) + \frac{4x^{1/2} - 8x + 8x^{3/2}}{1 + 4x^{1/2}} \right\}$$
(6)

(We follow the same notation as in ref. 5.)

In Figure 1s, we show below the scaled phase and amplitude for the sample with 15.71 K along with the predicted theoretical variation from eq. (5)-(8).



Figure 1s. Scaled (a) phase and (b) amplitude of the fluctuation conductivity for NbN thin film with $T_c \sim 15.71$ K along with theoretical predictions for AL and AL+MT theory in 2D and 3D.

The fact that our samples follow a 2D scaling behavior is not surprising, because even though their zero-temperature coherence length $\xi_0 \sim (4-8)$ nm is much smaller than the films thickness

t=50 nm, what matters for the dimensionality of the fluctuations in the temperature-dependent correlation length $\xi(T) = \xi_0/\sqrt{\varepsilon}$. Indeed, following Ref. [3,4], one can see that as soon as $\xi(T) >> t/\pi$ the film behaves effectively as a two-dimensional system. We can then estimate the crossover reduced temperature below which the system behaves as 2D as $\varepsilon_c = (\pi \xi_0/t)^2$. For the sample having $T_c \sim 15.71$ K, using $\xi_0 \sim 4$ nm [Ref. 6], this corresponds to $\varepsilon_c \approx 0.063$, i.e. T = 16.7K. Since the scaling works for much lower T and ε values (T<15.85 K, ε < 0.01) it is not surprising that we find 2D behavior. For the sample with $T_c \sim 3.14$ K, using $\xi_0 \sim 7$ nm [Ref. 6], one would obtain $\varepsilon_c \approx 0.19$, i.e. T = 3.75 K. However, the scaling in this sample occurs continuously over a much larger temperature range, and below $T \sim 4$ K one starts to see significant deviations from the 2D AL curve with ω_0 two order of magnitude smaller than ALtheory prediction, showing the complete failure of such a scaling in this case.





Figure 2s. (a) Superfluid stiffness of the T_c =3.14 K sample as given in Eq. (9) at various frequencies. The BKT transition is expected to occur when the zero-frequency stiffness intersect the universal line $2T/\pi$, see Eq. (9). Nonetheless, the effect of vortex-antivortex pairs is expected to occur at a lower temperature T_v , where the frequency dependence of the data starts to develop. (b) Superfluid stiffness of a thin 3nm NbN sample taken from Ref. [7]. In this case the temperature T_V can be easily identified by the rapid downturn of the data with respect to the BCS fit valid far from the transition. In both panels the dashed vertical line marks the dc T_c .

The possible fluctuation scenario that we propose in the manuscript focuses mainly on longitudinal phase fluctuations between domains. Indeed, as we mention in the manuscript, even

if transverse (vortical) phase fluctuations of BKT type are at play they are expected to be relevant only in a small range of temperatures above T_c , as we demonstrated in a recent analysis of the BKT transition in thin (*t*~few nanometers) films in Ref.[7]. Here we show in more details why not only the ordinary GL theory but also the standard BKT one fails in explaining the fluctuation regime at strong disorder. We focus thus on the most disordered sample, $T_c \sim 3.14$ K where deviations from AL theory are more evident. First of all, in Fig. 2s(a) we plot the superfluid density at various frequencies, to check for any signature of the so-called BKT jump, which is expected to occur⁷ in the zero-frequency limit when:

$$J = \frac{\hbar^2 n_s t}{4m} = \frac{2T}{\pi} \tag{9}$$

Notice that, with respect to Eq. (1) of the manuscript, we replaced a with the film thickness t, since if any BKT transition occurs it involves the whole film as an effective 2D system. When the superfluid stiffness is probed at finite frequency as in our case the universal jump (9) is expected to be smeared out^{8,9}. In particular, data taken at different frequencies start to deviate from each other at the temperature T_V where (bound) vortex-antivortex pairs become thermally excited. As we discussed in the case of thin films⁹, this temperature T_V is usually smaller than the real BKT critical temperature due to a small value of the vortex-core energy. This can be seen in Fig. 2s(b) where we report for comparison also the data of Ref. [7] in a 3nm thick NbN sample. Here T_V can be easily identified by the temperature where experimental data deviate from the BCS fit valid at lower temperatures. Notice that the downturn of J at T_V observed in this thin film (panel b) is much more pronounced than the smooth temperature dependence observed in our thick sample (panel a) even at the lowest accessible frequency. In the case of the 3nm sample one can also estimate the mean-field BCS critical temperature T_c^0 by extrapolating the BCS fit. As it has been demonstrated in Ref. [7], the BKT fluctuations extend only up to T_c^0 . As one can see in Fig. 2s(b), the BKT regimes is then less than 1K wide. Thus, in our much thicker samples this range of BKT fluctuations is expected to be even smaller, consistently with the fact that T_c approaches rapidly T_c^0 when the film thickness increases⁷.



Figure 3s. (a)-(b) Rescaled phase (a) and amplitude (b) of $\sigma_{\rm fl}(\omega)$ using the dynamic scaling analysis on the film with $T_{\rm c} \sim 3.14$ K. The solid line shows the prediction from BKT theory. The color coded temperature scale for the scaled curves is shown in panel (b). (c) Plot of the scaling frequency ω_0 for the $T_c = 3.14$ K sample as a function of $t_r^{-1/2}$. Notice that a BKT fit $\Omega(T) = \Omega_0 \exp\left(-2b/\sqrt{t_r}\right)$ (red line) works only asymptotically near to T_c , demonstrating the existence of a very small BKT fluctuation regime.

To further support this conclusion, we show explicitly how the BKT predictions for the scaling function *S* fails to reproduce the data, except eventually an extremely small temperature range near T_c . Let us recall that within BKT theory the fluctuation conductivity can be written as⁸:

$$\sigma = \frac{4e^2}{\hbar^2 d} \frac{J_0(T)}{\Omega(T)} \frac{1}{1 - i\omega/\Omega(T)} = \frac{4e^2}{\hbar^2 d} \frac{J_0(T)}{\Omega(T)} S(\omega/\Omega(T)), \quad S(x) = \frac{1}{1 - ix}$$
(10)

where

$$\Omega(T) = \frac{4\pi^2 J_0(T)}{k_B T} \frac{D_V}{\xi^2(T)}$$
(11)

Here $J_0(T)$ is the "bare" superfluid density, i.e. the one the system would have in the absence of vortex fluctuations, $D_V \sim \hbar/m \sim 10^{-4} m^2/s$ is the vortex diffusion constant⁹ and $\xi(T)$ is the BKT correlation length. A direct comparison with the data with *S* (eq. (10)) shows (Fig 3s(a-b)) that the scaled amplitude and phase do not follow the prediction from BTK theory. In passing, we also note that the value of the vortex diffusion constant is of the same order of the electron diffusion constant. In this respect, the estimate of L_0 reported in the manuscript is independent on the possible specific nature (longitudinal vs transverse) of the phase fluctuations in the pseudogap state. However, as we shall further argue below, the comparison with the data does not support such a vortex-fluctuations scenario far from the transition.

To get a better insight into the nature of the failure of the BKT scaling, let us analyze more quantitatively the predictions of Eqs. (10)-(11). For instance, from Eq. (10) one can also identify Ω with the scaling frequency ω_0 extracted from the experiments. Near T_c the main temperature

dependence of Ω comes from the BKT correlation length, which is expected to diverge exponentially^{7,8,10} at T_c as

$$\xi(T) = A\xi_0 \exp\left(b / \sqrt{t_r}\right), \qquad t_r = \frac{T - T_c}{T_c}$$
(12)

where the parameter *b* is related¹⁰ to the distance between the BKT temperature T_c and the mean-field one T_c^0 , and to the value of the vortex-core energy μ :

$$b = \frac{4}{\pi^2} \frac{\mu}{J_0} \sqrt{t_c}, \quad t_c = \frac{T_c^0 - T_c}{T_c}$$
(13)

By inserting Eq. (12) into eq. (11) we find that near T_c , $\Omega(T)$ is expected to scale as $\Omega(T) = \Omega_0 \exp\left(-\frac{2b}{\sqrt{t_r}}\right)$. In Fig. 3s(c) we show a plot of $\omega_0(T)$ versus $t_r^{-1/2}$ from which one can extract the value of *b* to be compared with Eq. (13) above. As one can see, in contrast to what found for example in InO_x^{-11} , $\Omega(T)$ does not seem to really follow the BKT functional form, except eventually for very few points near T_c . By fitting this regime one would obtain *b*~0.045. When compared with the relation (13) above, which has been successfully verified in thin NbN films⁷, one can estimate the BKT fluctuation range t_c . Indeed, as it has been shown in Ref. [7], in NbN $\mu/J_0 \sim 1$, so that *b*~0.045 corresponds to a value of $t_c \sim 0.012$, so that the BKT regime would be limited up to a temperature $T_c^0 \sim 3.18$. This result is consistent with the rapid shrinking of the BKT fluctuations regime with increasing film thickness reported in Ref. [7].

Let us also show that BKT physics cannot explain the slowing down of fluctuations observed in the pseudogap regime, i.e. the observation of a low value of the scaling frequency ω_0 at temperatures very far from T_c . If one wants to compare again ω_0 with the BKT scaling frequency $\Omega(T)$ (11), one must notice that the expression (12) for the BKT correlation length is only valid very near to T_c . Indeed, assuming that fluctuations retain a BKT character very far from it, the correct temperature dependence of the correlation length extracted from the renormalizationgroup analysis of the BKT transition is¹²

$$\xi(T) \cong \xi_0 \exp\left(\mu / k_B T\right) \tag{14}$$

To give a *lower-bound* estimate of the corresponding Ω for example at T=5K we can approximate J_0 in Eq. (11) with the one measured at the highest frequency accessible experimentally, i.e $J_0(T=5 \text{ K})\sim J(20 \text{ Ghz}, T=5 \text{ K})\sim 10^{-3} \text{ K}$. Nonetheless, by using $\xi_0 \sim 7 \text{ nm}$, the expected value $D_V \sim 10^{-4} \text{ m}^2/\text{s}$ and Eq. (14) for the correlation length (with $\mu/J_0 \sim 1^7$) we obtain

$$\Omega(T) \sim \frac{4\pi^2 J_0(T)}{k_B T} D_V \exp(-J_0(T)/k_B T) \sim 16 \text{ GHz}$$
(15)

This value, which is a lower bound for the true Ω , is still one order of magnitude *larger* than the scaling frequency ω_0 observed experimentally. This demonstrates that the enhanced fluctuation regime observed in the pseduogap regime cannot be accounted for by the standard BKT approach.

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