Correlated Conductance Fluctuations Close to the Berezinskii-Kosterlitz-Thouless Transition in Ultrathin NbN Films

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We probe the presence of long-range correlations in phase fluctuations by analyzing the higher-order spectrum of resistance fluctuations in ultrathin NbN superconducting films. The non-Gaussian component of resistance fluctuations is found to be sensitive to film thickness close to the transition, which allows us to distinguish between mean field and Berezinskii-Kosterlitz-Thouless (BKT) type superconducting transitions. The extent of non-Gaussianity was found to be bounded by the BKT and mean field transition temperatures and depends strongly on the roughness and structural inhomogeneity of the superconducting films. Our experiment outlines a novel fluctuation-based kinetic probe in detecting the nature of superconductivity in disordered low-dimensional materials.

The transition from a superconducting to a normal state in two dimensions is known to occur via the Berezinskii-Kosterlitz-Thouless (BKT) mechanism. Traditionally, the signature of the BKT transition is found by measuring the superfluid density where a discontinuity is observed at the critical temperature T_{BKT} [1–6]. This method is limited in its scope of application for systems exhibiting interfacial or buried superconductivity like oxide heterostructures due to the inability to measure the thickness of the superconducting layer accurately. Transport-based probes including discontinuity in the power law behavior of I-V characteristics or change in the curvature of magnetoresistance from convex to concave in the presence of a perpendicular magnetic field [7–12] have their limitations as real systems contain some degree of inhomogeneity which smear out the signatures of BKT type behavior. With growing interest in low-dimensional superconductivity, it is interesting to develop new probes or techniques that are not only sensitive enough to detect BKT transition but also compare its characteristics scales with the mean field description.

The BKT transition, as exhibited by ultrathin superconducting films, is characterized by the unbinding of vortex pairs beyond T_{BKT}. It is well known that these vortices exhibit long-range interactions which vary logarithmically with the distance between the vortices. Vortices also occur in bulk type-II superconductors in the presence of an external magnetic field. Earlier experiments probing vortices in bulk films through measurements of flux flow noise and voltage noise have reported the presence of broadband noise [13,14] as a function of driving current and magnetic field. The statistics of noise in these systems is non-Gaussian due to the fact that most of the noise arises from very few fluctuators. Noise measurements on quasi-2D MgB$_2$ films show a possibility of thermally induced vortex hopping

through measurements of flux noise close to BKT transition [15]. To the best of our knowledge, there are no reports on higher-order statistics of fluctuations looking into the possibility of interacting vortices in two dimensions.

Higher-order fluctuations in resistivity have been established as a useful tool to study the presence of long-range correlations in systems undergoing both electronic and structural phase transitions. Experiments on a wide variety of systems like doped silicon [16], metal nanowires [17], shape memory alloys [18,19], disordered magnetic alloys [20], etc., have shown that time dependent fluctuations (noise) in physical observables exhibit a strong non-Gaussian behavior due to the presence of long-range correlations originating from Coulomb forces, internal strain fields, or magnetic interactions. The origin of non-Gaussian noise can be understood from the following simple picture: In a system composed of many independent fluctuators, the resultant noise has a Gaussian nature as expected from the central limit theorem. However, the non-Gaussian component increases when the correlation length becomes larger, for example, close to a critical phase transition, becoming maximum when the fluctuators are correlated throughout the entire sample size. In reduced dimensions, non-Gaussian fluctuations have been predicted for magnetization of a 2D XY model which shows BKT transition as a function of temperature [21]. However, there have been very few experiments probing the statistics of fluctuations in systems undergoing BKT transition [22]. In this Letter, we study the statistics of resistance fluctuations to probe the nature of transition in thin film superconductors both in 3D and quasi-2D limits.

A characteristic feature of superconducting thin films is that when the film thickness (d) is reduced below the coherence length (ξ_l), the superconductivity crosses over to the 2D limit characterized by BKT transition [23,24].

The material system chosen for our study is a conventional
s-wave superconductor niobium nitride (NbN). The
Ginzburg-Landau coherence length ($\xi_l \sim 6$ nm) in these
films was estimated from the measurement of the upper
critical field (H_{c2}) as a function of temperature [25] and the
details are included in the Supplemental Material [26].
NbN thin films exhibit a crossover from BCS to BKT
type behavior, with reduction in d and are very good
candidates to study the BKT transition since ultrathin
films (down to 3 nm) can be synthesized with a high
degree of structural uniformity, while displaying moder-
ately large T_c. Thus, they provide us with an ideal platform
to investigate vortex correlations in low-dimensional
superconductivity.

The $T - d$ phase diagram of NbN films, showing the
critical temperature (T_{BKT} or T_{BCS}) vs normalized film
thickness is shown in Fig. 1(a). Films with $d \sim \xi_l$, undergo
BKT transition (black line and symbol) while the expected
mean field temperature is shown by the red line and sym-
bol. The thicker or 3D films ($d > \xi_l$) display BCS type
behavior and, hence, are characterized by only one tem-
perature scale (T_{BCS}) whereas films in the quasi-2D limit
($d \lesssim \xi_l$), which display BKT transition, are characterized
by two temperature scales (T_{BKT} and T_{BCS}). The mean field
critical temperature, T_{BCS}, decreases as the film thickness
is reduced due to the increase in electron-electron
Coulomb interactions which compete with the attractive

interaction required for superconductivity [27]. The
shaded region corresponds to the BKT fluctuation regime
dominated by interacting vortices.

The samples used in our experiments were grown on
lattice matched MgO substrate using dc magnetron sputter-
ing (for details, refer to [23,28,29]). Contacts (Cr/Au)
were defined by wire masking for four-probe measure-
ments. The normalized temperature dependence of resist-
ance is shown for all films in Fig. 1(b). The width of the
transition from the normal state to the superconducting
state is ≈ 1.1 K for thicker films and increases to ≈ 1.8 K
when the thickness is reduced below 6 nm. It is important
to note that T_c depends very strongly on the microstructural
details, and in spite of very similar deposition conditions,
variations in the microstructural details, particularly grain
size, can make T_c vary by nearly 30%. This also indicates
a limitation of standard time-averaged transport to charac-
terize the nature of ultrathin superconductivity without
ambiguity.

Our experimental procedure consists of measuring slow
time-dependent fluctuations in the sample resistance at
different temperatures for zero magnetic field. Typical
time traces of the fluctuations for a 3 nm thick NbN film
are shown in Fig. 1(c) and 2(a). The details of the noise
measurement technique are available in Ref. [19] as well as
the Supplemental Material [26]. As the temperature is
reduced, the fluctuations increase in magnitude [Fig. 2(a)].
The normalized power spectral density (PSD) for different
temperatures as shown in Fig. 2(b), exhibits $1/f^\alpha$ type
behavior. The PSD increases in magnitude as the tempera-
ture is lowered. We find that $\alpha \sim 2$–4 at low temperatures,
which eventually converges to ~ 1 at higher temperatures
restoring the $1/f$ nature of noise. We find similar behavior
in α even for thicker samples. The rather high values of α
might possibly arise due to the presence of vortices or
inhomogeneous percolative transport [30]. We believe
that further experiments are required to understand this
behavior of α. To explore the temperature dependence of
noise close to the superconducting transition, we integrate
the PSD over our measurement bandwidth (≈ 15 Hz) to
obtain the normalized variance which is shown along with
resistance as a function of temperature in Fig. 2(c). We find
that the normalized variance shows a divergent behavior at
low temperatures for all samples irrespective of thickness.
This behavior is best understood from the theory of perco-
lation. It is well known that for $T > T_{\mathrm{BKT}}$ ($T > T_{\mathrm{BCS}}$ for
$d \geq 18$ nm) thin film superconductors exhibit fluctuations
in the superconducting order parameter which leads to
formation of Cooper pairs for a very short time. These
fluctuations in the superconducting order parameter effec-
tively lead to the formation of a network of interconnected
superconducting and nonsuperconducting regions. As the
temperature is reduced, the superconducting phase grows.
The noise in these systems arises from two possible sce-
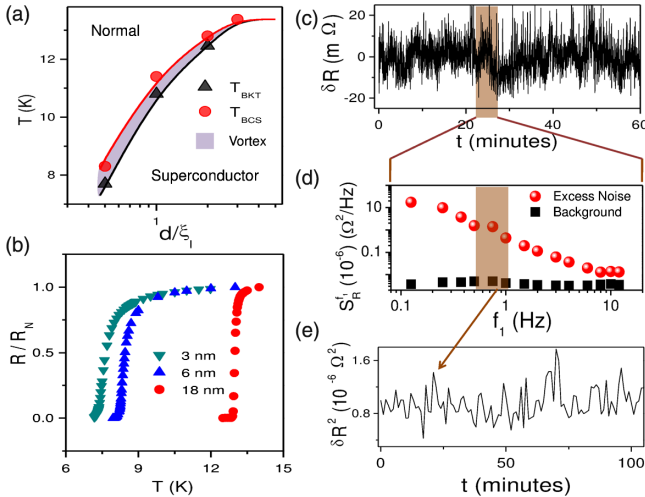narios: (a) resistance fluctuations in nonsuperconducting

FIG. 1 (color online). (a) Phase diagram of thin films of NbN
as a function of normalized film thickness, where ξ_l is the
superconducting coherence length (extracted from Table 1 in
Ref. [23]). Shaded region denotes presence of vortices due to
BKT transition in films in quasi-2D limit. The red and black
lines are a guide to the eye representing the variation in T_{BCS} and
T_{BKT}, respectively. (b) Normalized resistance for films of differ-
ent thickness as a function of temperature where R_N represents
the normal state resistance. (c) Time series of resistance fluctua-
tions at 7.3 K for $d = 3$ nm. (d) Power spectral density for a
given time window. (e) Fluctuations in noise power in an octave
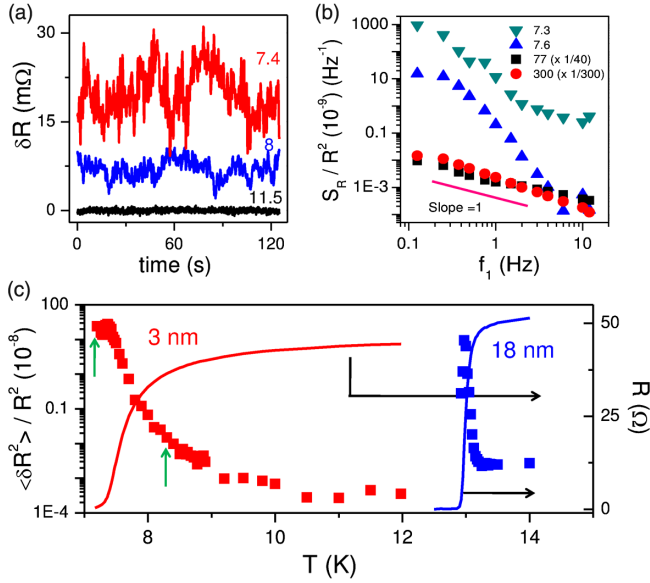spanning 0.5–1 Hz across different time windows.

FIG. 2 (color online). (a) Time traces showing the resistance fluctuations at different temperatures for $d = 3$ nm. The traces are displaced by 10 mΩ for clarity. The mean value of resistance at 7.4, 8, and 11.5 K are 5.3, 33, and 44.2 Ω, respectively (b) PSD at different temperatures for $d = 3$ nm. Scaling factors are indicated in the legend for $T = 77$ and 300 K. (c) Normalized variance of noise ($\langle \delta R^2 \rangle / R^2$) (filled squares) and resistance (R) (line) at different temperatures for $d = 3$ nm (red) and $d = 18$ nm (blue). The green arrows (pointing upwards) indicate the temperatures at which the second spectra are shown in Fig. 3(a).

regions, and (b) fluctuations in the number of superconducting regions or islands. It is well known from the theory of percolation that noise grows until the film becomes fully superconducting [31–34]. The noise follows the relation $\langle \delta R^2 \rangle / R^2 \propto R^{-l_{rs}}$ where the percolation exponent $l_{rs} \sim 1 \pm 0.4$ in our experiments (refer to the Supplemental Material [26]). We also do not rule out the contribution of generic critical fluctuations which are always present in thin film superconductors. The influence of temperature fluctuations ($\ll 0.5$ mK) was carefully evaluated and eliminated by varying the control parameters and observing its effect on the nature of the noise power spectrum.

To probe the presence of correlations, we measured higher-order statistics of noise using two independent methods: (a) calculating the probability distribution function (Supplemental Material [26]), and (b) second spectrum of the fluctuations. The second spectrum is a sensitive technique to estimate the presence of a non-Gaussian component (NGC) in the resistance fluctuations [18,35,36]. The experimental scheme employed to measure the second spectrum is shown in Figs. 1(c)–1(e). The time series shown in Fig. 1(c) is divided into successive windows of 2 minutes each. The power spectral density is calculated for each such window [Fig. 1(d)] and it is integrated over a chosen octave to obtain the noise power which is plotted as a function of time in Fig. 1(e). In effect,

the second spectrum is the Fourier transform of the four-point correlation of $\delta R(t)$ and is given by

$$S_R^{f_1}(f_2) = \int_0^\infty \langle \delta R^2(t) \delta R^2(t + \tau) \rangle \cos(2\pi f_2 \tau) \, d\tau,$$

where f_1 is the center frequency of the chosen octave and f_2 corresponds to the spectral frequencies. $S_R^{f_1}(f_2)$ represents the "spectral wandering" or fluctuations in the noise power in a chosen frequency band (f_L, f_H) [Fig. 1(e)]. Specifically, we have chosen an octave spanning 0.5–1 Hz, where the excess noise is considerably higher than the background, to minimize the effects of signal corruption by Gaussian background. The second spectrum of noise appears to display a rather weak dependence of frequency but shows an increase in overall magnitude with the lowering of temperature [Fig. 3(a)]. A convenient way to represent the second spectrum is to plot the normalized variance of the second spectrum $\sigma^{(2)} = \int_0^{f_H - f_L} S_R^{f_1}(f_2) df_2 / [\int_{f_L}^{f_H} S_R(f) df]^2$, which is a measure of the normalized fourth-order moment in noise, as a function of temperature. As shown in Fig. 3(b), $\sigma^{(2)}$ decreases monotonically from ≈ 9 at 7.2 K to a baseline value of ≈ 4 near 7.5 K. Comparison with the temperature dependence of resistance reveals that the enhanced $\sigma^{(2)}$ marks the onset of the normal state, which decays to a temperature-independent baseline value within about 10% of the normal state resistance (R_N). Importantly, $\sigma^{(2)}$ is reduced to the baseline value by $T \approx 7.6$ K, whereas the percolation dynamics of the resistance fluctuations is observable well up to ≈ 10 K [Fig. 2(c)]. This implies that the increase in $\sigma^{(2)}$ is due to an independent process, and unlikely to be due to critical or percolative fluctuations that dominate the measured noise (first spectrum).

To correlate the measured T dependence of $\sigma^{(2)}$ with other physical properties of the NbN thin films at the superconductor-normal transition, we have measured the magnetic penetration depth (λ) in the same sample as a function of T [37]. In such measurements, λ^{-2} is proportional to the superfluid density, whose deviation from the mean field BCS expectation [black line in Fig. 3(b)] has recently been interpreted as the BKT transition in ultrathin NbN films [23]. In the present sample [Fig. 3(b)], the maximum in $\sigma^{(2)}$ occurs at the same temperature, suggested as the BKT transition temperature T_{BKT}, where the superfluid density (λ^{-2}) drops to zero. Strikingly, $\sigma^{(2)}$ approaches the baseline value at the theoretically computed mean-field superconducting transition temperature [23] (T_{BCS}), suggesting a correlated kinetics of the charge carrying species between T_{BKT} and T_{BCS} [the shaded region in Fig. 3(b)].

To get a further understanding of the underlying mechanism, we have repeated the measurements for NbN films with different thickness. We subtract the baseline value from $\sigma^{(2)}$ in each case and plot the excess value as a function of normalized resistance R/R_N for each sample
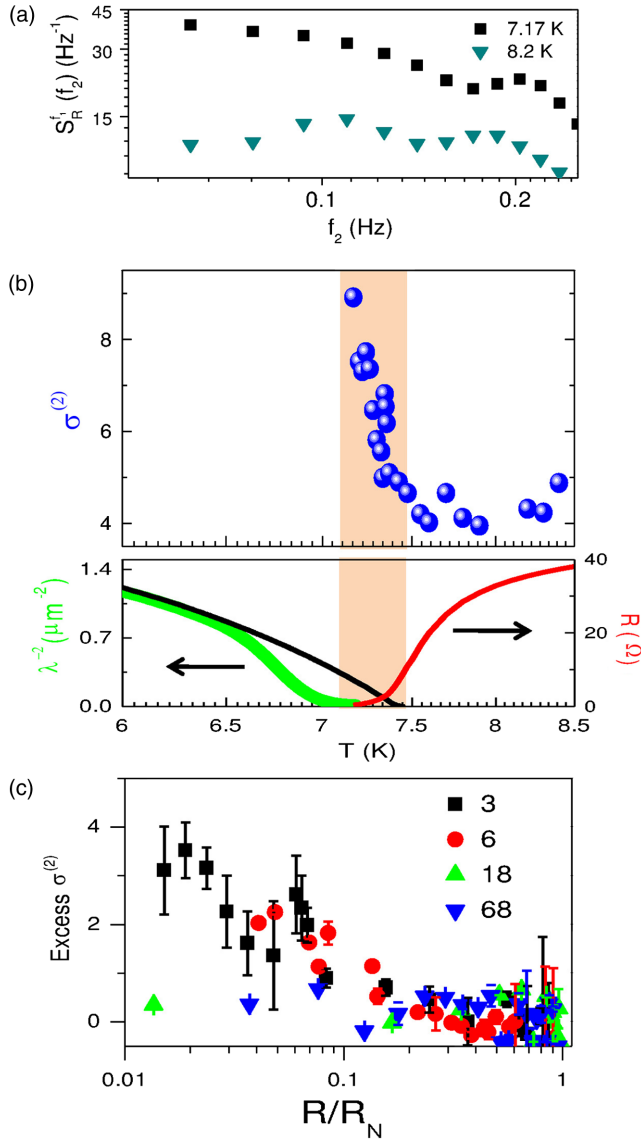
FIG. 3 (color online). (a) Second spectrum at different temperatures indicated by green arrows in Fig. 2(c) in an octave spanning (0.5–1) Hz. (b) Temperature dependence of normalized variance of the second spectrum $\sigma^{(2)}$ (blue points, upper panel), superfluid density λ^{-2} (green line, lower panel), and sample resistance R (red line, lower panel). Expected BCS variation of λ^{-2} (black line) obtained from a fit to the data in the temperature range 0.5 K–6.2 K is shown in the lower panel. (c) Excess $\sigma^{(2)}$ as a function of normalized resistance (R/R_N) for samples of different thickness. The error bars were calculated from measurements of $\sigma^{(2)}$ over 60 time windows.

[Fig. 3(c)]. We find that the representation in terms of R/R_N allows better comparison as the samples display a wide range of T_C. The dependence of $\sigma^{(2)}$ on R/R_N for thinner films ($d \sim \xi_l$) was found to be very different from the thicker ones ($d \geq 18$ nm). Most ($>80\%$) quasi-2D samples ($d \leq 6$ nm) show the remarkable increase in $\sigma^{(2)}$ close to the superconducting transition signifying the presence of long-range correlations, whereas the bulk films

display essentially no excess $\sigma^{(2)}$ which is indicative of Gaussian type fluctuations as expected in the case of Ginzburg-Landau fluctuations for a BCS superconductor. The thickness dependent behavior of $\sigma^{(2)}$ has two key implication: First, since all samples, irrespective of thickness, show critical fluctuations or percolative kinetics near the superconducting transition, we can eliminate the enhancement of $\sigma^{(2)}$ to arise from such kinetics. Second, appearance of excess $\sigma^{(2)}$ in the regime dominated by vortex fluctuation [Fig. 3(b) and schematics of Fig. 1(a)] strongly suggests the excess $\sigma^{(2)}$ to arise from long-range correlations among the vortices themselves.

Another phenomenon which can give rise to NGC in noise is dynamical current redistribution (DCR) which occurs due to large local resistivity fluctuations and strong transport inhomogeneities [38]. In our samples, $\delta R/R \ll 1$ and the noise mechanism is the same for both thin and thicker films. Had the origin of NGC been due to DCR, then all the films must have displayed strong NGC in noise which is not the case in our measurements.

Finally, to explore the sensitivity of non-Gaussian fluctuations to the structural details of the films, we have carried out measurements on two different samples of same nominal thickness 6 nm, but different structural morphology. We used an Atomic Force Microscope to characterize the roughness of the samples that were prepared at different times and deposition conditions. One of the samples (6 nm S3) shows an average roughness of 1.65 nm, whereas the relatively smoother sample (6 nm S4) has a much lower average roughness (0.31 nm) [Figs. 4(a) and 4(b)].
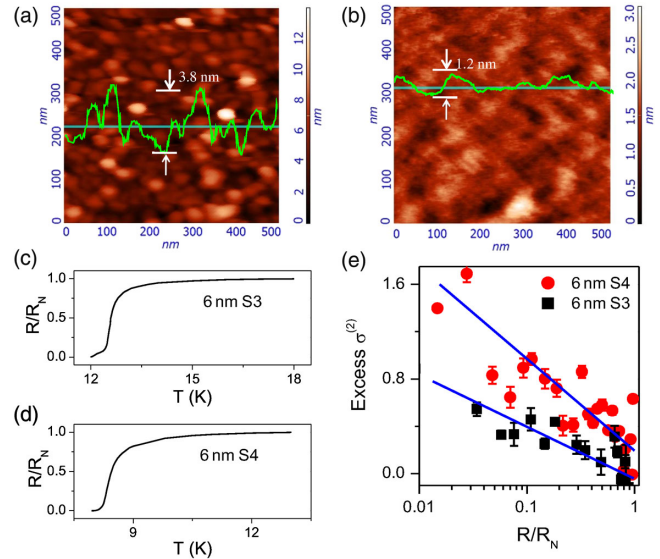

FIG. 4 (color online). (a),(b) Atomic Force Microscope images of samples ($d = 6$ nm) 6 nm S3, and 6 nm S4, respectively, with the insets showing a line scan (green line) indicating the roughness of respective samples. (c),(d) RT characteristics of both the samples. (e) Excess $\sigma^{(2)}$ for both the samples. Solid blue lines are a guide to the eye.

We observe a kink like feature (around 12.2 K) in the resistance vs temperature data [Fig. 4(c)] for 6 nm S3, possibly as a result of the surface roughness and strong thickness variation in the film. A clear distinction is observed in the second spectrum of noise [Fig. 4(e)] with the NGC becoming weaker as the surface roughness increases. These results can be viewed in the context of vortices in disordered or granular superconductors. Granular thin films are modeled as disordered arrays of Josephson junctions which can lead to a double transition [39,40]. Moreover, the pinning of vortices due to random potential fluctuations or defects is known to limit the long-range interactions among the vortices [41,42]. Further experiments on samples with controlled levels of disorder will be of great interest in evaluating the robustness of NGC in noise.

Thus, our experiments underline a new method to identify the BKT fluctuation regime in ultrathin superconducting films. The characteristics temperature scales T_{BKT} and T_{BCS} emerge as the range over which the NGC is nonzero, that could be useful to probe other low-dimensional superconductors as well.

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