Magnetoresistance studies of homogenously disordered 3-dimensional NbN thin films

Madhavi Chand1*, Mintu Mondal1, Sanjeev Kumar1, Anand Kamlapure1, Garima Saraswat1, S. P. Chockalingam12, John Jesudason1, Vivas Bagwe1, Vikram Tripathi1, Lara Benfatto3 and Pratap Raychaudhuri1

1 Tata Institute of Fundamental Research, Homi Bhabha Road, Navy Nagar, Colaba, Mumbai-400005, India
2 Columbia University, 538 West 120th Street, New York, NY 10027, USA
3 INFM-CNR Statistical Mechanics and Complexity Center, University of Rome “La Sapienza,” P.le A. Moro 5, 00185 Rome, Italy

*email: chand@tifr.res.in

Abstract. We present magnetoresistance studies on a series of homogenously disordered 3-dimensional NbN thin films with disorder ranging from the moderately clean limit \(k_F l \sim 10.12\) to the extremely dirty limit \(k_F l \sim 0.42\). We find that for samples with \(k_F l > 1\), the magnetoresistance is positive up to 12T and as disorder increases it decays more slowly with temperature. On the other hand, for samples with \(k_F l < 1\), we observe a peak in the magnetoresistance which vanishes at a temperature close to the pseudogap temperature of disordered superconducting samples. These observations are consistent with the idea of the disorder-driven non-superconducting state being comprised of pre-formed Cooper pairs.

1. Introduction

The nature and properties of the disorder-driven superconductor-insulator transition has been a subject of active research for many years. Detailed experimental investigations have been reported on a number of systems like quench condensed films of Bi [1,2], homogenously disordered TiN [3] and NbN [4] and amorphous InO [5,6], to name a few. In these experiments, many interesting phenomena have been observed such as the existence of a pseudogap state at high disorder [4], a large peak in the magnetoresistance [1,5], and the evidence of pairing in films with a non-superconducting ground state in the form of magnetoresistance (MR) oscillations [2]. Numerical studies also predict phenomena like the spontaneous formation of superconducting islands [7,8] and consequently a pseudogap state [7] and are therefore in agreement with many of the experiments. One of the main open questions is the nature of charge carriers at the SIT and in the regime where the system is driven into a non-superconducting ground state. The idea of the insulating state being comprised of Cooper pairs has received a lot of support in recent times [9,10,6] from tunneling and MR studies. Different explanations have been suggested recently for the peaks observed in the MR based on ideas of percolation [11] and localization [12]. In this paper we present MR measurements on a series of 3-dimensional, homogenously disordered NbN thin films. Samples with \(k_F l > 1\) have a positive MR up to 12T and as disorder increases it decreases at a slower rate with temperature. On the other hand, for samples with \(k_F l < 1\), there is a peak in the MR which vanishes at a temperature close to the pseudogap temperature.
2. Experimental Details

NbN thin films were prepared by reactive DC magnetron sputtering of a Nb target in a gas mixture of Ar and N₂ onto single crystal MgO (100) substrates. Stoichiometric NbN is a conventional BCS superconductor with a relatively high superconducting transition temperature \( T_c \) of 17K. However, the \( T_c \) is extremely sensitive to the growth conditions, which enables us to tune the disorder over a wide range. Details of fabrication and characterization can be found in References [13] and [14]. Disorder is characterized by the Ioffe Regel parameter \( k_F l \) \((k_F \equiv \text{Fermi wave vector and } l \equiv \text{mean free path})\), obtained experimentally from Hall effect and resistivity \( \rho \) measurements at 285K using free electron theory [13]. MR was measured in standard four-probe geometry, on a sample patterned using a shadow mask, in a conventional He3/He4 cryostat with a lowest temperature of 300mK and a highest field of 12T. Two kinds of measurements were made on the samples, namely resistance \( (R) \) vs. temperature \( (T) \) at different fields \( (H) \) and \( R \) vs. \( H \) at different temperatures.

3. Results and Discussion

Figure 1: (a) Resistivity vs. Temperature for samples with \( k_F l \approx 10.12, \ 8.82, \ 8.13, \ 8.01, \ 5.5, \ 4.98, \ 3.65, \ 3.27, \ 1.95, \ 1.68, \ 1.58, \ 1.23, \ 0.82, \ 0.49 \) and \( 0.42 \), on a logarithmic scale and. Inset: \( \sigma vs. T \) for the most disordered samples. (b) Phenomenological phase diagram showing superconducting critical temperature, \( T_c \) and pseudogap temperature \( (T^*) \) vs \( k_F l \). The labels I, II and III demarcate the regions conforming to mean field theory, with a pseudogap state and no superconducting ground state respectively.

Figure 1(a) shows \( \rho vs. T \) curves on a logarithmic scale for the full range of samples. We can see that barring the cleanest sample \((k_F l \approx 10.12)\) the rest have a negative temperature coefficient of resistance that gets more and more pronounced as disorder is increased. Simultaneously, the \( T_c \) decreases continuously till \( k_F l \approx 1 \) where it vanishes completely. This has been shown in Figure 1(b) in the form of a phenomenological phase diagram. We have shown previously through scanning tunnelling spectroscopy [4] that in the strongly disordered regime \((k_F l < 4)\), a gap is observed in the tunnelling density of states at temperatures higher than \( T_c \).

This gap, known as the pseudogap, finally vanishes at a higher temperature \( T^* \). This suggests that the superconducting transition is governed by phase fluctuations and Cooper pairs continue to exist at temperatures above which the phase coherence across the sample is lost.

In Figure 2 we present the MR data of samples with a well defined superconducting transition temperature. Figure 2(a) shows \( R \) as a function of applied magnetic field at different temperatures \( (T) \) above \( T_c \) as well as \( R \) vs. \( T \) for different applied fields. Figures 2(a) and 2(b) show the raw data for samples with \( T_c \sim 13.6K \) and 3.2K respectively.

In Figure 3 we show the MR, defined as the relative change in resistance with magnetic field \( R(12T) - R(0)/R(12T) \) as a function of \( T/T_c \) for a series of films with \( T_c \sim 16.2, \ 13.57, \ 11.5, \ 8.13, \ 7K, \ 4.5K, \ 3.2K \) and \( 1.65K \). This plot clearly indicates that as the system gets more disordered, the MR decreases less rapidly and persists up to higher temperatures above \( T_c \).

These MR observations are qualitatively consistent with the phase fluctuations and pseudogap scenario described in Ref [4]. In the pseudogap state the system can be viewed as consisting of
superconducting domains. Although the superconducting domains cannot produce a zero resistance state above $T_c$, because of phase incoherence between them, the superconducting correlations persist and lead to a lower resistance in this regime $T_c < T < T^*$. On the application of a magnetic field, these intra-domain superconducting correlations are also suppressed and the resistance increases to what it would have been in the absence of pairing correlations.

We now look at the samples that do not show a superconducting transition. Figures 4(a), (b) and (c) show $\rho$ vs. $T$ for samples with $k_B T < 0.2$, 0.49 and 0.42. Here we observe a prominent peak in the MR. We label the position of the MR peak in the data measured at the lowest temperature (300mK) as $H_{\text{peak}}$. It is evident from Figures 4(a), (b) and (c) that $H_{\text{peak}}$ shifts to lower fields with increasing disorder. In Figure 4(d) we plot these values of $H_{\text{peak}}$ along with the experimentally measured upper critical field ($H^\ast$) for the superconducting ones. We see that $H_{\text{peak}}$ evolves smoothly from $H^\ast$ across the STI. Details of measurement of $H^\ast$ can be found in references [13] and [15]. Figures 3(e-f) show the temperature evolution of the MR peaks. The peaks vanish at ~5K, which is close to $T^*$ in the disordered films.

We now try to understand the origin of the MR peak. We have seen in the phenomenological phase diagram (Figure 1(b)) that $T^*$ saturates at high disorder and remains ~6K even very close to $k_B T < 1$. This seems to suggest that Cooper pairing continues to exist even when $k_B T < 1$. Different theoretical frameworks have provided different explanations for this MR peak seen in many disordered superconductors. A recent paper suggests that the non-superconducting state comprises of localized Cooper pairs [12]. Such a scenario can also exist due to the fractal nature of the wavefunction in the insulating state [9]. However, we do not believe that this interpretation is applicable in the case of our films as even the non-superconducting films are not deep in the insulating regime. We have found that on plotting the conductivity ($\sigma$) vs. $T$ and extrapolating to zero temperature (inset of Figure 1(a)), $\sigma(0) \neq 0$ which indicates that these samples are dirty metals and not insulators. Therefore the interpretation more suitable here is the one due to Dubi et. al. [11] wherein the systems can be thought of as comprising of superconducting puddles separated by metallic regions of which shrink in size with increasing magnetic field due to suppression of superconducting correlations leading to a positive MR. Beyond a certain field, $H_{\text{peak}}$ these puddles shrink so much that it is no longer favorable for the current to pass through them resulting in the fact that the current now passes through only the normal regions, which

Figure 2: (a) Resistance ($R$) vs. applied magnetic field ($H$) for a sample with $T_c \sim 13.6K$, measured at different temperatures ($T$): 13.85K, 14K, 14.3K, 14.7K, 15K, 15.5K, 16K, 17K, 18K, 20K, 24K and 30K. The inset shows $R$ vs. $T$ for the same sample measured at different $H$ (b) $R$ vs. $H$ for sample with $T_c \sim 3.2K$ measured at 3.5K, 3.7K, 3.9K, 4.1K, 4.3K, 4.6K, 5K, 5.5K and 6K, again, inset shows $R$ vs. $T$ for the same sample measured at different $H$.

Figure 3: ($R(12T)-R(0T))/R(12T)$ as a function of $T/T_c$. For a series of films with $T_c$ 16.4K, 13.6K, 11.5K, 8.13K, 7K, 4.5K, 3.2K and 1.65K. The points are obtained from the MR scans and the solid lines are from the $R-T$ scans at $0T$ and $12T$. 
increase with further increase in $H$, resulting in negative MR. We therefore interpret $H_{\text{peak}}$ as the field beyond which superconducting correlations become insignificant, which is consistent with the fact that $H_{\text{peak}}$ evolves from $H_{c2}$. In addition, the fact that the peak vanishes at a temperature close to $T^*$ further corroborates the existence of Cooper pairs in the non superconducting state [16].

Conclusions
We have measured magnetoresistance for a series of NbN samples over a wide range of disorder and found it to be consistent with the idea of persistence of Cooper pairing above $T_c$ in strongly disordered superconductors where the superconducting state is destroyed by phase fluctuations and also in the regime where the ground state is no longer superconducting.

References

[1] H. Q. Nguyen et. al., Phys. Rev. Lett. 103, 157001 (2009).
[2] M. D. Stewart Jr. et. al., Science 318, 1273 (2007).
[3] B. Sacepe et. al., Nat Commun. 1, 140 (2010).
[4] M. Mondal et. al., Phys. Rev. Lett. 106, 047001 (2011).
[5] G. Sambandamurthy, et. al. Phys. Rev. Lett. 94, 017003 (2005).
[6] B. Sacépé et. al., Nature Physics 7, 239–244 (2011).
[7] A. Ghosal, M. Randeria, and N. Trivedi, Phys. Rev. Lett. 81 3940 (1998); Phys. Rev. B 65, 014501 (2001).
[8] Y. Dubi, Y. Meir, and Y. Avishai, Nature London 449 876 (2007); Phys. Rev. B 78, 024502 (2008).
[9] M. V. Feigel’man, L. B. Ioffe, V. E. Kravtsov, and E. A.Yuzbashyan, Phys. Rev. Lett. 98 027001 (2007).
[10] K. Bouadim, Y. L. Loh, M. Randeria and N. Trivedi, Nature Physics doi:10.1038/nphys2037.
[11] Y. Dubi, Meir, and Y. Avishai, Phys Rev. B 73, 054509 (2006).
[12] M. Müller, arXiv: 1109.0245v1
[13] S. P. Chockalingam et. al., Phys. Rev. B 77 214503 (2008).
[14] M. Chand et. al., Phys. Rev. B 80, 134514 (2009).
[15] M. Mondal et. al., J. Supercond. Nov. Magn. 24, 341 (2011).
[16] A more detailed discussion can be found in M. Chand et. al. arXiv: 1107.0705.