Effect of Phase Fluctuations on the Superconducting Properties of Strongly Disordered 3D NbN Thin Films

Madhavi Chand1*, Mintu Mondal1, Anand Kamlapure1, Garima Saraswat1, Archana Mishra2, John Jesudasan1, Vivas C. Bagwe1, Sanjeev Kumar1, Vikram Tripathi2, Lara Benfatto4, and Pratap Raychaudhuri1

1Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400005, India
2IIC, Indian Institute of Technology Roorkee, Roorkee, Uttarakhand 247667, India
3Department of Condensed Theoretical Physics, Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400005, India
4INFM-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 transport, Hall effect, electronic tunnelling and penetration depth studies in 3D homogeneously disordered epitaxial NbN thin films with disorder ranging from the moderately clean limit ($k_Fl\sim10.12$) to the very dirty limit ($k_Fl\sim1.24$). The superconducting transition temperature ($T_c$) decreases from ~17K to less than 350mK with increasing disorder. The $T_c$ and conductivity at the lowest temperature ($\sigma_0$) both asymptotically approach zero as $k_Fl\rightarrow1$, indicating a coincidence of the metal-insulator transition (MIT) and the superconductor-insulator transition (SIT). Close to critical disorder there is spatial inhomogeneity in the superconducting density of states (DOS) and the superconducting state is governed by quantum phase fluctuations. This results in suppression of the superfluid density ($n_s$) and a pseudogap state where the resistance is no longer zero but the energy gap ($\Delta$) remains finite.

Introduction

The disorder driven superconductor-insulator and metal-insulator transitions have been studied in a number of systems recently and the physics that governs the behaviour of superconducting and electronic properties close to these transitions is a topic of fundamental interest today.

It has been observed in studies on doped materials like Nb$_x$Si$_{1-x}$[1] and Au$_x$Si$_{1-x}$[2] that superconductivity vanishes on the metallic side of the MIT. Theories suggest [3,4] that in systems with homogenous disorder, superconductivity is suppressed primarily by increased effectiveness of Coulomb repulsion due to loss of screening and is therefore likely to vanish in the metallic regime.

On the other hand, in granular systems, superconductivity is found to persist on the insulating side of the MIT. Examples of materials studied are granular Al [5] and InO [6]. However, most of the experiments have been done on 2D systems where thickness is used to control the sheet resistance. This therefore poses an open question for 3D systems regarding whether or not superconductivity would persist on the insulating side of the MIT. It has been predicted [7] for strong localization that in the absence of el-el repulsion, superconductivity is possible below the mobility edge as long as
\[ \rho \Delta \ell \geq 1. \] Here \( \rho \) is the DOS at Fermi level, \( \Delta \) is the superconducting energy gap, \( \ell \) is the localization length and \( D \) is the dimension. This is tantamount to saying that superconductivity will survive as long as \( \Delta \) is larger than the level spacing for particles confined within \( \ell \).

Stoichiometric NbN is a conventional BCS superconductor with a relatively high \( T_c \) of 17K. However, disorder can be introduced without external doping so the properties are not dependent on composition. At the same time, the films are 3D and disorder does not depend on thickness. Disorder is characterized by the Ioffe Regel parameter \( k_F l \) (\( k_F \equiv \) Fermi wave vector and \( l \equiv \) mean free path), obtained experimentally from Hall effect and resistivity measurements at 285K using free electron theory [8,9]. The existence of strong interactions has been established in a temperature dependent study of Hall effect wherein it is clear that localization theories alone or even weak interaction theories are inadequate in explaining the normal state behaviour of this system[9]. In this paper we explore the superconductor-insulator transition in the context of the metal-insulator transition and study the superconducting properties in the region of critical disorder.

**Experimental Details**

NbN films were grown epitaxially on (100) oriented single crystalline MgO substrates using reactive D.C. magnetron sputtering of a Nb target in an Ar/N\(_2\) gas mixture. Films with \( k_F l \) ranging from 1.24 to 10.12 were deposited by controlling the sputtering power and Ar:N\(_2\) ratio.

The temperature dependence of resistivity (\( \rho \)) was measured using a standard A.C. four probe technique. To obtain \( k_F l \) experimentally, the Hall coefficient was measured on samples patterned in a Hall bar geometry by sweeping a magnetic field from -12T to 12T and then folding over the data to cancel the resistive component. Tunneling measurements were performed on NbN/insulator (oxide)/Ag planar tunnel junctions similar to those studied in [10]. The scanning tunnelling spectroscopy (STS) is done in a home-made low temperature STM. Details of the setup can be found in Ref [11]. The penetration depth (\( \lambda \)) as a function of temperature was measured down to 350mK using a “two coil” mutual inductance technique operating at 60 kHz. The advantage of this technique is that it allows measurement of the absolute value of \( \lambda \) over the entire temperature range up to \( T_c \) without any prior assumption about the dependence of \( \lambda \) on T. An explanation of this technique can be found in Ref [11].

**Results and Analysis**

Figure 1 (a) shows the temperature dependence of conductivity for films with different \( k_F l \). We can see that the positive temperature coefficient of conductivity gets stronger as we increase the disorder. A detailed analysis of this temperature dependence along with the corresponding temperature dependence of Hall coefficient can be found in Ref [9].

In the case of the most disordered sample (\( k_F l \sim 1.24 \)), where the zero resistance regime could not be reached, we verified that the upturn in conductivity is due to superconductivity by measuring the \( \sigma-T \) in presence of a magnetic field which shows the superconductivity getting quenched at 5T (inset of Figure 1(b)). Since neither the sign of the temperature coefficient of resistance nor the absolute value of \( k_F l \) can be used to exactly identify the metal insulator transition, the determining quantity used is the zero temperature conductivity. Therefore, we plot the lowest temperature conductivity (\( \sigma_0 \), measured at the peak value just above \( T_c \)) as a function of \( k_F l \) (Figure 1(b)). We can see that within experimental error, as \( k_F l \rightarrow 1 \), \( \sigma_0 \rightarrow 0 \) confirming that \( k_F l \rightarrow 1 \) does indeed correspond to the metal-insulator transition.

On the same plot we look at \( T_c \) as a function of \( k_F l \) and observe that \( T_c \) is suppressed as \( k_F l \) decreases and tends to zero as \( k_F l \rightarrow 1 \) indicating the coincidence of the metal-insulator and superconductor-insulator transitions at a single quantum critical point.
Now, with the dual quantum critical point established, it is interesting to explore the properties of the superconducting state close to the critical region.

Figure 2(a) shows a histogram of the variation in $\Delta$ (defined as half the difference between the coherence peak positions) measured via STS at different points on a highly disordered sample ($T_c$=4.1K) over a 150X30nm region. The observed spatial inhomogeneity demonstrates as a spatial distribution in the values of $\Delta$. Such a spontaneous segregation has been predicted in numerical studies [12] as well. The existence of this inhomogeneity brings into question the degree of phase stiffness in the superconducting state. In figure 2(b), the evolution of the DOS with temperature obtained by taking a spatial average of the local DOS shows that the gap feature persists even above the temperature at which resistance goes to zero. This can be understood in the following way: In most mean field theories, the electron-pairing and long range phase coherence occur at the same temperature, $T_c$. This means that the global phase coherence and $\Delta$ vanish at the same temperature, due mainly to the suppression of the gap with temperature. However, it has been shown [13] that for systems with low conductivity and small $n_s$, the phase ordering temperature is reduced significantly and becomes comparable to the pairing temperature. Therefore, the ‘pseudogap’ in figure 2(b) indicates that in this system, the superconducting state is possibly destroyed by phase fluctuations.

The phase fluctuations scenario is corroborated by measurement of $n_s$ (shown in terms of $1/\lambda^2(0)$ in figure 2(c)) at 350mK for a series of samples with different disorder which is compared with the value predicted from BCS: $1/\lambda^2_{BCS}(0) = \pi \mu_0 \Delta(0) \sigma_0 / \hbar$ where $\mu_0$ is the permeability of free space.

We observe that for samples with $T_c\geq$6K, the experimental and theoretical trends match closely, but for samples with $T_c<6$K there is strong suppression in the measured $n_s$ compared to the value predicted.

**Figure 1:** (a) $\sigma$ vs. $T$ for films of $k_Fl \sim 10.12, 8.82, 8.13, 8.01, 5.5, 4.98, 3.65, 3.27, 2.21, 1.68, 1.58$ and $1.24$. (b) Minimum $\sigma$ vs. $k_Fl$ as well as $T_c$ vs $k_Fl$. Inset of (b): zero field and in-field $\sigma$ vs. $T$ for the most disordered film with $k_Fl$=1.24.

**Figure 2:** (a) Histogram of observed energy gap obtained from measurement of local density of states. The value is taken as the average of the positions of the coherence peaks on either side of zero bias; (b) (top) averaged conductance curves measured at 3.3K and 4.5K for a sample of $T_c=4.1$K and (bottom) resistance vs. temperature for the same sample; (c) experimental and theoretical values of $1/\lambda^2(0)$ for different samples, (inset: superconducting energy gap ($\Delta$) vs. $T_c$ for different samples (magenta squares) along with a linear fit extrapolated to zero (black line)); (d) Conductance curves measured at 500mK for samples of different $T_c$. The black lines correspond to an attempted BCS fit to the density of states.
from BCS. In this calculation, $\Delta(0)$ is taken to be $2.05 k_B T_c$, corresponding to a strong coupling superconductor, which is obtained from a linear fitting of $\Delta$ vs. $T_c$ for samples with less disorder i.e. $T_c > 8$K (inset of figure 2(c)).

In a strongly interacting system like ours, we expect that number fluctuations, arising from the number phase uncertainty relation when there is long range phase coherence, would have a large Coulomb energy cost making it energetically favourable to have large quantum phase fluctuations in the system. However, in spite of large suppression of $n_s$ there will still be a phase coherent ground state as long as the conductivity at $T_c$ is greater than the quantum conductivity [13] i.e. $\sigma / \sigma_Q > 1$ where $\sigma_Q = (1/3)(2e)^2 / h a$. Here we take the characteristic length of phase fluctuations ‘a’ to be $\sim \xi$, which is the measured coherence length. We have found that this condition is true even for our most disordered samples with $T_c \sim 2$K where the ratio is $\sim 3.7$.

In addition to the spontaneous inhomogeneity, pseudogap feature and reduced $n_s$, we observe that the coherence peaks in the superconducting DOS are suppressed. In figure 2(d) we plot conductance curves for samples of different disorder measured at $\sim 500$ mK using planar tunnel junctions. We find that the coherence peaks are strongly suppressed compared to what is expected from the BCS DOS: $N(E) = E / \sqrt{E^2 - \Delta^2}$.

**Conclusions**

We have measured resistivity, Hall effect, electron tunnelling and penetration depth for a series of 3-dimensional homogenous NbN thin films of varying disorder. We observe that the metal-insulator and superconductor-insulator transitions coincide at a single quantum critical point. The superconducting state is governed by quantum phase fluctuations arising due to spontaneous inhomogeneity. The phase fluctuations result in a pseudogap region, suppression of the superfluid density and suppression of the coherence peaks in the DOS.

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