J Supercond Nov Magn
DOI 10.1007/s10948-010-1038-8

ORIGINAL PAPER

Phase Diagram and Upper Critical Field of Homogeneously Disordered Epitaxial 3-Dimensional NbN Films

Mintu Mondal · Madhavi Chand · Anand Kamlapure · John Jesudasan · Vivas C. Bagwe · Sanjeev Kumar · Garima Saraswat · Vikram Tripathi · Pratap Raychaudhuri

Received: 14 September 2010 / Accepted: 17 September 2010

Abstract We report the evolution of superconducting properties with disorder, in 3-dimensional homogeneously disordered epitaxial NbN thin films. The effective disorder in NbN is controlled from moderately clean limit down to Anderson metal–insulator transition by changing the deposition conditions. We propose a phase diagram for NbN in temperature-disorder plane. With increasing disorder, we observe that as $k_F l \rightarrow 1$ the superconducting transition temperature (T_c) and normal state conductivity in the limit $T \rightarrow 0$ (σ_0) go to zero. The phase diagram shows that in homogeneously disordered 3-D NbN films, the metal–insulator transition and the superconductor–insulator transition occur at a single quantum critical point, $k_F l \sim 1$.

Keywords NbN films · Upper critical field (H_{c2}) · Metal–insulator transition · Superconductor–insulator transition

1 Introduction

The effective of disorder on superconducting properties of a material is one of the most challenging problems in condensed matter physics. In 1959, Anderson [1] proposed that the disorder arising from nonmagnetic impurities does not affect the superconducting properties. It was, however, understood later that the Anderson theorem is only applicable for weakly disordered systems [2]. In case of strongly disordered systems, as the disorder strength increases, the Bloch states become localized and give rise to Anderson Metal–Insulator transition (MIT). Close to the MIT, the superconductivity is controlled by the competition between the superconducting energy gap and the localization length (l_{loc}). If the superconducting energy gap (Δ) is larger than the energy level spacing ($\delta\varepsilon$) of the confined eigenstates within a length l_{loc}, the pairing of localized eigenstates is favorable and leads to local superconductivity on the scale of the l_{loc}. Eventually, through Josephson coupling between these localized superconducting regions, it gives a phase coherent superconducting ground state even in an insulating system. In this scenario, superconductivity persists in the insulating regime [3–7], as long as $N(0)l_{loc}^3\Delta > 1$, where $N(0)$ is the density of states at Fermi level. Eventually, as the disorder is further increased the system is expected to undergo transition from a superconducting to insulating ground state. However, there is some experimental evidence [8–10] that in many homogeneously disordered systems the superconductivity is completely suppressed on the metallic side of the metal–insulator transition while the system is still a metal. It has been argued [11] that the suppression of T_c in the disordered metallic state can arise from the increase in electron–electron interactions due to loss of effective screening in the presence of disorder. The enhanced coulomb interaction competes with the electron–electron (e–e) attractive interaction in the Cooper channel leading to the destruction of superconductivity in the metallic state.

Here, we study the evolution of superconducting transition temperature (T_c) and upper critical field (H_{c2}) as a

M. Mondal (✉) · M. Chand · A. Kamlapure · J. Jesudasan · V.C. Bagwe · S. Kumar · G. Saraswat · P. Raychaudhuri
Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400005, India
e-mail: mondal@tifr.res.in

P. Raychaudhuri
e-mail: pratap@tifr.res.in

V. Tripathi
Department of Theoretical Physics, Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400005, India

Published online: 12 October 2010

function of disorder in homogeneously disordered epitaxial 3D-NbN [12, 13]. All the films are in the 3-D limit with thickness (>50 nm) much larger than the dirty limit coherence length ($\xi \sim 5$ nm). The disorder spans a large range from moderately clean ($k_F l \sim 10$) to the Mott limit of minimum metallic conductivity, i.e., $k_F l \sim 1$. In this system, we present evidence of a coinciding metal–insulator and superconductor–insulator transition as the disorder approaches $k_F l \sim 1$. In this range of disorder, we observe that the Ginzburg–Landau coherence length (ξ_{GL}) estimated from upper critical field ($H_{c2}(0)$) increases by a factor of 2 with increasing disorder in contrary to the expectation in the weak disorder regime where ξ_{GL} decrease with increasing disorder.

2 Experimental Details

Epitaxial NbN films were grown on (100) oriented single crystalline MgO substrates using reactive d.c. magnetron sputtering of Nb in Ar/N_2 gas mixture. The effective disorder of these films was controlled by changing the sputtering power or the Ar/N_2 ratio, which allowed us to grow films over a large range of disorder. Details of preparation and characterization of these samples have been reported in Refs. [12, 13]. The temperature dependence of resistivity (ρ) was measured using standard four probe technique. To determine $H_{c2}(0)$, we have measured resistivity in presence of magnetic field. The $H_{c2}(T)$ was determined from $\rho(T, H)$ versus T data taken at different applied magnetic field. Hall coefficient (R_H) was measured using a standard four-probe ac technique on films patterned in Hall-bar geometry. R_H was calculated from Hall voltage deduced from reversed field sweeps from +12 to −12 T after subtracting the resistive contribution. The thickness of the films was measured using a stylus profilometer. For each film, $k_F l$ was determined from the ρ(285 K) and R_H(285 K) using the free electron formula $k_F l = \{(3\pi^2)^{2/3}\hbar[R_H(285K)]^{1/3}\}/[\rho(285K)e^{5/3}]$, where e is the electron charge.

3 Results and Discussion

Figures 1(a) and (b) show the resistivity as a function of temperature and Fig. 1(c) shows the conductivity (σ) as a function of temperature for a set of films with $k_F l$ ranging from $k_F l = 1.24$ to $k_F l = 10.12$. The superconducting transition temperature, T_c, of the films is determined from the temperature at which resistance falls to 10% of its normal state value. All the films with $k_F l \leq 8.13$ show a positive $d\sigma/dT$ up to room temperature. Here, we define σ_0 as the d.c. conductivity just above T_c. For films $k_F l \leq 8.13$, σ_0 is the minimum conductivity. All our films show finite σ_0 including the highest disordered film with $k_F l = 1.24$, indicating that all the films are in metallic regime.

Figures 2(a) and (b) show the variation of T_c and σ_0 as a function of $k_F l$ for all the films. From the trend of variation of T_c and σ_0 with $k_F l$, it is clear that both T_c and σ_0 are going to zero as $k_F l \to 1$. Thus, the MIT and the SIT in this system coincide at a single quantum critical point, $k_F l \sim 1$. Based on T_c vs. $k_F l$ and σ_0 vs. $k_F l$ data presented here, and the normal state data reported in Ref. [13], we propose the phenomenological phase diagram in Fig. 3.

The nearly exact coincidence of MIT and SIT observed in NbN is intriguing. While superconductivity can get destroyed either in the insulating or the metallic side of the MIT depending on the relative contribution of localization and e–e interactions, so far no theory explicitly predicts that these two transitions will coincide at a single point. Previous studies have indicated that both localization and Coulomb effect are present in our system [13]. While it is possible in

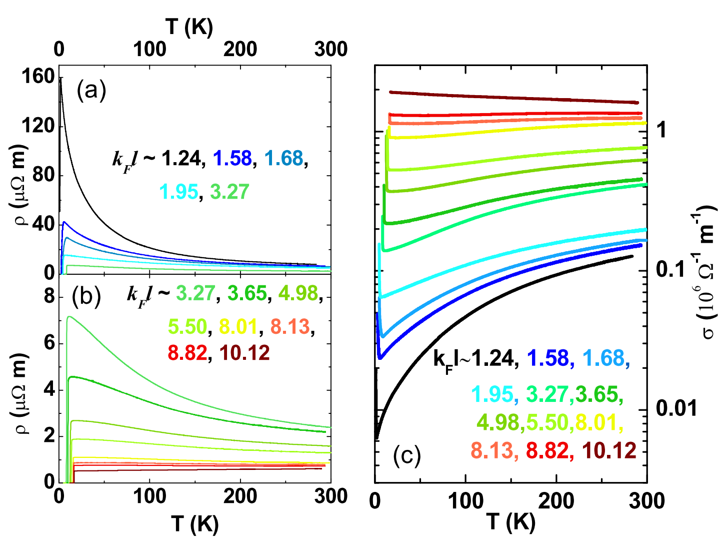

Fig. 1 (**a**) Resistivity (ρ) as a function of temperature (T) for a set of films with $k_F l$ value, from $k_F l = 1.24$ to $k_F l = 3.27$. (**b**) Resistivity (ρ) as a function of temperature (T) for a set of films with $k_F l$ value 3.27 to $k_F l = 10.12$. Note that the resistivity data for the films with $k_F l = 3.27$ is plotted in both (**a**) and (**b**). (**c**) Conductivity (σ) as a function of temperature (T) for a set of films with $k_F l$ values from $k_F l = 1.24$ to $k_F l = 10.12$

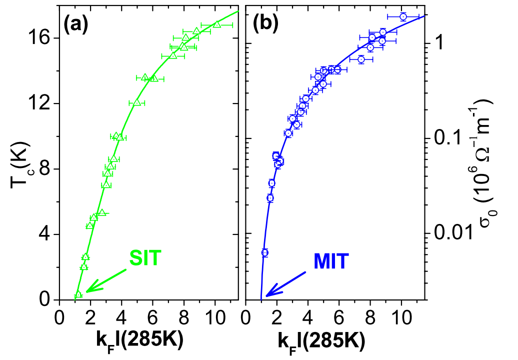

Fig. 2 (**a**) T_c vs. $k_F l$ for $k_F l \sim 1.23$ to 10.12, showing $T_c \to 0$ as $k_F l \to 1$; (**b**) σ_0 vs. $k_F l$ where σ_0 is in log scale showing $\sigma_0 \to 0$ as $k_F l \to 1$

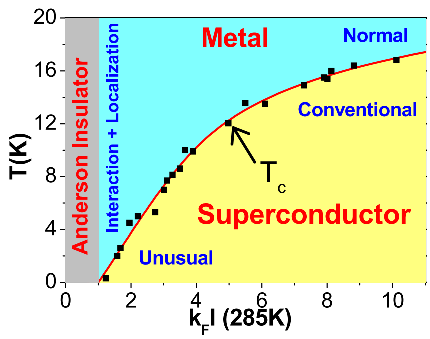

Fig. 3 Phase diagram of homogeneously disordered epitaxial 3-dimensional NbN films. The Metal–Insulator transition (MIT) and Superconductor–Insulator transitions (SIT) coincide at a single quantum critical point, $k_F l \sim 1$

principle for this coincidence to be purely accidental, such a possibility seems extremely unlikely. On the other hand, a coinciding transition has also been observed before in epitaxial boron-doped diamond films [14]. All previous studies on strongly disordered superconductors have been performed on amorphous or granular films. It is therefore possible that homogeneously disordered epitaxial superconductors behave qualitatively in a different way compared to their granular/amorphous counterparts.

Figure 4(a) shows the representative $\rho(H,T)$ as function of T for a film with $T_c \sim 4.2$ K at different magnetic field. $H_{c2}(T)$ is determined from the $\rho(H,T)$ vs. T data where $\rho(H,T)$ goes to 50% of its normal state resistivity just above T_c. $H_{c2}(T)$ as a function of T for a series of films is shown in Fig. 4(b). Since all our films are in the dirty limit, $l \ll \xi_{\mathrm{GL}}$, we have estimated $H_{c2}(0)$ and ξ_{GL} from the dirty limit relations [15]:

$$H_{c2}(0) = 0.69 T_c \frac{dH_{c2}}{dT}\bigg|_{T=T_c} \quad \text{and} \quad \xi_{\mathrm{GL}} = \left[\frac{\phi_0}{2\pi H_{c2}(0)}\right]^{1/2}. \tag{1}$$

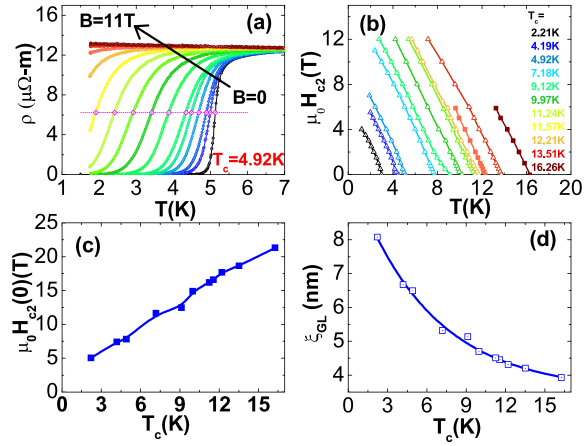

Fig. 4 (**a**) Resistivity (ρ) as a function of temperature (T) for film of $T_c \sim 4.92$ K in different magnetic fields. The successive magnetic field values are 0, 0.25, 0.5, 1, 1.5, 2, 2, 3, 4, 5, 6, 7, 8, 9, 10, and 11 T; (**b**) The upper critical field (H_{c2}) as a function of temperature (T) for series of films with different superconducting transition temperature (T_c). (**c**) $H_{c2}(0)$ as a function of T_c; (**d**) GL coherence length (ξ_{GL}) as a function of (T_c)

Figures 4(c) and (d) show the variation of $H_{c2}(0)$ and ξ_{GL} as a function of T_c obtained using (1). It is interesting to note that ξ_{GL} increases monotonically by a factor of 2 while T_c decreases by one order of magnitude. In case of weak disorder regime where Anderson theorem is valid, ξ_{GL} decreases with increasing disorder due to the reduction in electronic mean free path (l). In our case, however, the BCS coherence length (ξ_{BCS}) itself increases due to the decrease in T_c. Therefore, $\xi_{\mathrm{GL}} = (\xi_{\mathrm{BCS}} l)^{1/2}$, is determined by a competition between ξ_{BCS} and l. We have independently estimated ξ_{BCS} from the measured superconducting energy gap for $T \to 0$ (Δ_0) from tunneling measurement, Fermi velocity (v_{F}) and electronic mean free path (l) using the following dirty limit BCS relation:

$$\xi_{\mathrm{BCS}} = \frac{\hbar v_F}{\pi \Delta_0} \quad \text{and} \quad \xi_{\mathrm{GL}} = (l \xi_{\mathrm{BCS}})^{1/2}. \tag{2}$$

The calculated numbers reproduce the experimental trend while the estimated coherence length (ξ_{GL}) using dirty limit BCS relation is approximately twice the experimentally measured value for the respective disordered film. The relative insensitivity of ξ_{GL} to disorder is because the increase in ξ_{BCS} due to reducing T_c is partially compensated by the decrease in mean free path.

Further investigations on the superconducting properties show that for low disorder NbN follows the conventional BCS behavior [16, 17]. However, as we approach the SIT the superconducting state is characterized by a gradual loss in the coherence peak in the tunneling density of states and a linear variation of the superfluid density with temperature over a large temperature range. The unusual behavior in tunneling density of states and superfluid density will be discussed in a future publication [18].

4 Conclusion

We have established a phase diagram for homogeneously disordered 3-dimensional NbN films where MIT and SIT coincide at single quantum critical point, $k_F l \sim 1$. At this moment, it is not clear whether this coincidence is merely accidental or has a deeper significance. We have seen that the coherence length (ξ_{GL}) monotonically increases with disorder in contrast with expected behavior from weak disorder limit.

References

1. Anderson, P.W.: J. Phys. Chem. Solids **11**, 26 (1959)
2. Ma, M., Lee, P.A.: Phys. Rev. B **32**, 5658 (1985)
3. Sacépé, B., Chapelier, C., Baturina, T.I., Vinokur, V.M., Baklanov, M.R., Sanquer, M.: Phys. Rev. Lett. **101**, 157006 (2008)
4. Sambandamurthy, G., Engel, L.W., Johansson, A., Peled, E., Shahar, D.: Phys. Rev. Lett. **94**, 017003 (2005)
5. Feigel'man, M.V., Ioffe, L.B., Kravtsov, V.E., Yuzbashyan, E.A.: Phys. Rev. Lett. **98**, 027001 (2007)
6. Ghosal, A., Randeria, M., Trivedi, N.: Phys. Rev. Lett. **81**, 3940 (1998)
7. Ghosal, A., Randeria, M., Trivedi, N.: Phys. Rev. B **65**, 014501 (2001)
8. Furubayashi, T., Nishida, N., Yamaguchi, M., Morigaki, K., Ishimoto, H.: Solid State Commun. **55**, 513 (1985)
9. Bishop, B.J., Spencer, E.G., Dynes, R.C.: Solid-State Electron. **28**, 73 (1985)
10. Hertel, G., Bishop, D.J., Spencer, E.G., Rowell, J.M., Dynes, R.C.: Phys. Rev. Lett. **50**, 743 (1983)
11. Finkelshtein, A.: JETP Lett. **45**, 46 (1987)
12. Chockalingam, S.P., Chand, M., Jesudasan, J., Tripathi, V., Raychaudhuri, P.: Phys. Rev. B **77**, 214503 (2008)
13. Chand, M., Mishra, A., Xiong, Y.M., Kamlapure, A., Chockalingam, S.P., Jesudasan, J., Bagwe, V., Mondal, M., Adams, P.W., Tripathi, V., Raychaudhuri, P.: Phys. Rev. B **80**, 134514 (2009)
14. Klein, T., Achatz, P., Kacmarcik, J., Marcenat, C., Gustafsson, F., Marcus, J., Bustarret, E., Pernot, J., Omnes, F., Sernelius, Bo E., Persson, C., da Ferreira Silva, A., Cytermann, C.: Phys. Rev. B **75**, 165313 (2007)
15. Werthamer, N.R., Helfland, E., Honenberg, P.C.: Phys. Rev. **147**, 295 (1966)
16. Kamlapure, A., Mondal, M., Chand, M., Mishra, A., Jesudasan, J., Bagwe, V., Benfatto, L., Tripathi, V., Raychaudhuri, P.: Appl. Phys. Lett. **96**, 072509 (2010)
17. Chockalingam, S.P., Chand, M., Kamlapure, A., Jesudasan, J., Mishra, A., Tripathi, V., Raychaudhuri, P.: Phys. Rev. B **79**, 094509 (2009)
18. Mondal, M., et al.: arXiv:1006.4143