Superconducting Fluctuations above $T_c$ and pair breaking parameters of two dimensional Niobium Nitride Films

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Transport properties have been investigated for the epitaxial superconducting NbN thin films. We analysed the excess conductance $\sigma' = \sigma(T) - \sigma^N$ by the sum of the Aslamazov-Larkin (AL) and Maki-Thompson (MT) terms for thermal fluctuations above $T_c$, where the $\sigma^N = 1/R_{sq}^N$ is the normal state sheet conductance. We have found that the theoretical expression $\sigma'_{\text{theo}}(T) = \sigma'_{\text{AL}}(T) + \sigma'_{\text{MT}}(T, \delta)$ can be well fitted to $\sigma'_{\text{exp}}(T)$ with use of the suitable value of the pair breaking parameter $\delta$ in the MT term relating to the inelastic scattering rate $1/\tau_{\text{in}}(T)$ as $\delta = \pi \hbar / 8 k_B T \tau_{\text{in}}$. The rate $1/\tau_{\text{in}}(T)$ given by the sum of $1/\tau_{\text{fluc}}(T)$, $1/\tau_{\text{e-e}}(T)$ and $1/\tau_{\text{e-ph}}(T)$ is determined from the analysis of the magneto-conductance $\Delta \sigma = \sigma(H) - \sigma(0)$ by the sum of AL, MT and the localization terms, where the first, second and third terms correspond to the rate due to the superconducting fluctuation effect, electron-electron and electron-phonon interactions, respectively. The $R_{sq}^N$ dependence of $\delta$ is expressed by $\delta = \delta_0 + \alpha R_{sq}^N$, where the first term $\delta_0$ due to $1/\tau_{\text{e-ph}}(T)$ and the second term due to the sum of $1/\tau_{\text{fluc}}(T)$ and $1/\tau_{\text{e-e}}(T)$. Although we obtained a reasonable value of Debye temperature $\Theta_D \approx 630$ K from the $\delta_0$, the magnitude of the $\alpha$ is about 5 times larger than the theoretical value.

1. Introduction

Transport properties of low-dimensional superconductors (LDSs) as ultra-thin films and nanowires are strongly affected by the thermal and quantum fluctuations. Then LDSs have been widely investigated from the viewpoints of not only device physics [1] but also fundamental physical interests as $R-T$ characteristics due to the excess conductances above superconducting transition temperature $T_c$, thermal [2] and quantum Phase slips. As for superconducting-insulator transition in two dimensional
(2D) weakly disordered NbN systems, we have already reported the transport properties of NbN films with different normal state resistance \( \sigma_{sq}^N \) prepared by tuning the film thickness \( d \) of epitaxial film on MgO substrates.[3] We analyzed the suppression of \( T_c \) by Finkel’stein formula[4] from the localization theory called as fermionic scenario. This theory shows that the \( T_c \) depression is determined by the bulk \( T_{cb} \) and elastic scattering time \( \tau \), depending on materials.

Decades since 1970, superconducting thermal fluctuations at temperatures above \( T_c \) have been intensively investigated. Conventionally, the temperature dependence of excess conductance \( \sigma'(T) = \sigma(T) - \sigma^N \) for 2D superconductors has been mainly analyzed by the sum of the two dominant Aslamazov-Larkin (AL) [5] and the Maki-Thompson (MT) [6] terms, where the \( \sigma^N \) is the normal state conductance. One of the purposes for investigations of the \( \sigma'(T) \) due to the superconducting fluctuations is the determination of the some inherent characteristics because the theoretical formula for \( \sigma'(T) \) to fit the data contains several microscopic quantities as parameters. The MT term includes the pair breaking parameter \( \delta \) introduced for removing the divergence from the Maki diagram.[7] The progress in the electron localization theory[8] has revealed that \( \delta \) is essentially the same as the inverse of the inelastic scattering time \( 1/\tau_{in}(T) \), that is,

\[
\delta = \frac{\pi \hbar}{8k_B T \tau_{in}(T)}.
\]

From fitting the theory to the experimental data of \( \sigma'(T) \), we can determine \( \delta(T) \), that is, \( \tau_{in}(T) \). On the other hand, we can obtain \( \tau_{in}(T) \) from the analysis of the magneto-conductance \( \Delta \sigma \), defined as \( \Delta \sigma(T,H) = \sigma(T,H) - \sigma(T,0) \). Following the previous investigations of \( \sigma_{sq}^{N} \) dependence of \( T_c \) for the homogeneous 2D NbN/MgO films [3], in the present study we report the detailed result of the investigation of \( \sigma'(T) \) at \( H = 0 \) and \( \Delta \sigma(T,H) \).

2. Sample Preparation and Experimental Procedures

NbN specimen films were prepared by deposition on (100) single crystal MgO substrate with use of the DC magnetron sputtering method. Film thickness \( d \) is in the range from 2.0 nm to 25.8 nm. The \( d \) was estimated from the sputtering time using the calibrated relation between the sputtering time and exact thickness by the atomic force micrometer in thick region. We measured the resistance \( R(T,H) \) using a standard dc four-probe technique. We applied a magnetic field perpendicular to the film surface up to \( \pm 5 \) T. The details of the preparation procedures and microcrystal structures have been shown in previous report.

3. Experimental result and discussion

Firstly, we discuss the temperature dependence of the excess conductance \( \sigma'(T) = \sigma(T) - \sigma^N \) in the absence of the external magnetic field. To clarify the characteristic of \( \sigma'(T) \) near \( T_c \), Fig.1 shows the \( T \) dependence of \( \sigma^N/\sigma'(T) = R_{sq}(T)/[R_{sq}^N - R_{sq}(T)] \) for the typical film with \( d = 5.2 \) nm and \( R_{sq}^N = 257.7 \) \( \Omega \), where the normal sheet resistance \( R_{sq}^N = 1/\sigma^N \) is defined as a resistance determined from the extrapolation of the nearly linear relation between \( 1/H \) and \( 1/R_{sq}(T) \) to \( 1/H = 0 \) at temperatures near \( T_c \). The excess conductance \( \sigma' \) above \( T_c \) for 2D superconductors is given by the sum of AL and MT terms as follows,
\[ \sigma' = \sigma_{\text{AL}} + \sigma_{\text{MT}} = (e^2/16\hbar) \times \left[ \frac{1}{\eta} + \frac{2}{\eta - \delta} \ln \frac{\eta}{\delta} \right], \]

where \( \eta = \ln(T/T_c) \) is the reduced temperature. Solid line is calculated from eq. (2) to fit the data for relatively wide temperature region with suitable values \( T_c = 11.11 \) K and the constant \( \delta = 0.33 \). Although the slight disagreements between the theory and the experimental data near \( T_c \), the good fit of eq.(2) to data means the existence of the MT term, that is, the pair breaking of the cooper pairs in the present NbN films. The slight disagreements suggest the temperature dependence of \( \delta \). To investigate the \( \delta(T) \) in detail at temperature near \( T_c \), the value of \( \delta \) was determined at each temperature by fitting \( \sigma_{\text{MT}} \) in eq.(2) to data on the excess conductance. Before discussion of \( \delta(T) \), we analyze \( \Delta \sigma(T, H) \) for determination of \( \tau_{\text{in}}(T) \) relating to the pair breaking parameter through eq.(1) in order to compare the \( \delta^2(T) \) estimated from eq.(2) with \( \delta^3(T) \) obtained from the combination of eq.(1) and the following eq.(3) including the \( \tau_{\text{in}}(T) \). The \( \Delta \sigma \) for 2D superconductors is given by the sum of the three terms as follows,

\[ \Delta \sigma = \Delta \sigma_{\text{L}}(D \tau_{\text{in}} H, D \tau_{\text{so}} H) + \Delta \sigma_{\text{MT}}(D \tau_{\text{in}} H, DH, \eta, T) + \Delta \sigma_{\text{AL}}(DH, \eta, T), \]

where each term corresponds to localization, Maki-Thomson and Aslamazov-Larkin terms, and \( \tau_{\text{so}} \) and \( D \) are the spin-orbit scattering time and diffusion constant, respectively. The exact expressions are given in the previous paper.[9] The quantity \( \tau_{\text{in}}(T) \) can be determined from the fitting procedure of eq.(2) to the experimental data with \( \tau_{\text{in}}(T) \) and \( T \) independent \( D \) regarding as fitting parameters.

Figure 2 shows the \( H \) dependence of \( \Delta \sigma \) for the same film as that shown in Fig. 1 at various temperatures from two typical temperatures \( T = 11.21 \) K and \( 11.35 \) K at bottoms to two typical temperatures \( 20.0 \) K and \( 25.0 \) K at uppers. From the experimental results of \( \Delta \sigma < 0 \) even at high temperatures and that the present system has a strong spin-orbit interaction, namely, \( \tau_{\text{so}}/\tau_{\text{in}}(T) \ll 1 \) due to relatively heavy atoms of Nb and N, we assume that the magnitude of \( \tau_{\text{so}} \) is negligibly small compared with that of \( \tau_{\text{in}}(T) \). The solid lines are calculated from eq. (3) with a suitable value \( \tau_{\text{in}}(T) \) at each temperature and \( D = 8.5 \times 10^{-3} \) m/s. This value is almost the same as the value \( D = 9.0 \times 10^{-3} \) m/s obtained from the magnitude of \( dH_{\text{c2}}(T)/dT \) at temperatures near \( T_c \) using the formula \( D = (4k_B/\pi e) \times |dH_{\text{c2}}(T)/dT|^{-1} \). The inset shows the \( T \) dependence of \( \tau_{\text{in}} \) obtained from fitting procedures. The inelastic scattering rate \( 1/\tau_{\text{in}} \) is given by the sum of rates due to superconducting fluctuation, electron-electron (e-e) scattering [9] and electron-phonon (e-\( \phi \)) scattering [10] mechanism as follows;
1/T_m = 1/T_{ph} + 1/T_{el-ph} + 1/T_{el-phon}

$$= \left( \frac{k_B^2 R_{sq}^N}{2 \pi \hbar^2} \right) [2 \ln 2 / (\eta + \gamma) + \ln(4 \pi \hbar / e^2 R_{sq}^N)] \lambda \Theta_D^2 \left[ 1 + \frac{4 \pi^2 \epsilon^2(\gamma) k_B \hbar}{\pi \hbar} \right]$$

(4)

where $\gamma = 4 \ln 2 / [(c^2 + 128 \hbar e^2 R_{sq}^N) - \epsilon]$, $\epsilon = \ln(\pi \hbar / e R_{sq}^N)$, $\lambda$ is the electron-phonon coupling constant and $\Theta_D$ is the Debye temperature. The solid line shows the relation $T_m \propto 1/T$ expected from the $el-el$ scattering, the second term in eq. (4). The characteristic of $T_m \propto 1/T^n$ with $n > 1$ at high temperatures above $T \geq 17$ K comes from the combination of the $el-ph$ and $el-el$ scattering mechanisms. At the lowest temperatures, $T_m(T)$ shows drastic changes with decreasing temperature. The steep decrease may come from the first term, in eq.(4) that is, the superconducting fluctuation. However, the rapid increase below $\sim 15K$ cannot be understood by eq.(4). Such anomalous behaviors of $T_m(T)$ are frequently observed for other films with different $R_{sq}^N$. At the present stage, we have not exact idea to explain the $T_m(T)$ data at low temperatures.

Figure 3 summarizes the temperature dependence of two pair breaking parameters $\delta^F(T)$ and $\delta^M(T)$. The $\delta^F(T)$ is obtained from the analysis of $\sigma^N(H=0)$ at $H=0$ in Fig. 1 with eq. (2) and the $\delta^M(T)$ is obtained from the eq.(1) with use of $T_m(T)$ in Fig. 2 determined from the analysis of $\Delta\sigma(H,T)$ by eq.(3). As temperature approaching $T_c$, both magnitudes of the $\delta^F(T)$ and $\delta^M(T)$ decrease and then seem to strongly increase. Although the ratio $\delta^F(T)/\delta^M(T)$ maximally has about 1.4 at high temperatures, two series of $\delta$ show qualitatively almost the same temperature dependence. The characteristic of $\delta$ depending on temperature shows that the inelastic scattering rate $1/T_m(T)$ is given by the sum of different scattering mechanisms.

Figure 4 shows the $R_{sq}^N$ dependence of $\delta^F$ determined at $\eta = 0.1$ for all films in the present work. The magnitude of $\delta^F$ shows the gradual change at the low $R_{sq}^N$ region and seem to approach a certain value $\delta_0$. From the combination of eq. (1) and eq. (4), when $R_{sq}^N = 0$ only the $el-ph$ inelastic scattering mechanism contributes to the theoretical value $\delta(0) = (7\pi^2 \lambda \zeta(2)/4) (T/\Theta_D)^2$. This means that the Debye temperature $\Theta_D$ can be estimated form the experimentally extrapolated value $\delta_{exp}(0)$. By assuming $\delta_{exp}(0) \approx 0.09$ and typical value $\lambda = 1.5$ for superconducting materials, we obtain $\Theta_D \approx 630$ K with use of $T = 1.1 \times T_{el-ph} (14.85 K)$ in the above relation. Here, the factor 1.1 comes from the experimental condition of $\eta = \ln(T/T_d) \approx 0.1$ and $T_{el-ph}$ is the bulk superconducting transition temperature determined from the $R_{sq}^N$ dependence of $T_c$ in the previous report for the same NbN series. [3] Although the present value of $\Theta_D \approx 630$ K is lower than the bulk value $\Theta_D_{bulk} \approx 750$ K [11], it is quite reasonable that the $\Theta_D_{film}$ for thin films is generally low because of the specimen size effect. [12] With use of the value $\Theta_D \approx 630$ K obtained from the above estimation, the full expression for $\delta(R_{sq}^N)$ is given by

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**Figure 3.** $T$ dependence of $\delta^F$ and $\delta^M$ determined from the analysis of $\sigma^N(H=0)$ and $\Delta\sigma$, respectively. Lines are guides for eyes.

**Figure 4.** $R_{sq}^N$ dependence of $\delta^F$. Solid line is calculated from eq.(5) to fit the data with use of pre-factor $\alpha = 4.8$. 

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\[
\delta(R_{sq}^N) = \alpha (e^2 R_{sq}^N / 16h) \times \left[ \ln(n / e^2 R_{sq}^N) + 2 \ln 2 / (\eta + \gamma) \right] \\
+ (10.5 \pi^3 \zeta(2)/4) \times [1.1 \times T_c (R_{sq}^N) / 630]^2,
\]

where the factor \( \alpha \) is the suitable parameters to fit the theory to data and \( T_c(R_{sq}^N) \) is given in the previous report. [3] The solid line in Fig.4 is fit of eq.(5) to data with use of the pre-factor \( \alpha \approx 5 \) and the other lines show each contribution due to the el-el, el-ph and fluc. inelastic scattering rates. Although the obtained value of \( \Theta_D \approx 630 \) K for NbN film series is reasonable, at the present stage we have no explanation for this difference of the coefficient \( \alpha \) between the theory and experiment.

4. Conclusion
We prepared homogeneous NbN thin films with various \( R_{sq}^N \) to investigate the superconducting transport properties above \( T_c \). We measured the excess conductance \( \sigma' \) and magnetoconductance \( \Delta \sigma \) to estimate the pair breaking parameter \( \delta \) in the MT term and inelastic scattering time \( \tau_{in}(T) \). We analyze the \( R_{sq}^N \) dependence of \( \delta \) determined from \( \sigma' \) at \( H = 0 \) with the sum of three contributions due to the electron-electron, electron-phonon and superconducting fluctuation inelastic scattering rates, simply expressed as \( \delta = \delta_0 + aR_{sq}^N \). From the value of \( \delta_0 \) estimated form extrapolation of \( R_{sq}^N \) to \( R_{sq}^N = 0 \) in the \( \delta - R_{sq}^N \) data, we obtained the Debye temperature \( \Theta_D \approx 630 \) K for the present NbN films. However, the pre-factor \( \alpha \) is about 5 times larger than the theoretical value.

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