Normal-state transport in superconducting NbN films on r-cut sapphire

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Abstract. High-quality thin NbN films are very crucial for realizing quantum devices. Here, we investigated electrical transport and noise properties of a series of thin NbN films of various thicknesses grown on r-cut sapphire substrate using a DC magnetron sputtering technique. The films exhibit non-uniform thickness dependencies for superconducting transition temperature ($T_c$) and normal-state resistivity. Morphological characterization of NbN samples of various thicknesses reveals uniform structure in thin films and granular structure in thick films. By measuring transport and noise properties in a normal state, we observe that the granular structure of NbN films does not have a strong effect on resistivity and does not cause an additional source of current noise.

1. Introduction

Niobium nitride (NbN) has become a material that is increasingly important for development of superconducting electronic devices for high-frequency, high-speed, and high-temperature operations because it has a high transition temperature ($T_c$), and good thermal cycling stability. Thin NbN films are widely used in low-temperature superconducting devices, such as single-photon detectors [1] and hot-electron bolometers [2], and it is considered as a potential material for application in superconducting quantum circuits [3-5]. Great interest on the fundamental level is related to effects of disorder on superconducting and normal state properties of NbN thin films [6-9] as well as a demonstration of thermal and quantum phase slips in NbN nanowires [10], an important phenomenon for understanding one-dimensional superconductivity. Therefore, there has been a growing demand for high-quality thin NbN films for a variety of applications.

Extensive studies of NbN during the past decades [11-14] show that properties of NbN are strongly dependent on deposition-related conditions such as crystal structure and lattice constant. The latter is usually a direct result of the choice of a substrate material. NbN films grown on sapphire ($\text{Al}_2\text{O}_3$) substrates, preferably with c-cut and a-cut orientations, demonstrate a prospective performance with a high $T_c$ and low resistivity in a normal state [12, 15, 16]. Here, we focus on study of transport and electronic properties of NbN thin films grown on r-cut sapphire, which is the least studied system. As will be shown further, morphological characterization of NbN samples of various thicknesses on r-cut sapphire reveals uniform structure in thin films and granular patterns in thick films.
2. Fabrication and experiment

Fabrication of samples includes the following steps: (1) NbN film deposition, (2) fabrication of gold metallic pads, and (3) patterning of NbN film. NbN film deposition is performed in an ultra-high vacuum system (AJA Orion series). The r-cut sapphire substrates are preheated up to 400°C with a resistive heater supplied with a PID controller. After 10-minute temperature stabilization niobium target (2” diameter, 99.95% purity) is first pre-conditioned in argon atmosphere for 5 minutes followed by 5-minute discharge stabilization in a gas mixture containing 20% of nitrogen. Set current of 600 mA resulted in approx. 300 W power at operating pressure of 3.6 mTorr. The deposition of variable thicknesses is produced by opening the magnetron shutter and controlling the deposition time. The resulting thicknesses are calculated from pre-calibrated deposition rate obtained with a quartz crystal nanobalance detector. The calculated deposition rate is 0.065 nm/s, which is the optimal sputtering rate to control thickness below 10 nm. In the following step, the gold pads are fabricated by means of photolithography, e-beam evaporation, and lift-off processes. To pattern NbN film a negative resist mask is formed with electron-beam lithography, then unwanted NbN areas are removed using plasma-chemical etching in SF$_6$ atmosphere. The parameters of NbN films are presented in Table 1.

| $d$ (nm) | $R_s$(300 K) ($\Omega$/sq) | $r_R$ | $T_c$ (K) | $D$ cm$^2$/s |
|----------|-----------------------------|-------|-----------|--------------|
| 5        | 340                         | 0.83  | 11.7      | -            |
| 10       | 151                         | 0.85  | 12.6      | 0.474        |
| 30       | 55.9                        | 0.83  | 13.2      | -            |
| 50       | 37.6                        | 0.8   | 12.9      | 0.33         |
| 100      | 20.76                       | 0.74  | 13.3      | -            |
| 200      | 10.56                       | 0.74  | 14        | -            |
| 300      | 7.78                        | 0.72  | 13.9      | 0.29         |

The morphological characterization of the samples is performed using atomic force microscopy (AFM) setup from NT-MDT (INTEGRA series) in classical contact mode. Figure 1 reveals morphologies of NbN films for thicknesses 10, 100 and 300 nm. AFM images, presented in Figure 1(a-c), show a pronounced granular structure in the thickest film. Figure 1(d-e) presents analysis of granular structure of NbN films, measured by AFM. The effective diameter of a grain $D_s$ is defined as diameter of cross-sectional area of a grain. It is measured to be 2.66±0.02, 10.30±0.23, and 33.50±2.1 nm for 10-, 100- and 300-nm-thick NbN films, respectively. The change of film morphology with thickness increase may be related to a change of a growth mode, which is suggested to change from substrate-confined growth at the beginning (NbN phase) to the thermal dynamic growth Nb$_3$N$_3$ phase as the deposition time increased [14].

Further, we consider the effect of granular morphology on resistivity in a normal state. Figure 2 (a) shows the temperature dependence of normalized resistance for NbN films of different thicknesses. At decreasing $T_c$ the resistance increases and drops to zero at critical temperature $T_c$. The resistance slope above $T_c$ can be described by resistance ratio $r_R=R_{300}/R_{10}$. The parameters of $T_c$ and $r_R$ are presented in Table 1. Interestingly, we observe a noticeable increase of resistance above the resistive transition with increasing $d$ (see Figure 2 (a)). This effect cannot be explained by weak-localization phenomena, predicting such behaviour for films thinner than $L_w$, where $L_w \approx 4 - 10$ nm is the phase-breaking length in NbN at 15-20 K [17]. Normal-state resistivity $\rho$, obtained as $\rho = R_s d$, as a function of film thickness is presented in Figure 2 (b).
Figure 1. Morphological characterization of NbN films. (a-c) AFM images of the NbN films of 10, 100, 300 nm thickness, respectively. (d) Grain diameter distribution for one of the studied 100-nm NbN film. Solid line indicates the best-fitting normal grain diameter distribution. (e) Average grain diameter $D_g$ as a function of the film thickness $d$. The experimental dependence is fitted with a linear function (dashed line).

One can see that resistivity of the 5-nm NbN film is higher than for the 10-nm and 30-nm films, meanwhile for films with $d > 10$ nm it also gradually increases at increasing $d$. The change in $\rho$ is usually associated with an increase of disorder or emergence of additional scattering mechanisms. For example, the presence of grains can affect resistivity through scattering at grain boundaries. To define the effect of grains on $\rho$ we estimate the mean free path $l$ for electrons. Figure 2 (c) shows the temperature dependence of resistance at different values of an applied perpendicular magnetic field. From the temperature slope of upper critical magnetic field $B_{c2}$ (see inset of Figure 2 (c)) we estimate the diffusion coefficient $D = -\frac{4k_B}{\pi e} \left( \frac{dB_{c2}}{dT} \right)^{-1}$, where $k_B$ is the Boltzmann constant, $e$ is the electron charge.

Figure 2. Electrical and transport properties of NbN films in a normal state. (a) The temperature dependencies of normalized resistance of NbN films in a wide temperature range. (b) The thickness dependence of resistivity. The blue circles correspond to resistivity at 20 K, the orange circles correspond to resistivity at 300 K. (c) Main: the temperature dependence of resistance per square for the 50-nm thick film in an external magnetic field B applied perpendicular to the film. Inset: the temperature dependence of the upper critical magnetic field $B_{c2}$ determined at resistance $R = R_s/2$. 
The values of $D$ for 10, 50, and 300 nm thick films are presented in Table 1. Here, it is instructive to estimate the mean free path as $l=3D/v_F$, where $v_F$ is Fermi velocity. This yields estimation of mean free path $l=0.043-0.067$ nm at $v_F=2\times10^6$ m/s in NbN [15], where the shortest value of $l$ is determined for 300 nm film. Since obtained $l$ is much smaller than thickness and average grain size $D_g$, the Fuchs–Sondheimer’s surface scattering model [18-19] and the Mayadas-Shatzkes’ grain boundary scattering model [20] are not applicable for describing resistivity data in the studied NbN samples. We assume that an increase in resistivity and a decrease in the mean free path can be related to the growth of film disorder due to a long duration time of film deposition. In fact, a long deposition process is accompanied by absorption of nitrogen by the magnetron target and degassing of the magnetron chamber. Thus, the effect of grains on resistivity can be negligible.

Figure 3. (a) The current dependence of current shot noise $S_I$ for the 300-nm NbN sample (blue circles) is in comparison with the noise caused by granular structure $S_g$ (red dashed line). (b) Increase of temperatures $T_R$ and $T_N$ versus joule power ($P$) dissipated in the NbN sample. The $T_R$ is retrieved from the $R(T)$ dependence of the sample. The black dashed line corresponds to the fit by equation $P\sim T_e^{-3}T_b^{-3}$.

Next, we consider the effect of granular morphology on spectral density of current fluctuations ($S_I$) in a sample patterned from the 300-nm thick NbN film. The experimental setup for measurement of current fluctuations is described in Ref. [21]. We investigate the current noise in the normal state above $T_c$. Experiments are performed at a resonance frequency of 45 MHz, high enough to ignore possible $1/f$-like noise contributions. In this measurement, the sample is maintained in the sample holder in a vacuum. The samples are biased with a DC current at bath temperature ($T_b$) of 20-21 K. The Joule power ($P$), dissipated in NbN film is defined as $P=IV$, where $I$ is the bias current, $V$ is the measured voltage. In scenario of intrinsically inhomogeneous transport in a granular material, the fluctuations can be dominated by a partition noise of a network of nanoscopic weak links. Weak links could be modeled as tunnel barriers or constrictions, separated by large conductive regions with strong energy relaxation [21-22]. Here we suppose that the spectral density of current fluctuations can be determined as $S_I=2e|I|/N \coth(2e|V|/2k_BT_dN)$, where $N$ is a number of weak links defined as $N=L/D_g$. Figure 3 (a) shows the experimental dependence of $S_I$ for the NbN sample (0.33×64 μm²) in comparison with an estimate of the noise of weak links $S_g$ at $N=L/D_g=64$ μm/33 nm ~ 2000. One can see that the estimated $S_g$ predicts less noise than we observe here. In contrast, the experimental data can be described by electron heating in NbN film. In noise thermometry, the noise temperature $T_N$, which is equivalent to electron temperature $T_e$, can be found with the Johnson-Nyquist formula $T_N=S_I/(dV/dI)/4k_b$, where $dV/dI$ is a differential resistance of a sample. Figure 3 (b) shows the $P$ dependence of $T_N$ on a log-log scale. With intense heating, when the regime $T_e\gg T_b$ is achieved, we observe the $P(T_e)$ dependence with the heat power-law $P\sim T_e^{-3}T_b^{-3}$ (the dashed black line). In addition, we plot the electron temperature of the sample ($T_K$), which is reestablished from the $R(T)$ dependence and sample resistance $R=V/I$ data. With small heating, the electron temperatures obtained by means of noise
thermometry and secondary (resistive) thermometry give similar results. Meanwhile, at intense heating, we observe a small deviation $T_R$ from $T_N$, which can be related to the fact that the $R(T)$-dependence measured at a low current bias. Thus, we assume that the electron heating effect describes $S_I$ better than the current noise caused by granular structure of the sample.

In conclusion, our preliminary results demonstrate that granular structure of thick NbN films does not have a strong effect on resistivity and does not cause an additional source of current noise.

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