Monocrystalline NbN nanofilms on a 3C-SiC/Si substrate

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The authors have realized NbN (100) nanofilms on a 3C-SiC (100)/Si(100) substrate by dc reactive magnetron sputtering at 800 °C. High-resolution transmission electron microscopy (HRTEM) is used to characterize the films, showing a monocrystalline structure and confirming epitaxial growth on the 3C-SiC layer. A film ranging in thickness from 3.4 to 4.1 nm shows a superconducting transition temperature of 11.8 K, which is the highest reported for NbN films of comparable thickness. The NbN nano-films on 3C-SiC offer a promising alternative to improve terahertz detectors. For comparison, NbN nanofilms grown directly on Si substrates are also studied by HRTEM. © 2007 American Institute of Physics. [DOI: [10.1063/1.2766963](https://doi.org/10.1063/1.2766963)]

The ability to grow superconducting NbN films of several nanometer thick is of significant importance to the development of modern photon detector technology. Superconducting hot electron bolometer (HEB) mixers based on such nanofilms are the only sensitive heterodyne detectors for high-resolution spectroscopy at frequencies between 1.5 and 6 THz.^{1–4} These detectors will be used on the Herschel space telescope⁵ and are required in various future conceptual space missions.⁶ Another type of detector, the superconducting single photon detector (SSPD),⁷ is based on similar films and is ultrafast and sensitive for the detection of both visible and infrared photons. SSPDs can perform high speed photon counting which has many applications, for example, optical communications and quantum information.

To date, HEB mixers are based on ultrathin NbN films grown primarily on substrates such as Si with its native oxide,^{2–4} MgO,⁸ and Si with a buffering MgO film.⁹ SSPDs are based on NbN films grown on sapphire substrates. For films with an intended thickness of 3.5 nm (not directly measured), the highest superconducting transition temperatures (T_c) are reported to be 9.5–11 K.⁹ Among them, NbN films on MgO, MgO buffer layers and sapphire substrates have higher T_c than NbN films on Si substrates. These substrates allow for epitaxial growth of the NbN films,^{8–10} resulting in a monocrystalline structure. For HEB mixers, Si is a preferred substrate because of its low loss at terahertz frequencies, well-established processing technology, and inherent reliability. However, the drawback to using Si in this case is the limited intermediate frequency bandwidth, which is set by the thermal time constant.

In this letter, we demonstrate superconducting NbN nanofilms on a 3C-SiC buffered Si substrate. The films were characterized by high-resolution transmission electron microscopy (HRTEM). In addition, the superconducting properties were measured.

The 3C-SiC buffer layers were heteroepitaxially grown on Si (100) substrates by atmospheric pressure chemical vapor deposition at 1280 °C using a process described in detail elsewhere.¹¹ To ensure reasonably good crystal quality near the top surface of the 3C-SiC layer given its lattice mismatch with Si, we choose a thickness of 1 μ m for 3C-SiC layer. As reported previously,¹¹ the resulting films are (100) oriented, single crystalline 3C-SiC films. 3C-SiC has a zinc blende (cubic ZnS) structure with a lattice constant (a_0) of 4.36 Å,¹² while NbN has a face centered cubic (fcc) structure with $a_0=4.41$ Å,¹³ resulting in only a nominal lattice mismatch (1%) between the two materials. The previous group to exploit the small lattice mismatch between 3C-SiC and NbN was Shoji *et al.*,¹³ however, their NbN films were not ultra-thin (<10 nm), but rather 100-nm-thick films.

The NbN films were deposited on the 3C-SiC coated Si substrates by dc reactive magnetron sputtering of a Nb target in a reactive (Ar/N₂) gas mixture using a Z-400 Leybold Heraeus system. The target, having a diameter of 3 in. was positioned 50 mm from the substrate. The base pressure prior to deposition was 1.2×10^{-4} Pa. During deposition, the Ar partial pressure was 0.5 Pa, while the N₂ partial pressure was 1×10^{-2} Pa. The dc magnetron power was provided by current regulated power supplies set at 0.3 A with a resulting voltage of 270 V. The substrate was actively heated to 800 °C. The deposition rate was 0.5 nm/s, calibrated by sputtering a thick NbN layer and measuring its thickness. Prior to deposition, oxides on the 3C-SiC surface were removed by immersion in a diluted HF solution. The NbN

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FIG. 1. Cross-sectional HRTEM micrograph of an NbN nanofilm grown on a 3C-SiC/Si substrate: (a) viewed along the $\langle 110 \rangle$ direction for both NbN and SiC showing from the top, the glue (used to prepare the specimen), the NbN film, and the 3C-SiC layer. The bar is 5 nm. (b) Close-up view of the NbN film on 3C-SiC and (c) a magnified view of only the NbN film (from another specimen) which shows the monocrystalline structure of the NbN layer.

films were deposited for 7 s, targeting a thickness of 3.5 ± 0.5 nm. Two small samples from closely spaced regions of the substrate were extracted for analysis: one for HRTEM and the other for electrical characterization.

The crystalline structure and thickness of the NbN film were characterized by HRTEM. The specimen was prepared by mechanical grinding, polishing the cross section to a thickness of ~ 10 μm , and subsequently, by thinning to electron transparency using a Gatan PIPS 691 Ar ion milling tool. A CM300UT-FEG Philips transmission electron microscope operated at 300 kV was used for high-resolution imaging.

Figure 1(a) shows a cross-sectional HRTEM micrograph taken from a NbN film grown on 3C-SiC. Additionally, a zoomed view of the crystalline structure of the NbN and 3C-SiC layer is given in Fig. 1(b). It can be seen that the NbN exhibits the same lattice structure as the underlying 3C-SiC, suggesting that the film growth was epitaxial. In contrast with the bulk regions of NbN and SiC films, the NbN/SiC interface in the HRTEM micrograph is, however, not as clearly defined, yet still suggests an epitaxial transition between the 3C-SiC buffer layer and NbN substrate. To emphasize the monocrystalline structure in the NbN layer, Fig. 1(c) shows a cross-sectional HRTEM micrograph at larger magnification, taken from a second specimen.

The lattice constants can be determined in the HRTEM images for both NbN (200) and SiC (200) layers. The NbN-to-3C-SiC lattice constant ratio is 0.98–1.01, as determined from three distinct HRTEM images. As such the NbN film has the same lattice plane spacing as the underlying 3C-SiC film, even for a NbN film thickness of < 5 nm. This also shows that the electron-beam direction in the NbN is along a crystallographic direction similar to that of 3C-SiC, i.e., $\langle 110 \rangle$ with a cubic lattice with $a_0 = 4.36$ Å.

A complementary structural characterization was performed on NbN sputtered and unsputtered substrates by x-ray diffraction (XRD). In this case, the thickness of the NbN layer was 10 nm. We found a sharp peak in the XRD spectrum from both the NbN sputtered and unsputtered samples to be virtually at the same location at 41.4° , corresponding to a reflection from both (200) NbN and (200) 3C-SiC and further indicating an epitaxial relationship between the NbN film and 3C-SiC buffer layer.

FIG. 2. Bright field image of the NbN layer on a 3C-SiC/Si substrate. The inset shows a cross-sectional HRTEM micrograph of the 3C-SiC on Si. Electron beam direction is the $\langle 110 \rangle$ in both crystals.

For NbN, film thickness is a crucial parameter with respect to its superconducting properties and therefore detector applications. To determine the nominal NbN film thickness, we measured the film thickness using the HRTEM images taken at different locations. As shown in Fig. 1(a), the thickness varies from 3.4 to 4.1 nm roughly as would be expected based on the deposition rate (3.5 nm). However, in some other locations, the thickness was measured to be as high as 5 nm. The variations are due to the roughness of the NbN film originating from roughness and crystalline defects in the underlying 3C-SiC.

Although the NbN is monocrystalline, there are stacking faults and twinning defects present in the film. These defects were found by HRTEM imaging in combination with the fast fourier transformation (FFT) and are likely to be due to defects in the 3C-SiC film.¹² Figure 2 shows a HRTEM micrograph of a 3C-SiC/Si sample (in the inset) and a bright field (BF) image of the complete NbN/3C-SiC/Si structure, confirming the presence of stacking faults and other crystal faults in the 3C-SiC buffer layer. Also, the BF image indicates that the SiC surface is not entirely smooth on the microscopic scale. This may ultimately limit some device applications, however, it is not likely to be an issue for HEB mixers.

The measured resistance versus temperature (RT) of the NbN film on 3C-SiC is shown in Fig. 3. The transition temperature T_c , taken from the middle point of the resistive transition, is 11.8 K. The 10%–90% transition width ΔT_c is 1.1 K. In the same figure, the RT curve measured from a 5 nm NbN film on Si (to be discussed later) is also included, giving in this case a T_c of 9.5 K and a ΔT_c of 1.8 K. It becomes clear that the transition temperature of the NbN on 3C-SiC is higher, indicating enhanced superconductivity, while the transition width is slightly smaller, implying more uniform superconductivity. The sheet resistance ($R_\square$) of the NbN film is 600 Ω determined above T_c . The $R_\square$ could not be measured at room temperature because of the parallel electric conduction in the 3C-SiC layer.¹³ The $R_\square$ is nearly the same as a typical value obtained for the NbN films grown on Si, being 600 Ω at about 16 K and 540 Ω at room temperature.

We also studied NbN films on Si (100), essentially the same ones as used for HEB mixers,^{2–4} using HRTEM. These films were prepared using the same sputtering process as for those on 3C-SiC. Figure 4 shows two cross-sectional micrographs taken from different NbN films. The lattice plane

FIG. 3. Resistance as a function of temperature for both a NbN nanofilm on a 3C-SiC/Si substrate and a NbN film on a Si substrate.

spacings in the NbN layers were determined by performing FFT for a number of regions. We found that the NbN layers on Si were polycrystalline with a cubic NbN phase¹⁴ and a lattice constant $a_0=4.41$ Å. The crystal size was in the range of 2–5 nm. As for film thickness, the HRTEM image in Fig. 4(a) indicates an average thickness of 5 nm for the film exhibiting a T_c of 9.5 K (the detailed RT curve is given in Fig. 3), while the HRTEM image in Fig. 4(b) suggests a thickness of 6 nm for the film with a T_c of 9.8 K. In both cases, the films, however, are supposed to be 3.5 nm according to the deposition rate. The difference suggests that the deposition rate of NbN on Si (with native oxide), probably due to the sticking coefficient difference, is faster than that calibrated from thick NbN layers. Furthermore, in both micrographs the native oxide layer is clearly visible at the interface between NbN and Si.

FIG. 4. Cross-sectional HRTEM micrographs of two thin NbN films grown directly on Si substrates with native oxide, viewed along the $\langle 110 \rangle$ direction in Si. The film with a T_c of 9.5 K is shown in (a). The film with a T_c of 9.8 K is shown in (b). The bars are 5 nm.

In summary, epitaxy of monocrystalline, sub-5 nm-thick NbN on Si substrates has been realized by using 3C-SiC buffer layers. The film microstructure was studied using HRTEM in combination with XRD. Lattice defects present in the NbN films, such as stacking faults, are attributable to defects in the 3C-SiC buffer layer. A film with a thickness of 3.4–4.1 nm demonstrated a transition temperature of about 12 K, the highest temperature yet reported for NbN films of similar thickness. Further improvement in the performance of the NbN films can be expected with improvements in the crystal quality and surface characteristics of the 3C-SiC buffer layer. In terms of HEB mixers, the nanoscale NbN films on 3C-SiC have potential to reduce the thermal time by a factor of 2 because of the increased T_c and the small film thickness as compared with NbN films on Si substrates.¹⁵

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