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Epitaxial NbN/AlN/NbN tunnel junctions on Si substrates with TiN buffer layers

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We have developed epitaxial NbN/AlN/NbN tunnel junctions on Si (100) substrates with a TiN buffer layer. A 50-nm-thick (200)-oriented TiN thin film was introduced as the buffer layer for epitaxial growth of NbN/AlN/NbN trilayers on Si substrates. The fabricated NbN/AlN/NbN junctions demonstrated excellent tunneling properties with a high gap voltage of 5.5 mV, a large \( I_cR_N \) product of 3.8 mV, a sharp quasiparticle current rise with a \( \Delta V_g \) of 0.4 mV, and a small subgap leakage current. The junction quality factor \( R_s/R_N \) was about 23 for the junction with a \( J_c \) of 47 A/cm\(^2\) and was about 6 for the junction with a \( J_c \) of 3.0 kA/cm\(^2\). X-ray diffraction and transmission electron microscopy observations showed that the NbN/AlN/NbN trilayers were grown epitaxially on the (200)-orientated TiN buffer layer and had a highly crystalline structure with the (200) orientation.

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NbN-based Josephson tunnel junctions are promising superconducting components for use in high-frequency and high-speed Josephson devices because of their large gap voltage (5.0 mv) and high transition temperature (16 K).1–5 Epitaxial NbN/AlN/NbN junctions fabricated on single-crystal MgO substrates have excellent junction quality,6–8 and demonstrate good performance as superconducting electronics devices.9–12 In most superconducting electronics applications, the junctions must be prepared on various substrates. For example, superconductor–insulator–superconductor mixers are usually fabricated on substrates with low dielectric constants and single flux quantum circuits are prepared on Si wafers.13–15 However, because the superconducting properties of NbN films are dependent on the crystal structures of the film,16,17 it is difficult to fabricate high-quality NbN tunnel junctions on substrates other than MgO. Despite several attempts to develop NbN tunnel junctions using Si substrates,18,19 to date, there have been no reports of junctions with a high quality similar to that of epitaxial NbN junctions fabricated on MgO substrates. Therefore, to enable practical application of NbN junctions in superconducting electronics, it is important to develop epitaxial NbN tunnel junctions on Si or other substrates.

In order to improve the epitaxial growth of NbN films on Si substrates, a possible approach is to introduce a buffer layer between the NbN film and the Si substrate. Various thin films such as MgO, AlN, and TiN have been used as the buffer layers for the epitaxial growth of NbN films.20–23 Among these, TiN is a promising material to be used as the buffer layer because TiN and NbN have the same crystal structure with a lattice mismatch of only 3.5%. Recently, we fabricated (200)-oriented TiN films on Si (100) substrates using DC magnetron sputtering and showed that TiN films deposited at

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high temperature exhibit highly crystalline structures and good superconducting properties. In this study, we have tried to employ the (200)-oriented TiN films as a buffer layer for the epitaxial growth of NbN films and NbN/AlN/NbN trilayers on Si substrates. We fabricated epitaxial NbN/AlN/NbN tunnel junctions on Si substrates with TiN buffer layers and investigated their crystal structures and electrical properties.

TiN buffer layers were prepared by DC magnetron sputtering in a load-lock sputtering system with the background pressure below $3 \times 10^{-6}$ Pa. To improve (200)-oriented growth of the TiN films, the TiN buffer layers were deposited at $800\,^\circ\mathrm{C}$. The buffer layer thickness and roughness were 50 and 0.5 nm, respectively. A detailed description of the fabrication process and properties of TiN films is provided in our previous study. Following the deposition of the TiN buffer layer on the Si (100) substrates, the samples were transferred to a multichamber sputtering system for the deposition of the NbN/AlN/NbN trilayers. The sputtering system consists of four deposition chambers, a load-lock chamber, and a plasma cleaning chamber. The background pressure for each chamber was around $2 \times 10^{-5}$ Pa. NbN/AlN/NbN trilayers were deposited in situ by reactive DC magnetron sputtering from an 8-inch diameter Nb and Al targets at an ambient substrate temperature. The NbN films were deposited in an Ar + N$_2$ mixture with a 29:10 ratio of Ar (29 sccm) and N$_2$ (10 sccm), and the AlN films were deposited in pure nitrogen atmosphere. The total pressure was set to 0.25 Pa.

Fig. 1 shows the X-ray diffraction (XRD) patterns for the NbN/AlN/NbN trilayers with different AlN thicknesses (1 and 2 nm) and for a 150-nm-thick NbN film, all of them fabricated on Si (100) substrates with a 50-nm-thick TiN buffer layer. To characterize the crystal structure of the counter NbN electrode film, the thickness of the base NbN electrode films was set to 20 nm such that the XRD peaks originating from the base NbN thin films became negligibly small, while the thickness of the counter NbN films was fixed at 150 nm. As shown in Fig. 1, both the NbN/AlN/NbN trilayer and NbN single layer display (200) peaks with high intensities near the TiN (200) peak. The mismatch between NbN (200) and TiN (200) was only 4.2%, suggesting that NbN/AlN/NbN trilayers can be grown epitaxially on TiN buffer layers.

NbN/AlN/NbN tunnel junctions were fabricated using photolithography, reactive-ion etching, and a lift-off process. Fig. 2 shows the $I$–$V$ characteristics of the NbN/AlN/NbN junctions on TiN buffer layers with different values of critical current density $J_c$ measured at 4.2 K. The $J_c$ of the junction is defined as $I_c/A$, where $I_c$ is the Josephson critical current and $A$ is the junction area. Because it is difficult to accurately define $I_c$, we calculated $I_c$ as $I_g \pi/4$, where $I_g$ is the quasiparticle current measured at the inflection point of the quasiparticle and normal currents. The gap voltage $V_g$ is determined by the voltage at the $I_g/2$, and the gap voltage width $\Delta V_g$ is given by the difference between the voltages of the start and end of the superconducting transition of junction. In a wide

![XRD scans for NbN/AlN/NbN trilayers with different AlN thickness fabricated on Si substrates with TiN buffer. The thickness of the base and the counter NbN films are fixed to be 20 nm and 150 nm. The black line is the XRD pattern of 150-nm-thick NbN film.](image-url)
FIG. 2. $I−V$ characteristics of NbN/AlN/NbN tunnel junctions for different values of $J_c$. (a) 47 A/cm$^2$, (b) 193 A/cm$^2$, (c) 614 A/cm$^2$, (d) 1.7 kA/cm$^2$ and (e) 3.0 kA/cm$^2$ measured at 4.2 K.

range of $J_c$, the junctions demonstrated excellent tunneling properties with a high gap voltage, large $I_cR_N$ product, sharp quasiparticle current rise, and small subgap leakage current. The junction quality factor $R_{sg}/R_N$, where $R_{sg}$ is the subgap leakage resistance measured at 4 mV and $R_N$ is the normal resistance measured by the slope of the normal current, was about 23 and 6 for the junction when $J_c = 47$ and 3.0 kA/cm$^2$, respectively. For all junctions, the junction quality is similar to that of the epitaxial NbN/AlN/NbN junctions fabricated on single-crystal MgO substrates.\cite{6-8} These results suggest that our NbN/AlN/NbN junctions fabricated on the Si substrate with the TiN buffer layer are also epitaxial junctions.

Figure 3 shows the current density $J_c$ values of the junctions with different thicknesses of the AlN barriers. Junction parameters are listed in Table I, junction size was the designed value on mask. Because the junction area was defined by contact exposure photolithography and RIE etching process, the size of junction was smaller than the mask size due to overexposure and over etching of junction side. Therefore, actual $J_c$ values may be slightly larger than that measured from the mask size. The $J_c$ can be controlled by the thickness of the AlN barrier deposited at a very low sputtering
rate of 0.05 nm/s and varied from a few A/cm² to kA/cm² by changing the deposition time in the range of several tens of seconds. As shown in Fig. 3, \( J_c \) increases exponentially with decreasing barrier thickness \( d_{\text{AlN}} \) according to \( J_c = C \exp(-0.84 \, d_{\text{AlN}}) \), where \( C \) is a constant. The average barrier height \( \phi \) of the junctions is estimated to be about 0.7 eV by comparing the observed \( J_c - d_{\text{AlN}} \) relationship with the theoretical formula given by Simons\(^2\)

\[
J_c = 3.16 \times 10^{10} \frac{\sqrt{\phi}}{d} \exp(-1.025 \sqrt{\phi} d),
\]

where \( d \) is the barrier thickness measured in angstroms. The barrier height is about the same value as the epitaxial NbN/AlN/NbN tunnel junctions fabricated on single crystal MgO substrates (0.9 eV),\(^8\) suggesting epitaxial growth of NbN/AlN/NbN trilayers on Si substrates with TiN buffer layers.

To confirm the epitaxial growth of the NbN/AlN/NbN trilayers on Si substrates, we investigated the interface properties via transmission electron microscopy (TEM). Fig. 4 shows cross-sectional TEM images of NbN/AlN/NbN junctions on Si substrates with TiN buffer films. Fig. 4(a), 4(b), and 4(c) show the interfaces of Si and TiN, TiN and NbN and the NbN/AlN/NbN trilayer, respectively, and Fig. 4(d)–4(f) present the selected area electron diffraction patterns for each interface. As shown in Fig. 4, the TiN buffer layers on the Si substrates, the NbN films on the TiN buffer layer, and NbN/AlN/NbN trilayers display the same crystal structure with the (200) face orientation. These results are consistent with the XRD analysis results shown in Fig. 1, indicating that NbN/AlN/NbN trilayers can be grown epitaxially on Si substrates with TiN buffer layers.

In conclusion, we have fabricated epitaxial NbN/AlN/NbN Josephson tunnel junctions on a Si substrate with a TiN buffer layer. The junctions showed good Josephson tunnel properties with a high gap voltage, a large \( I_c R_N \) product, sharp quasiparticle current rise, and small subgap

| TABLE I. Electrical parameters of NbN/AlN/NbN tunnel junctions measured at 4.2 K. |
|---------------------------------|-------------|-------------|-------------|-------------|-------------|-------------|
| Barrier Thickness (Å) | \( J_c \) (A/cm²) | \( V_g \) (mV) | \( \Delta V_g \) (mV) | \( V_m \) (mV) | \( R_{2g}/R_N \) | \( I_c R_N \) (mV) | Junction Size (µm x µm) |
|----------------------|--------------|-------------|-------------|-------------|-------------|-------------|----------------|
| 19.8                 | 47           | 5.2         | 0.5         | 82          | 23          | 3.5         | 8 x 8         |
| 18                   | 193          | 5.4         | 0.4         | 76          | 21          | 3.6         | 6 x 6         |
| 17.1                 | 350          | 5.5         | 0.4         | 67          | 18          | 3.6         | 6 x 6         |
| 16.2                 | 614          | 5.5         | 0.4         | 64          | 17          | 3.8         | 6 x 6         |
| 15.3                 | 1700         | 5.0         | 0.3         | 15          | 6           | 2.6         | 4 x 4         |
| 14.4                 | 3000         | 5.0         | 0.3         | 17          | 6           | 2.9         | 6 x 6         |
leakage current and are of similar quality to the epitaxial NbN/AlN/Nb junctions fabricated on single-crystal MgO substrates. The crystal structures of NbN/AlN/Nb trilayers were investigated by XRD and TEM observations. Both the base and counter NbN films exhibit the same crystal structure with a (200) face orientation on the Si substrate with the TiN buffer. Our results demonstrated that the epitaxial NbN/AlN/Nb junction can be fabricated on Si substrates using a TiN buffer layer. This is suitable for many applications in superconducting electronics.

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