Epitaxial niobium nitride superconducting nanowire single-photon detectors

ABSTRACT

Superconducting nanowires used in single-photon detectors have become an indispensable resource for a range of quantum and classical applications due to their high detection efficiency over a broad spectrum, fast ultra-fast speed, exceptional timing performance, and ultra-low dark count noise. In this Letter, we demonstrate SNSPDs made from single-crystal NbN thin films grown by molecular beam epitaxy (MBE) on nearly lattice-matched AlN-on-sapphire substrates. This substrate platform is attractive for the integration of SNSPDs with several other elements of nitride-based photonic integrated circuits. The fabricated device consisting of 20 nm-wide and 6.3 nm-thick nanowire shows saturated internal efficiency at wavelengths of 780 nm and 1050 nm, while a further reduction in the achievable thin film thickness holds promise for saturating the efficiency further.

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Superconducting nanowire single-photon detectors (SNSPDs) have become an indispensable resource for a range of quantum and classical applications due to their high detection efficiency over a broad spectrum, fast ultra-fast speed, exceptional timing performance, and ultra-low dark count noise. In this Letter, we demonstrate SNSPDs made from single-crystal NbN thin films grown by molecular beam epitaxy (MBE) on nearly lattice-matched AlN-on-sapphire substrates. This substrate platform is attractive for the integration of SNSPDs with several other elements of nitride-based photonic integrated circuits. The fabricated device consisting of 20 nm-wide and 6.3 nm-thick nanowire shows saturated internal efficiency at wavelengths of 780 nm and 1050 nm, while a further reduction in the achievable thin film thickness holds promise for saturating the efficiency further.
generated using a radio frequency plasma source fed by ultrahigh-purity N₂ gas, which is further purified by an in-line purifier. Aluminum (99.9999% purity) is supplied using a Knudsen effusion cell. The Nb flux is generated using an in situ electron-beam evaporator source with 3N5-pure (excluding tantalum, Ta) Nb pellets in a tungsten hearth liner. The NbN films are grown at the temperature of 1100 °C, measured using a thermo-couple behind the substrate, and at a growth rate of approximately 1.0 nm/min.

The MBE film growth is monitored in situ using a reflection high-energy electron diffraction (RHEED) system operated at a voltage of 15 kV and a current of 1.5 A. Figure 1(a) shows sharp and streaky patterns formed by electron diffraction from the smooth surface of the NbN film, indicating the epitaxial nature of the single-crystal NbN film. As shown in Fig. 1(c), the in situ observation of oscillations of the RHEED intensity vs the growth time confirms that NbN grows in a 2D layer-by-layer growth mode on the AlN surface. The thicknesses of the NbN film and its native oxide layer are 6.3 nm and 0.29 nm, respectively. RHEED intensity monitored throughout the NbN thin film growth. The exhibited oscillations of the specular spot brightness indicate the 2D layer-by-layer growth mode of NbN. (d) Cross-sectional sketch of the thin film layer structure. (e) Measured sheet resistance of the NbN thin film vs temperature with the inset showing the Tc value of 12.1 K.

We fabricate SNSPD devices by patterning the MBE-NbN thin film. The nanowires are defined by the exposure of negative-tone 6% hydrogen silsesquioxane (HSQ) resist using 100 keV electron-beam lithography (Raith EBPG 5000+) and the subsequent development in 25% tetramethylammonium hydroxide (TMAH) for 2 min at room temperature. The HSQ resist is spun at the speed of 4000 rpm, resulting in an approximate thickness of 90 nm. In a second electron-beam lithography step, contact electrodes are defined using double-layer polymer methacrylate (PMMA) positive-tone resist. After the development in the mixture of methyl isobutyl ketone (MIBK) and isopropyl alcohol (IPA), we lift off the electron-beam evaporated 10 nm-thick Cr adhesion layer and 100 nm-thick Au in acetone overnight to form the contact pads. The HSQ nanowire pattern is then transferred to the NbN layer in a timed reactive-ion etching (RIE) step employing CF₄ chemistry and 50 W RF power. The HSQ resist is left on top of the NbN nanowires after fabrication, serving as a barrier to oxidation.

For initial tests, we fabricate short-nanowire detectors with widths ranging from 20 nm to 100 nm for comparison of the internal efficiencies. As shown in Figs. 2(a) and 2(b), the active detection parts of the devices are made of 20 μm-long straight nanowires, which are suitable for future waveguide integration. All the nanowires are serially connected to long 1 μm-wide meandered wires to prevent the detector latching at high bias currents. The sheet resistance of the devices is measured to be around 180 Ω/sq, which slightly increases compared to the value measured on the bare film prior to fabrication.

In order to characterize the optical response of the fabricated detectors, the detector chip containing multiple devices is mounted on a 3-axis stack of Attocube stages inside a closed-cycle refrigerator and cooled down to a base temperature of 1.7 K. Continuous wave (CW) laser light with varying wavelengths is attenuated to the single-photon level and sent to the detector chip via a standard telecommunication fiber (SMF-28) installed in the refrigerator. The detectors are flood-illuminated by fixing the fiber tip far away from the surface of the detector chip. We control the Attocube stages to move the detector chip at low temperature and make an electrical contact between the RF probes and the gold pads of the detectors. The RF probes are connected to a semi-rigid coaxial cable installed in the refrigerator, while the room-temperature end of the cable is attached to a bias-tee (Mini-Circuits ZFBT-6GW+±) to separate the DC bias current and RF output pulses for the detectors. The bias current is supplied by a programmable sourcemeter (Keithley 2401) in conjunction with a low-pass filter (1 kHz cut-off frequency). The output pulses of the detectors are amplified by a low-noise RF amplifier (RF bay LNA-650) and sent to a 4 GHz oscilloscope for the pulse observation or a pulse counter (PicoQuant PicoHarp 300) for the photon counting measurement. Figure 2(c) shows a single-shot trace measurement of the output voltage pulse from the 20 nm-wide detector. The decay time constant extracted from the exponential fitting (red dashed line) is 5.4 ns, which view of the superconducting transition region. The transition temperature of the film is measured to be Tc = 12.1 K, defined as the temperature where the normal state resistance of the film drops to 50% of that measured at 20 K. This value is higher than the previous results of NbN[10] and NbTiN[11] thin films sputtered on AlN substrates for SNSPD fabrication. The high Tc value is also consistent with the significantly low resistivity of the film, which is calculated to be only ~100 Ω·cm obtained by multiplying the thickness with the room-temperature sheet resistance.
translates into a sheet kinetic inductance of 24 pH/sq of the NbN film, assuming 50 Ω input impedance of the readout amplifier.

Figure 3 demonstrates the normalized photon counting rates (PCRs) as a function of the relative bias current to the switching current ($I_{bias}/I_{SW}$) for 20 nm-wide and 30 nm-wide nanowire detectors. $I_{SW}$ values of the devices are measured to be 25.5 µA and 38.8 µA, respectively, indicating a critical current density of ~20 MA/cm². We also measure $I_{SW}$ for wider nanowires (40–100 nm width), which show an excellent linear dependence on the wire width. The extrapolation of the data indicates a negligible “dead” width of the wires within the measurement errors, thereby confirming the absence of the edge damage effect during the fabrication process in contrast to the results reported by Charaev et al. As expected, detectors made of narrower nanowires with reduced $I_{SW}$ show better saturated internal efficiencies at a shorter wavelength. For the 20 nm-wide nanowire detector, we observe a clear saturation plateau at a wavelength of 780 nm, while the efficiency is only nearly saturated at a wavelength of 1050 nm. The minor fluctuation in the curve corresponding to a wavelength of 780 nm is due to the polarization instability of the laser since the photon absorption of the nanowire is significantly dependent on the polarization status of the incident photons. Neither the 20 nm-wide nor the 30 nm-wide nanowires show saturation behavior at a wavelength of 1550 nm. We attribute the inefficiency of the detectors to the possibly larger diffusion coefficient ($D$) of the single-crystal MBE-NbN material in comparison with sputtered or atomic-layer-deposited (ALD) poly-crystal NbN that has been utilized for the fabrication of high-efficiency SNSPDs. As indicated by the theory of photon detection mechanism based on hot-spot and vortex assistance, a larger $D$ allows faster diffusion of electrons, which favors a relatively low “temperature” of the hot electrons, because the energy of absorbed photons is confined to a relatively large volume at the initial stage of hot-spot formation. This, in turn, reduces the internal efficiency of the detectors owing to the decreased probability of superconducting state collapse under the assistance of the vortex-antivortex pair, which nucleates in the region with a less-suppressed superconducting order parameter. However, we expect that by further reducing the MBE-NbN film thickness down to 2–3 nm, this effect could be compensated, and saturated efficiency can be obtained at longer wavelengths with relaxed nanowire widths up to > 100 nm, as recently demonstrated with ultra-thin WSi and MoSi films. The growth of high-crystal-line-quality NbN films of 3 nm thick or less is achievable by MBE although a method to protect such thin films from oxidation upon exposure to the ambient air is necessary and under investigation. Future work will explore the suitability of such ultra-thin films for SNSPDs.

In summary, we have demonstrated the first SNSPDs patterned from MBE-grown single-crystal NbN thin films on AlN substrates. The 20 nm-wide SNSPDs show saturated internal efficiency at the wavelength of 780 nm and near-saturation at 1050 nm. We expect that single-crystal MBE-NbN could address the limited fabrication yield problem, which conventional poly-crystalline-Nb(Ti)N detectors suffer from, by removing the grain boundaries and thus reducing the defect area in the film to the minimum level. It is also worth mentioning that the AlN-on-sapphire substrate, which the epitaxial growth of
NbN relies on, is particularly attractive due to its potential of the on-chip integration of SNSPDs with versatile AlN nanophotonic circuits. The excellent optical functionalities of AlN, such as strong $\chi^{(2)}/\chi^{(3)}$ nonlinearity and large electro-optic effect, render NbN on AlN-on-sapphire a very attractive material platform for realizing fully integrated quantum photonic circuits with the generation, routing, active manipulation, and the final detection of single photons on a single chip.

**AUTHORS’ CONTRIBUTIONS**

R.C. and J.W. contributed equally to this work.

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**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

**REFERENCES**


