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Ultra-thin TaN films for superconducting nanowire single-photon detectors

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Ultra-thin films of superconducting tantalum nitride are deposited by reactive magnetron sputtering on heated sapphire substrates. The critical temperature $T_C = 10.5$ *K is reached for films thicker than 10 nm. A superconducting nanowire single-photon detector in the form of a meander line with a width of 110 nm was made from 5 nm thick TaN film. The detector had a transition temperature of 8.3 K and a critical current density of 4 MA/cm*^{2} *at 4.2 K. A photon detection efficiency of 20 % has been obtained for the detector with a filling factor of 0.55 at wavelengths up to 700 nm.*

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1. INTRODUCTION

At present Superconducting Nanowire Single-Photon Detectors (SNSPD) are under intensive development, where increasing the detection efficiency (DE) and extending the operational spectral range are two of the most important directions. There are several possible ways for the realization of these tasks. As it has been shown theoretically[1] and proofed experimentally[2] the cut-off wavelength of the spectral independent DE can be shifted to longer wavelength by reduction of the wire thickness or width, increase of the bias current or by using superconducting material with smaller energy gap. The first two geometric approaches can be realized by further improvement of the film fabrication technique and patterning procedure. The third one is quite delicate since it requires realizing experimentally the critical current of the nanowire close to the critical current due to breaking of Cooper pairs. Different materials like NbN[2], NbTiN[3], MgB_2[4] have been tested so far. There are several requirements for a material, which could be the next promising candidate for SNSPD:

- Suitability for fabrication of ultra-thin films. The films have to be mechanically and temporally stable, tolerating multiple thermo-cycles without degradation of their properties.
- Small electron diffusion coefficient D to slow down out-diffusion of excited non-equilibrium electrons from the photon absorption site. This requirement limits the use of Nb, which typically has a diffusion coefficient several times larger than that of NbN.
- Low density of electron states at Fermi level to increase the efficiency of an absorbed photon in suppressing superconductivity.
- Small energy gap to increase number of quasiparticles created by an absorbed photon.

It is rather difficult to find superconducting materials which would fulfill all mentioned requirements. Conventional low temperature superconductors with small energy gap, like Al or Ti are low resistive, relatively clean materials, which are characterized by large electron diffusivity. A certain compromise can be found among nitrides and carbides of transition metals. Nitrides and carbides are hard and chemically inert compounds that is why they are widely used for various mechanical and microelectronic applications[5,6]. Their properties can be varied in a wide range (from insulator to superconductor for instance) by the fine adjustment of their stoichiometry via variation of fabrication conditions. Thin and ultra-thin films of NbN (currently the most popular material for SNSPD) can be made with a thickness as small as 3-4 nm nevertheless showing superconducting, normal state and mechanical properties suitable for detector fabrication. A disadvantage of NbN though is the large superconducting gap, which is one of the largest among conventional superconductors.

Here we describe the technology of growing and patterning of ultra-thin TaN films. We report on their normal state and superconducting properties and analyze the perspective of using this material for fabrication of SNSPD devices.

2. TECHNOLOGY

Films of TaN were deposited on 10×10 mm^2 R-plane cut, one-side polished sapphire substrates, which were placed on the heater surface without any thermal glue. During deposition the temperature of the heater was kept at 750°C. Before deposition the surface of the Ta target (99.95%) was pre-cleaned by sputtering of the target material in a pure Ar atmosphere (see Fig.1 dashed line) at the pressure $p_{\mathrm{Ar}} \approx 3.5\times10^{-3}$ mbar. After the pre-cleaning the reactive gas (N_2) with a partial pressure $P_{\mathrm{N2}} \approx 3.8\times10^{-4}$ mbar was introduced into the vacuum chamber. After 10-12 min stabilization of the discharge voltage at $U \approx 375$ V a TaN film was deposited at the deposition current $I = 110$ mA (Fig. 1 solid line). At these conditions the deposition rate of TaN was ≈ 0.12 nm/s. The rate was inferred from the thickness and the deposition time of the test-film. The thickness was measured with a stylus profilometer at the film edge, which was formed by means of standard photolithography and reactive ion etching with Ar+SF_6 gas mixture. The thickness of all subsequent films was controlled by the deposition time.

3. TaN THIN FILMS

The temperature dependence of the resistance of all TaN films was measured right after deposition by means of the standard four-probe technique. For all studied thicknesses in the range from ≈ 2.3 to 18 nm the normal state resistance increases with decrease in temperature. Therefore, the resistivity ratio between 295 K and 10 K (R_{295}/R_{10}) remains less than one for all films. It decreases with the decrease in the film thickness from 0.99 at 18 nm to 0.8 at 2.3 nm. The zero resistance critical temperature T_C and the width of the transition ΔT were defined from $R(T)$ curves by $10^{-3}R_{10}$ and 0.1/0.9 R_N criteria, correspondingly, where R_N is the normal state resistance extrapolated in the transition region. The decrease in the TaN film thickness results in a decrease of T_C from 10.25 K measured on films thicker than 10 nm down to 6 K, which we measured for the 2.3 nm thick TaN film (Fig. 1). The width of the transition increases with the decrease in the thickness from less than 0.3 K for the thickest films to 2.5 K for the thinnest one. The decrease in T_C with the decrease in thickness has been already reported for Nb and NbN superconducting thin films and explained by the proximity effect[7]. The real film is represented by a model that considers superconducting films as NSN structures, where normal layers are formed at the film/substrate interface and at the surface of the film. The solid line in Fig. 1 is the best fit of our experimental results by Eq. (1) in Ref. 7. From the fit we have obtained a maximum critical temperature $T_C^0 = 10.9$ K of TaN films with thickness much larger than the thickness $d_N = 0.35$ nm of the surface layers with destroyed superconductivity. The surface layers with partly unbounded Ta atoms absorb oxygen and other impurities thus forming oxides and other non-superconducting Ta compounds. The thickness of this layer is close to the size of the unit cell of TaN[8] that confirms the stability of thin films of this material.

In the studied range of thicknesses, the resistivity of TaN films increases with the decrease in thickness from 125 to 250 μΩ cm. In contrast, the temperature dependences of the second critical magnetic field, which were measured in magnetic fields up to 5 T, demonstrate almost the same slope $dB_{C2}/dT \approx 2.15$ T/K independently of the film thickness. This gives the electron diffusivity of TaN films $D = -1.097(dB_{C2}/dT)^{-1}_{T=Tc} \approx 0.5$ cm^2/s. According to the Einstein relation $N(0)=(2e^2\rho_0 D)^{-1}$ this results in a twofold decrease of the electron density of states $N(0)$ at the Fermi level for one spin direction from $3*10^{47}$ J^{-1} m^{-3} to $1.5*10^{47}$ J^{-1} m^{-3} with the decrease in the film thickness,comparable to that in NbN films[9].

4. TaN SNSPD

SNSPDs with the standard meander-line structure have been made by means of electron-beam lithography and ion milling technique from a 5 nm thick TaN film. The width of the line $W = 110$ nm has been measured using scanning electron microscopy. The meander covers an area of 4x3.5 μm^2 with a 55 % filling factor and is embedded into a 50 Ω impedance coplanar readout line (see Fig. 2aThe studied devices demonstrate $T_C \approx 8.3$ K that is slightly smaller than that of the non-patterned film. The critical current at $T = 4.2$ K was close to 22 μA resulting in the value of the critical current density $j_C(4.2\text{ K}) = 4$ MA/cm^2. The T_C and $j_C(4.2\text{ K})$ values of the meander structure are close to those of micrometer wide bridges, which were made from the same film. This confirms a pretty good stability of TaN ultra-thin films during fabrication of sub-micrometer structures. Multiple thermo-cyclesof fabricated detectors either in the He-gas atmosphere or in vacuum did not lead to any distinguishable degradation of their superconducting and normal-state properties.

Photon count rates of TaN SNSPD have been measured in a helium bath cryostat at $T \approx 5$ K and $I_{\text{bias}}/I_C \approx 0.9$. Details of the experimental set-up and the method of extracting the detection efficiency (DE) and the intrinsic DE can be found in Ref. 2. The DE spectrum is shown in Fig. 2b. At wavelengths smaller than ≈ 700 nm the device DE is approximately 20 %. In this range of photon energies the response of SNSPD is determined by the hot-spot mechanism. Formation of a normal belt across the nanowire occurs due to destruction of Cooper pairs by the energy of the absorbed photon and acceleration of the remaining pairs above the critical velocity by the applied bias current[1]. Increasing the wavelength λ above 700 nm results in a decrease of the DE as $\lambda^{-8.5}$. In this part of the spectrum the photon energy is not sufficient to create the normal belt across the nanowire. The photon detection appears as a local enhancement of the probability of a dark count event. The dark counts in current biased nanowires are due to thermal excitation of magnetic vortices over the energy barrier, which prevents vortex entry[10,11]. The energy of an absorbed photon suppresses the barrier and facilitates the entry of a vortex, which is further driven across the line by the Lorentz force. Vortex movement causes energy dissipation and formation of the normal belt. These photon-activated vortex jumps have been used to explain similar DE spectra, which were observed in SNSPD from NbN ultra-thin films[2].

Using normal state and superconducting properties of the studied TaN structures we estimated the cut-off wavelength λ_0 of the hot-spot mechanism[1,2] via the corresponding photon energy

$$\varepsilon = \frac{hc}{\lambda_0} = \frac{3\sqrt{\pi}}{4\varsigma}\Delta^2 W d N(0)\sqrt{D\tau}\left(1-\frac{I_B}{I_C^d}\right). \quad (1)$$

In (1) ζ and τ are the effectiveness and the time scale of the energy transfer process from the absorbed photon to the electrons, $\Delta = 1.75 k_B T_C$ is the superconducting energy gap and I^d_C is the de-pairing critical current. We found a cut-off wavelength of 750 nm, which is in good agreement with experimental observations. Using the method which is described in detail in Ref. 2, we estimated for the 5 nm thick TaN SNSPD in the hot-spot regime the intrinsic detection efficiency ≈ 100 %. The overall experimental accuracy was better than 15% mostly defined by uncertainties in the measured device parameters.

CONCLUSIONS

We have developed a fabrication technology for thin and ultra-thin superconducting TaN films allowing us to produce stable films with T_C up to 10.5 K for thicknesses larger than 10 nm. SNSPDs with the meander-line structure, the line width 110 nm and the thickness 5 nm demonstrated 20 % detection efficiency at $\lambda < 700$ nm. We have estimated in this regime a 100 % intrinsic detection efficiency of the meander. Due to a smaller superconducting energy gap TaN SNSPDs offer an alternative to NbN SNSPD devices in the IR and NIR spectral ranges.

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REFERENCES

1 A.D. Semenov, A. Engel, H.-W. Hübers, K. Il'in, M. Siegel, *Eur. Phys. J. B* **47**, 495 (2005).

2 M. Hofherr, D. Rall, K. Ilin, M. Siegel, A. Semenov, H.-W. Hübers, N.A. Gippius, *J. Appl. Phys.* **108**, 014507 (2010).
3 B. Baek, J. A. Stern, S.W. Nam, *Appl. Phys. Lett.* **95**, 191110 (2009).
4 H. Shibata, H. Takesue, T. Honjo, T. Akazaki, Y. Tokura, *Appl. Phys. Lett.* **97**, 212504 (2010).
5 S.M. Rossnagel, *J. Vac. Sci. Technol. B* **20**, 2328 (2002).
6 R. Setzu, E. Baggetta, J.-C. Villegier, *J. of Phys.: Conf. Ser.* **97**, 012077 (2008).
7 K.S. Il'in, M. Siegel, A.D. Semenov, A. Engel, H.-W. Hübers, *Inst. Phys. Conf. Ser.* **181**, 2895 (2004).
8 E.I. Isaev, S.I. Simak, I.A. Abrikosov, R. Ahuja, Yu.Kh. Vekilov, M.I. Katsnelson, A.I. Lichtenstein, B. Lohansson, *J. Appl. Phys.* **101**, 123519, (2007).
9 A. Semenov, B. Günther, U. Böttger, H.–W. Hübers, H. Bartolf, A. Engel, A. Schilling, K. Ilin, M. Siegel, R. Schneider, D. Gerthsen, N.A. Gippius, *Phys. Rev. B* **80**, 054510 (2009).
10 H. Bartolf, A. Engel, A. Schilling, K. Il'in, M. Siegel, H.-W. Huebers, A. Semenov, *Phys. Rev. B* **81**, 024502 (2010).
11 L.N. Bulaevskii, M.J. Graf, C.D. Batista, V.G. Kogan, *Phys. Rev. B* **83**, 144526 (2011).

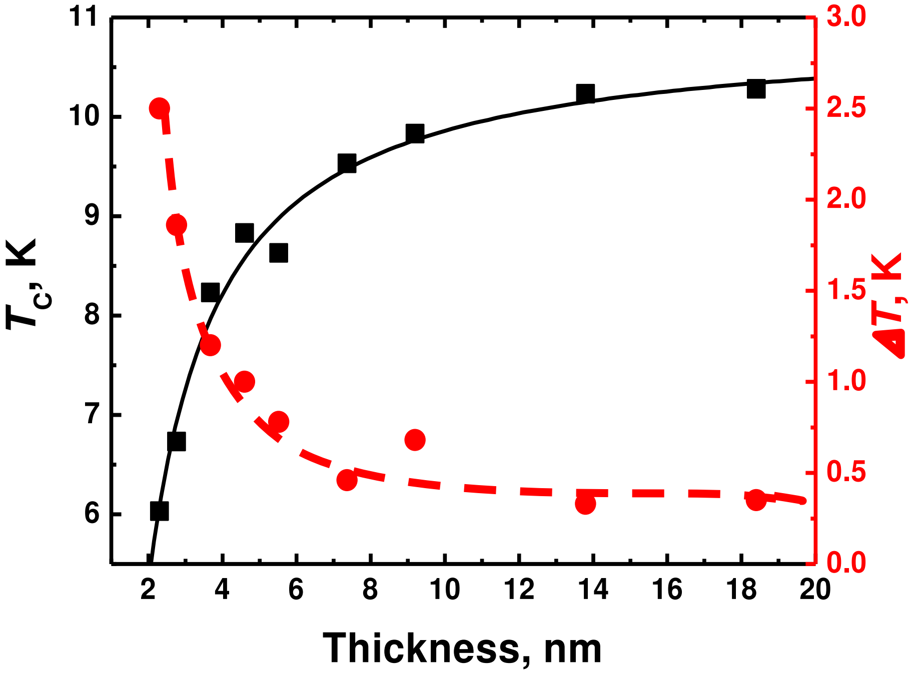

Fig. 1. (Color online) Dependence of the zero resistance critical temperature T_C (squares) and the width of superconducting transition ΔT (circles) on the thickness of TaN thin films. The solid line is the fit of $T_C(d)$ with Eq. 1 from Ref. 7. The dashed line is to guide the eye.

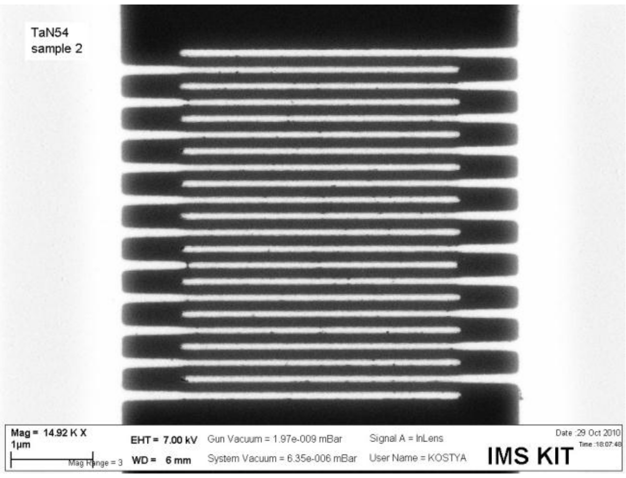

Fig. 2a

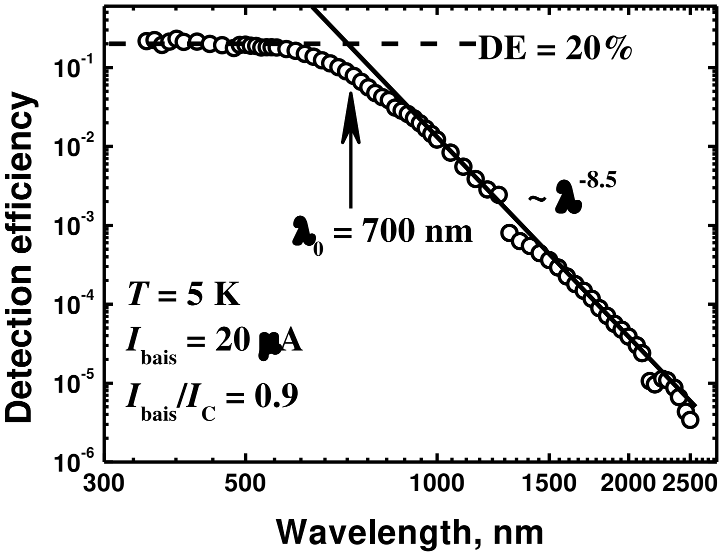

Fig. 2b

Fig. 2 (a) SEM image of TaN SNSPD; (b) spectrum of the detection efficiency of the 5 nm thick TaN SNSPD. The measurement conditions are indicated in the graph.