

National Research University Higher School of Economics

as a manuscript

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**ULTRA-FAST SUPERCONDUCTING SINGLE-PHOTON
DETECTOR FOR THE MID-IR RANGE BASED ON
NIOBIUM NITRIDE FILMS**

Dissertation summary
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Doctor of Philosophy in Engineering

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Relevance of the research topic

Superconducting single-photon detectors (SSPD)¹ represent a modern class of photon detectors cable of operation in the visible and infrared ranges demonstrating an excellent specifications: >90% system detection efficiency at <100 Hz dark count rates together with temporal resolution of the order of 30 ps and 50 MHz counting rates [0]. These characteristics are the key component for implementation of these devices to a wide range of quantum technologies [0]. First demonstration of SSPD operation was in 2001 at Moscow State Pedagogical University [0]. Back then demonstrated characteristics of the detector made this technology a strong rival for conventional single-photon detectors. Taking into account spectral limitations of the avalanche single-photon detectors and low speed of photomultiplier tubes, SSPDs became the only alternative that meets all of the possible requirements to the single photon detectors in the telecommunication wavelengths [0]. Moreover, it was demonstrated that dark count rate of SSPDs could be below 0.1 Hz and photoresponse could be extended to the mid-IR range but at a cost of detection efficiency reduction down to 1% [0, 0, 0, 0, 0, 0].

Listed strong sides of SSPDs brought the attention of scientists in the context of telecommunications, both fiber-optic-based and free-space. As a result, SSPDs were used in a quantum key distribution experiments [0] and in a free-space telecommunication channel between Earth and Moon with a data transfer rate of 622 Mbit/s [0]. Among other large projects that involved SSPDs are PICA (Picosecond Imaging Circuit Analysis) which in 2004 allowed inspection of the semiconductor microchips for malfunctions [0, 0]. Moreover, SSPDs demonstrated an excellent performance in a LIDAR system operated at 2.3 μm that demonstrated depth resolution of the order of 1 mm [0, 0].

Sensitive element in a SSPD is typically made of ultrathin superconducting film with 4 – 10 nm thickness, which is then structured into long strip (wire) that forms a sensel (sensor element) of the detector. Multi-element SSPD prototypes with hundreds of elements were made using different superconducting materials [0, 0]. It should be noted that even though superconductivity was observed in a variety of materials, only a small amount of them was found to be suitable for SSPD fabrication

¹Modified appellations could be found in the literature. For example, standard detectors with a nonosized active area are often denoted as superconducting nanowire single photon detectors and recently appeared devices with micron-scale strips as superconducting microwire single-photon detectors. WSSPD abbreviation could be found for SSPDs fabricated over a waveguide

[0]. In superconducting films used for SSPD fabrication processes of heat exchange, diffusion and relaxation play a critical role [0]. Today, for SSPD fabrication *dirty* and *disordered* materials are mainly used. In this categories two main groups of materials could be highlighted: noncrystalline films of transition metals (NbN, NbTiN, TiN, TaN, VN) and amorphous films based on silicides (WSi, MoSi). Besides the structure, another significant peculiarity of this materials is their transition temperature to the superconducting state T_c . Niobium nitride and tungsten silicides, two of the most used materials for SSPD fabrication, have critical temperature that differ by almost three times. Nevertheless, experiments showed that detection mechanisms in these two materials are identical, but have different time and space scales, which influence detection efficiency and speed of fabricated detectors [0].

Critical temperature is able to make a significant influence over functionality of the superconducting single-photon detectors. SSPDs based on WSi require operating temperatures in the range 300 -700 mK, while NbN SSPDs operate at temperatures 0.9-2.5 K that are obtained with compact closed-cycle refrigerators, which simplifies their integration into real-life applications. However, a low value of T_c could also be a benefit when detectors are required to operate in the mid-IR range — low-energy photons would cause a more probabilistic disruption of superconducting state in a superconductor with a lower energy gap $\Delta = 1.76k_B T_c$. Therefore, on the one hand, materials with a low energy gap simplify extending operation range toward mid-IR, and on the other, complicate and make more expensive application of such detectors on practice. However, a critical characteristic for most of the practical applications besides (spectral) detection efficiency is speed. Due to larger electron thermalization time in WSi films, as well as low operating currents of WSi SSPD, these devices have low temporal resolution and low achievable counting rates, even though their detection efficiency is comparably high [0]. These critical differences make niobium nitride a more preferable material for fabrication of fast and efficient detectors for practical applications.

Extending the operation range of single-photon detectors towards long-wave region of the IR is a relevant scientific task. By solving this task a significant progress could be made in realization and optimization of communication channels in a free-space, LIDAR, and medical visualization [0, 0, 0]. In case of SSPDs, extending the operation range is one of the main direction of this technology evolution. When photon energy decreases, detection efficiency of SSPD drops down to $\sim 1\%$ range and there was minor progress towards overcoming this problem [0, 0]. Partly, this

is due to the fact that this solutions should not significantly limit the speed of the detectors, which is a very valuable characteristic for practical implementation [0, 0]. On this basis, it is required to study the possibility of implementing superconducting ultra-thin niobium nitride films, that demonstrate superiority over amorphous WSi ultra-thin films from the point of speed, for fabrication of high-efficiency SSPDs aimed for mid-IR range [0, 0]. Detection efficiency, according to the model of SSPD operation, is mostly determined by the sheet resistance, transition temperature, and diffusion coefficient of the films [0, 0, 0, 0]. Consequently, research of the relationship between these parameters and detection efficiency of the detectors is possible via adjustment of the parameters via film deposition process. In the past, this approach already showed positive results, however, due to limitations of parameters variation it didn't demonstrate any major benefits [0, 0].

Besides the optimisation of already known materials for SSPD fabrication, it is important to research the new ones. One of such materials could be vanadium nitride, that has similar with NbN crystal structure and high transition temperature compared to WSi [0, 0]. Similar diffusion coefficient together with lower energy gap suggests a potential benefit for VN SSPDs compared to both NbN and WSi [0]. This hypothesis requires a focused research of achievable performance of VN SSPDs.

In conclusion, even though superconducting single-photon detectors have a potential to efficiently operate in the mid-IR range, currently this is only possible at a price of their speed reduction. Therefore, the **purpose** of the study is the research and development of single-photon detector the demonstrates high detection efficiency at the spectral range $0.7 - 3 \mu\text{m}$ together with high speed of operation. The purpose is achieved by consistently solving the following logically related tasks:

1. To study the relationship between SSPD detection efficiency and the level of disorder of initial superconducting thin films.
2. To research reactive magnetron sputtering process in the context of the relationships between deposition conditions (deposition time, substrate temperature, operating gas pressure, nitrogen content, magnetron current), films' parameters (sheet resistance, transition temperature), and SSPD characteristics (detection efficiency, counting rate, jitter).
3. To research possible causes for the reproducibility of the deposition process of NbN films obtained using reactive magnetron sputtering.

4. To obtain current dependencies of the detection efficiencies of SSPDs in the range $1.3 - 3 \mu\text{m}$ for the devices fabricated out of films with various sheet resistance values.
5. Obtaining ultra-thin superconducting films of niobium nitride that allow fabrication of superconducting single-photon detectors that demonstrate detection efficiency $\geq 80\%$ at $1.8 \mu\text{m}$ and $\geq 50\%$ at $2 - 3 \mu\text{m}$ while operating at 2.2 K.
6. To apply optimized fabrication root on ultra-thin vanadium nitride films in order to fabricate VN SSPDs.
7. To obtain detection efficiency values for SSPDs based on NbN and VN with various values of sheet resistance.

Scientific novelty of the research is in follows:

1. An approach, which differs from the well-known one by the implementation of disordered NbN films for increasing detection efficiency of SSPDs while maintaining detectors' speed.
2. For the first time a superconducting single-photon detector that simultaneously demonstrate 41% detection efficiency and 20 ns dead time, together with 59 ps temporal resolution is obtained.
3. For the first time a large statistic that includes more than 800 deposited NbN and 120 VN films under various conditions, and having various values of R_s and T_c (0.02 – 6.1 kOhm/square and 14 – 1.7 K correspondingly) is presented.
4. For the first time superconducting NbN films with $T_c = 7.8$ K and $R_s = 1.2$ kOhm/square are presented.
5. Detection efficiency *vs.* bias current dependencies are presented for VN and NbN films with various sheet resistance values. Systematic increase of the detection efficiency for increasing sheet resistance values is obtained.
6. An SSPD based on disordered NbN film with $R_s = 0.8$ kOhm/square ($T_c = 8.1$ K), integrated into optical resonator, achieve system detection efficiency of 94% at $1.3 \mu\text{m}$ and 2.2 K, for a strip width equal to 100 nm and bias current equal to $0.85I_c$. Temporal resolution reached 52 ps and dead time 11 ns.
7. An SSPD based on disordered NbN film with $R_s = 1.2$ kOhm/square ($T_c = 7.8$ K), integrated into optical resonator, achieve system detection efficiency of 92% at $1.5 \mu\text{m}$ and 2.2 K, for a strip width equal to 100 nm

and bias current equal to $0.85I_c$. Temporal resolution reached 59 ps. At 2.8 K internal detection efficiency of this device reached 74 and 41% at 2 and 3 μm correspondingly.

Scientific novelty of the research

The practical significance of the work consists in conducting a systematic study of the correlation between the deposition parameters and the properties of NbN and VN superconducting films, as well as the characteristics of detectors based on them; developing a reproducible route for obtaining ultrathin NbN films suitable for fabrication of highly efficient single-photo detectors for visible, near- and mid-IR ranges operating at a temperature of 2.2 K and preserving high-speed performance. Obtained results are actively used in the detection systems developed and implemented by "Superconducting Nanotechnology"(SCONTEL), which justifies the successful **implementation result**.

Provisions presented for defense

1. When depositing on silicon and silicon nitride substrates by reactive magnetron sputtering of ultrathin films of niobium nitride with a surface resistance of more than 200 Ohm /square, the highest values of critical transition temperatures to the superconducting state occur at substrate heating temperatures of 573 – 873 K, nitrogen concentration in the gas mixture of 20 – 22%, argon pressure of 3 mTorr and the film growth rate 0.7 Å/s.
2. An increase in the surface resistance of initial NbN films from 0.3 to 1.2 kOhm/square (with a concomitant change in the critical temperature from 10.7 to 7.8 K) leads to an increase in the internal quantum efficiency of SSPD from 18 to 97% at a wavelength of 1.5 μm at an operating temperature of 2.2 K, strip width 100 nm and detector bias current $0.9I_c$.
3. Increasing the surface resistance of the initial NbN films to 1.2 kOhm/square (at $T_c = 7.8$ K) contributes to an increase in the detection efficiency of SSPD in the mid-IR range, reaching 41% at a wavelength of 3 μm at an operating temperature of 2.8 K, the width of the strip 100 nm and detector bias current of $0.9I_c$. The calculated value of detection efficiency increases up to 90% when the detector is cooled down to a temperature of 2.2 K, provided by a closed-cycle refrigerators in the absence of direct background illumination.

4. SSPDs based on vanadium nitride films with surface resistance 0.17 kOhm/square and a critical temperature of 5.5 K do not achieve saturation of the internal detection efficiency at a wavelength of 1.3 μm at an operating temperature of 1.7 K, a strip width of 100 nm and detector bias current of $0.9I_c$ and thus are inferior in terms of detection efficiency to superconducting single-photon detectors based on niobium nitride films with a surface resistance of 0.7 kOhm/square and a critical temperature of 9.5 K under the same conditions.

The degree of reliability of the obtained results

The reliability of obtained results is ensured by the use of trusted experimental techniques used in the laboratory of quantum detectors in the Moscow State Pedagogical University and LLC SCONTEL; by the agreement with more recent data of similar studies conducted in other leading scientific centers, as well as by the creation of devices based on the proposed approaches and achieving high efficiency of the detectors.

Work approbation

The main results of the work were reported and discussed at the following scientific conferences:

1. "Micro-, nanotechnologies and their applications Chernogolovka, Moscow region, Russia, November 24-27, 2014. Report "Research of an SSPD receiver optimized for operation in the wavelength range from 700 to 1200 nm";
2. Third International Conference on Quantum Technologies (ICQT 2015), Moscow, Russia, July 13-17, 2015 The report "Capability investigation of superconductive single-photon detectors optimized for 800 – 1200 nm spectrum range»;
3. 6th International Conference on Photonics and Information Optics. Moscow, Russia, February 3-5, 2016 Report "Technology development creation of resonator structures to increase the quantum efficiency of NbN IR photon detectors";
4. Superconductors-based sensors and quantum technologies workshop, Moscow, Russia, April 18-21, 2016 Report "High-efficiency singlephoton detectors based on NbN films";
5. Micro-nanotechnologies and their application. Chernogolovka, Moscow region, Russia, February 6-10, 2017 Report "Superconducting IR detector

- with the ability to determine the number of photons in a short pulse of radiation";
6. 4th International School and Conference on Optoelectronics, Photonics, Engineering and Nanostructures "Saint Petersburg OPEN 2017 Saint Petersburg, Russia, 3-6 April, 2017 Report "Development of fast and high-effective single-photon detector for spectrum range up to 2.3 μm »;
 7. International Scientific Conference of Students, postgraduates and young scientists "Lomonosov-2017 Moscow, Russia, April 10-14, 2017 Report "Superconducting detector of single photons with an input optical radiation band up to 2.3 microns";
 8. Fourth International Conference on Quantum Technologies ICQT 2017 Moscow, Russia, July 12-17, 2017 The report "Photon-Number-Resolving SSPDs with system detection efficiency over 60
 9. VII International Conference on Photonics and Information Optics, Moscow, Russia, January 24-26, 2018. Report "Application of thin superconducting vanadium nitride films for the manufacture of single IR photon counters";
 10. 5th International School and Conference "Saint-Petersburg OPEN 2018, Saint Petersburg, Russia, April 2-5, 2018 Report "Influence of sputtering parameters on the main characteristics of ultra-thin vanadium nitride films";
 11. Annual Interuniversity Scientific and Technical Conference of students, graduate students and young specialists named after E.V.Armensky, Moscow, Russia, February 18-28, 2019 Report "Features of deposition of disordered ultrathin vanadium nitride films";
 12. Interaction between Radiation and Quantum matter IRQ 2019, Moscow, Russia, July 2-5, 2019 The report "Vanadium nitride – a promising material for superconducting nanodevices»;
 13. 3rd International Symposium on "Single Photon based Quantum Technologies", September 15-17, 2020, virtual. Report "Near-unity photon detection with ultra-low dark count rate at telecom c-band range";
 14. Applied Superconductivity Conference 2020, October 23 - November 7, 2020, virtual. Report "A comparison of VN and NbN thin film properties towards optimal SNSPD efficiency".

List of published articles reflecting the main scientific findings of the dissertation

The author's publications in peer-reviewed scientific journals included in the international citation system Scopus and WoS:

- A1. Korneeva, Y.P., Manova, N.N., Dryazgov, M.A., Simonov, N.O., **Zolotov, P.I.** and Korneev, A.A., 2021. Influence of sheet resistance and strip width on the detection efficiency saturation in micron-wide superconducting strips and large-area meanders. *Superconductor Science and Technology*, 34(8), p.084001.
- A2. **Zolotov, P.**, Semenov, A., Divochiy, A. and Goltsman, G., 2021. A Comparison of VN and NbN Thin Films Towards Optimal SNSPD Efficiency. *IEEE Transactions on Applied Superconductivity*, 31(5), pp.1-4.
- A3. Moshkova, M., Divochiy, A., Morozov, P., Vakhtomin, Y., Antipov, A., **Zolotov, P.**, Seleznev, V., Ahmetov, M. and Smirnov, K., 2019. High-performance superconducting photon-number-resolving detectors with 86% system efficiency at telecom range. *JOSA B*, 36(3), pp.B20-B25.
- A4. Divochiy, A., Misiaszek, M., Vakhtomin, Y., Morozov, P., Smirnov, K., **Zolotov, P.** and Kolenderski, P., 2018. Single photon detection system for visible and infrared spectrum range. *Optics letters*, 43(24), pp.6085-6088.
- A5. Smirnov, K., Divochiy, A., Vakhtomin, Y., Morozov, P., **Zolotov, P.**, Antipov, A. and Seleznev, V., 2018. NbN single-photon detectors with saturated dependence of quantum efficiency. *Superconductor Science and Technology*, 31(3), p.035011.

The author's personal contribution to the research

The content of the dissertation and the main provisions submitted for defense reflect the personal contribution of the author in the published works performed by the author personally or in collaboration with colleagues. The author's personal contribution included the formulation of tasks, deposition of the thin films and subsequent measurement of their main parameters, the adaptation of the process of reactive magnetron sputtering to obtain films with high values of surface resistances and critical temperatures, measurement of the spectral efficiency of fabricated detectors, participation in discussion and analysis the obtained data, as well as the preparation of scientific articles. All the results submitted for defense were obtained by the author personally or with his direct participation.

Contents

The dissertation consists of an introduction, 4 chapters, a conclusion and 3 appendices. The full volume of the dissertation is 115 pages, including 46 figures and 3 tables. The list of references contains 104 titles.

In the **introduction** the purpose and objectives of the dissertation research are formulated, its relevance and novelty are determined, the protected provisions are presented, the practical significance of the work is described.

The **first chapter** presents a review of the literature on superconducting single-photon detectors, describes theoretical and experimental work, devoted to the study of the effect of single-photon detection in superconducting nanostructures; the main methods of obtaining thin films are considered.

The **second chapter** presents thin film deposition setup and technological route for fabrication of superconducting single-photon detectors; describes experimental setup for their initial testing.

The **third chapter** is devoted to the study of the influence of various parameters of the reactive magnetron sputtering process on the parameters of the resulting films of niobium nitride. The chapter also describes theoretical and experimental investigation of the possibility of increasing the detection efficiency of SSPDs by using disordered films with different values of surface resistance and critical temperature.

The **fourth chapter** presents an experimental technique for studying the spectral detection efficiency of NbN SSPD. The maximum achievable values of the system quantum efficiency of manufactured detectors at telecommunication wavelengths are demonstrated. A study of detection efficiency at near-field wavelengths is described. A significant increase in the quantum efficiency of SSPD in the mid-IR range is shown when using initial films with a surface resistance of more than 1 kOhm/square.

In **conclusion**, the results of the work are presented and the conclusions of the study are made.

The **appendices** contain auxiliary data on the mathematical description of the process of reactive magnetron sputtering of NbN films, obtained on the basis of experimental data.

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