National Research University Higher School of Economics

As a manuscript

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Investigation of the critical temperature of the superconducting transition of thin aluminum films

Dissertation summary

For the purpose of obtaining academic degree

Doctor of Philosophy in Engineering

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Professor

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In Physics and Mathematics

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Relevance of the research topic

Behind each finished electronic product there is a large amount of scientific work to study the parameters and characteristics of individual components and elements that make up such a device. With the advent of superconducting electronics and nanoelectronics, as well as devices operating using quantum-size effects, a need arose to study the various characteristics of superconducting mesoscopic and nanostructures. In addition, the mechanisms that influence the key characteristics of such objects and the possibility of controlled production of superconducting objects with specified parameters are of great interest.

One of the most important parameters of a superconductor is its critical temperature Tc. But, unfortunately, most materials suitable for the role of electronic components have a very low transition temperature. For example, aluminum, which is considered the most promising superconductor for use as a qubit in a quantum computer, has a Tc equal to 1.19 K. Only the study of the superconducting properties of aluminum objects requires quite expensive refrigeration units. For example, fairly widespread and relatively inexpensive refrigerators of the CFMS type are cooled only to 1.6 K in the standard version, and obtaining lower temperatures requires either specialized inserts with third helium, which leads to their significant rise in price, or the presence of large and powerful cryogenic setups, price which is not comparable to the price of CFMS. The use of aluminum elements in superconducting electronics and nanoelectronics will require significant complication and increased cost of the finished device. Therefore, of particular interest is the possibility of increasing the Tc of superconducting low-dimensional structures made of aluminum.

State of the matter

The fact of a change in the critical temperature in quite small objects made of aluminum relative to a bulk material has been known for quite a long time [1], moreover, such a change is universal and manifests itself in all superconductors with a decrease in their size [2]. However, there is still debate about the reasons causing this change. Adding to the complexity is the fact that in some superconductors (aluminum, indium) Tc increases [3], while in others (niobium, vanadium) it decreases [4].

For a long time, one of the main and general reasons for the deviation of Tc of thin films from the value in bulk material has been considered the quality of the films being manufactured [5]. However, with improvements in manufacturing techniques, as well as in the quality of the thin films themselves, a shift in the critical temperature is still present in them. From a theoretical point of view, the answer to this question seems to lie on the surface: based on the BCS theory (1), a change in the critical temperature of a superconductor can be achieved either by modifying the phonon spectrum [6] or by changing the density of electronic states near the Fermi level [7].

$$T_c = 1.14\hbar\omega exp\left[-\frac{1}{N(0)V}\right], \tag{1}$$

where, $\hbar$ is Planck's constant, ω is a certain characteristic frequency of phonons, N (0) is the density of states of electrons on the Fermi surface in the normal state, and V is a parameter, the physical meaning of which is the characteristic binding energy between two electrons arising due to interaction with lattice vibrations (electron-phonon interaction constant). Over time, an assumption also appeared around the influence of the quantum limitation of charge carriers on the change in Tc [8]. However, despite the apparent simplicity of the theoretical interpretation, scientists have not been able to come to a general conclusion regarding the shift in critical temperature with decreasing object size. Moreover, if in theory three clear directions have been formed, then in practice, when producing thin films, even more factors arise that influence the final value of Tc. As a result, it becomes very difficult to determine the decisive contribution that has the maximum impact on the change in Tc. And the main problem is the fact that each of the emerging mechanisms quite strongly influences each other, which leads to their rather non-trivial interaction [9]. The most pronounced factors influencing the final value of the critical temperature of low-dimensional objects are the size of the granules that make up the object itself [10], mechanical defects [11], the presence of magnetic impurities [12], the proximity effect [13], and surface roughness [14], surface effects [15] and the size effect itself [16].

Based on such a wide range of different influences that influence the change in Tc in thin films, the main task of the researcher is to determine the decisive contribution to the change in Tc. Therefore, **the main goal of the dissertation work** is to establish the dependence of the critical temperature of a low-dimensional superconductor on its characteristic dimensions. To achieve this goal, it was necessary to decide to complete the following tasks:

1. Experimentally study the effect of film thickness on the change in the critical temperature of the superconductor.

2. Experimentally study the influence of various substrates on the change in the critical temperature of the superconductor.

3. Experimentally investigate the influence of the film production method on the change in the critical temperature of the superconductor.

4. Experimentally study the influence of the size of film granules on the change in the critical temperature of the superconductor.

5. Experimentally investigate the influence of the residual resistance of films on the change in the critical temperature of the superconductor.

6. Explore the possibility of describing experimental results in terms of the quantum size effect.

Scientific novelty is based on a systematic study of the dependence of the critical temperature of thin aluminum films on film thickness, film morphology and substrate material. In particular:

• For the first time, superconducting aluminum films were produced on sapphire substrates using molecular beam epitaxy and thermal sputtering.

• For the first time, the Tc of low-dimensional films of pure aluminum prepared on sapphire substrates was studied.

• For the first time, the Tc of low-dimensional films of pure aluminum, produced by molecular beam epitaxy, on gallium arsenide substrates was studied.

• For the first time, comparative experiments were carried out to observe the effect of changing the critical temperature on films of various qualities.

• It was shown for the first time that in higher quality epitaxial films of aluminum there is an effect of changing the critical temperature with film thickness.

• It was shown for the first time that, regardless of the substrate material (gallium arsenide or sapphire), there is an effect of changing the critical temperature with film thickness.

• A connection has been established between the quantum size effect and an increase in Tc in films with a decrease in their thickness.

Theoretical and practical significance of the work

The theoretical significance of the dissertation research lies in systematic measurements of the critical temperature of aluminum films depending on various factors and obtaining a large amount of data for the analysis of those mechanisms that can influence the shift of Tc in aluminum. A relationship has been established between changes in the critical temperature and the thickness of the films, substrate material, granule size, residual resistance of thin films, charge path length, and manufacturing quality of the objects under study.

The practical significance of the work lies in the development of a technique for producing high-quality aluminum films using molecular beam epitaxy on sapphire and gallium arsenide substrates. The experimental data obtained make it possible to highlight the influence of various factors affecting the Tc of a superconducting aluminum film during its manufacture, which makes it possible to produce film structures with a given superconducting transition temperature for use in various devices operating on superconducting elements.

Methodology and research methods

Aluminum was chosen as the material under study as the material showing the greatest increase in Tc relative to a volumetric object compared to other superconductors [17]. Superconducting film objects were produced by electron beam deposition and molecular beam epitaxy. Analysis of the fabricated structures was carried out using a high-resolution transmission electron microscope, as well as using a scanning transmission electron microscope using dark-field ring imaging.

Experimental studies of Tc were carried out on three cryogenic setups and with different thermometers. Sample resistance measurements were carried out using a four-point circuit at direct current. The current was applied to the samples in the range from 0.1 μA to 10 μA. Processing of experimental data was carried out using the Gnuplot program.

Main provisions presented for defense

1. An increase in the critical temperature of the superconducting transition Tc in aluminum films is a true size effect, independent of the morphology of the sample and/or substrate material.

2. An increase in the average granule size from 40 nm to 200 nm in superconducting aluminum films does not lead to a significant decrease in the superconducting transition temperature

3. The use of sapphire substrates leads to a noticeable decrease in the critical temperature of the superconducting transition in films with a thickness in the range from 15 to 45 nm.

4. The effect of increasing the critical temperature of the superconducting transition Tc in aluminum films can be qualitatively and quantitatively explained by the quantum size effect.

The degree of reliability of the obtained results

The reliability of the experimental data obtained is confirmed by quantitatively identical measurements obtained on three installations using three different thermometers. Reproducibility of results when re-manufacturing samples with specified parameters. There is the same trend towards an increase in Tc, with a decrease in the thickness of the object, in films, regardless of their quality and the method of their manufacture. As well as the agreement of the obtained data with existing works and with theoretical calculations. The quality of the films was controlled by three independent laboratories using various research techniques.

Author's personal contribution

The author's personal contribution is presented at all stages of the dissertation research, except for microanalysis of the structure of aluminum films, carried out by colleagues from the Aristotle University of Thessaloniki and colleagues from the National Research University MIET. The production, measurement and interpretation of the results obtained were carried out by the author of the dissertation or with his direct active participation.

Approbation of results

The results of the dissertation work were presented by the dissertation author at the following conferences:

1. Sedov E.A., Zavyalov V.V., Arutyunov K.Yu. Quantum size effect in semimetals and superconductors. XI International School-Conference of Students, Postgraduate Students and Young Scientists. Ufa, November 11-14, 2020

2. Sedov E.A., Zavyalov V.V., Arutyunov K.Yu. Quantum size effect in superconductors. Physics. Technologies. Innovation. Physicotechnical Institute-

2020. VII International Youth Scientific Conference dedicated to the 100th anniversary of the Ural Federal University, Yekaterinburg, May 18-22, 2020

3. Sedov E.A. Change in the critical temperature of superconducting aluminum films. Scientific and technical conference of students, graduate students and young specialists of the National Research University Higher School of Economics named after. E.V. Armensky. Moscow city. 2020

4. Sedov E.A., Zavyalov V.V., Arutyunov K.Yu. Quantum size effect in thin films of aluminum. Physics. Technologies. Innovation. Physicotechnical Institute-2019. VI International Youth Scientific Conference, Yekaterinburg, May 15-19, 2019

5. E. Sedov, I. Golokolenov, G. Konstantinidis, A. Stavrinidis, G. Stavrinidis, V. Zavialov, K. Arutyunov. The Enhancement of the Critical Temperature in Thin Aluminum Films. 15th Cryogenics 2019 / IIR International Conference / April 7 – 11, 2019 / Prague / Czech Republic.

6. Sedov E.A. Quantum size effect and aluminum films. Scientific and technical conference of students, graduate students and young specialists of the National Research University Higher School of Economics named after. E.V. Armensky. Moscow city. 2019

7. Sedov E.A. Increase in Tc of a superconductor in aluminum films. Scientific and technical conference of students, graduate students and young specialists of the National Research University Higher School of Economics named after. E.V. Armensky. Moscow city. 2018

List of publications on the topic of the thesis:

- Sedov E. A., Zavyalov V. V., Arutyunov K. Yu. Quantum size effect in superconductors // Bulletin of the Ufa Scientific Center of the Russian Academy of Sciences. 2021.1, 39 - 43.

- Arutyunov K., Zavialov Vitalii, Sedov E., Golokolenov I., Заруднева А. А., Шеин К. В., Trun’kin I. N., Vasiliev A. L., Konstantinidis G., Stavrinidis G., Croitoru M. D., Shanenko A. A. Nanoarchitecture: Toward Quantum-Size Tuning of Superconductivity. // Physica Status Solidi - Rapid Research Letters. 2019, 1800317, 1-5.
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- Sedov E., Riikonen K., Arutyunov K. Quantum size effect in single-crystalline bismuth nanorods // Journal of Physics: Conference Series. 2017. 929. 012088. P. 1-6.
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Scope and structure of the dissertation

The dissertation consists of an introduction, 5 chapters and a conclusion. The full volume of the dissertation is 124 pages, including 4 tables and 56 figures. The bibliography includes 216 sources.

The introduction presents the relevance of the research, formulates the goal and defines the objectives of the dissertation work, presents the provisions to be defended, and describes the practical and theoretical significance of the work.

The first chapter describes existing approaches to solving the problem of changing the critical temperature of superconducting films relative to reducing their thickness.

The second chapter provides a description of the method for manufacturing superconducting objects studied in this work.

The third chapter presents the results of crystallographic and microstructural analysis, as well as the results of studying the surface of the samples.

The fourth chapter concentrates on the results of experimental measurements and analyzes and identifies patterns in the dependence of Tc of aluminum films depending on various factors.

The fifth chapter presents the discussion of results and comparing them with theoretical models.

In conclusion presented the results of the work.

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