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Morphology and electrical parameters of thin aluminum films deposited on substrates at temperatures from 77 to 800 K

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Experimental studies of the basic parameters of 150 nm aluminum films on Si(111), SiO_2/Si(001) substrates have been performed. The films were obtained by magnetron sputtering and thermal evaporation in the temperature range from 77 to 800 K. A vacuum insert was made for the Z400 setup to cool the substrate to liquid nitrogen temperature, and a standard heater of the Kurt Lesker setup was used to heat it to 800 K. It was found that cryogenic deposition of aluminum adatoms allows decreasing the size of the formed grains from 280 to 20 nm and the roughness from 4.2 to 1.7 nm. The specific resistance of the films and the superconducting transition temperature T_c increase from 27 to 260 Ω·nm and from 1.2 to 2.3 K, respectively. This is associated with an increase in the number of intergrain boundaries in cryogenic Al- films and can lead to an increase in their kinetic inductance by 20 times or more.

Keywords: thin films, aluminum, resistivity, superconducting transition temperature, SEM, AFM.

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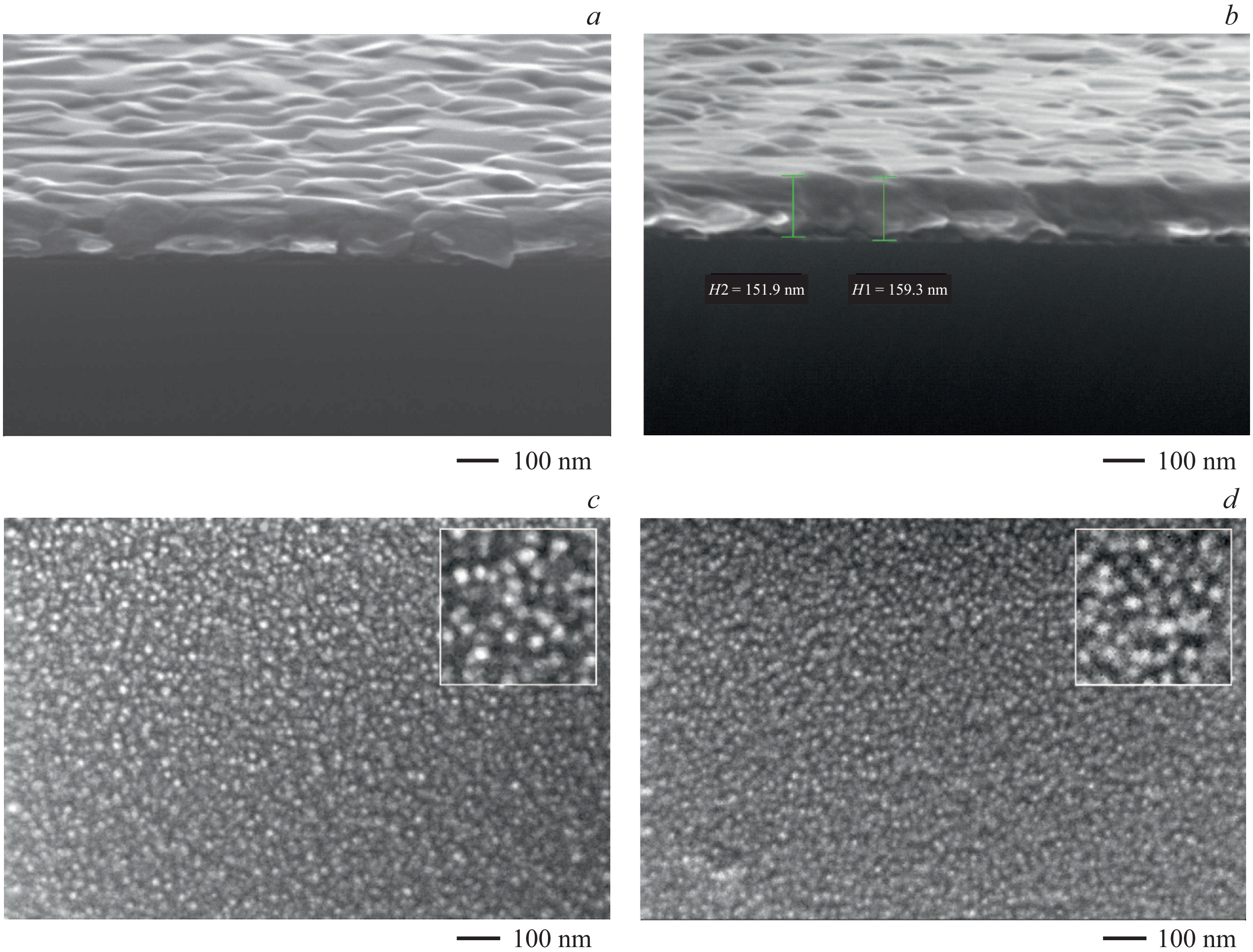
Thin aluminum-based films are widely used in various applications: from anti-corrosion coatings, micro- and optoelectronics to quantum computer components. Special interest to them is prompted by the need to create compact superconducting devices based on tunnel junctions. Increasing the critical transition temperature of metallic films to the superconducting state is one of the priority tasks in the development of growth techniques. It is known that a number of methods can be used to achieve this goal in the formation of aluminum films: thermal evaporation in an oxygen atmosphere, sputtering with silicon doping, magnetron sputtering in a nitrogen atmosphere. An important electrophysical parameter of thin films is their kinetic inductance, which depends on their microstructure. In our study [1] the possibility of controlling the microstructure of thin aluminum films by forming seed homobuffer layers on the surface of a silicon substrate was shown. In this paper we present the results of measuring the electrical parameters and surface morphology of aluminum films with a total thickness of 150 nm. Samples of M-series films grown by magnetron sputtering on Si(111) substrates at 300 K had seed homobuffer layers with a thickness of 20−40 nm formed at temperatures from 300 to 800 K similarly to [1]. Series E aluminum films were formed by thermal evaporation on Si(111) and oxidized silicon SiO_2/Si(001) substrates at 77 and 300 K. Electron microscope images of aluminum film chip crack are shown in the figure, and their characteristic dimensions are presented in the table.

The study of the relationship between the resistivity of thin films and their morphology and structure has been going on for decades [2–11]. In the earliest Thomson [3] model, a geometric model was considered for electrons reflecting off two film surfaces of thickness t at a constant mean free path length λ. The later Fuchs-Sondheimer [4,5] model already took into account the statistical distribution of λ and the morphology of the film surface. The modern Mayadas–Shatzkes [6,7] model added the transparency of grain boundaries and their average grain size. An alternative phenomenological Andrews model [9], according to which the resistivity is inversely proportional to the average grain diameter and the constant of proportionality is called the Andrews parameter, is discussed in a detailed overview on the contribution of grain boundaries in metal [8].

In the Mayadas-Schatzkes model, the reflection coefficient from the intergrain boundary $R$is used. The inverse resistivity dependence

$$\rho_0/\rho_{ms} = 1 - 1.5\alpha + 3\alpha^2,$$

where ρ_0 — resistivity of solid aluminum, ρ_{ms} — resistivity according to the Mayadas–Shatzkes model, $\alpha = (l_0/d)(R/(1-R))$, l_0 — electron mean free path length, d — grain size in fine-grained film. For aluminum, the reflection coefficient is generally considered to be $R = 0.7-0.9$ and depends on orientation [10]. The influence of the intrinsic size effect due to the polycrystalline structure of metal films on the reflection, transmission,



SEM images of cracked aluminum films after magnetron sputtering (*a, b*) and thermal evaporation (*c, d*). *a* — M-300/300/Si(111), *b* — M-700/300/Si(111), *c* — E-77/Si(111), *d* — E-300/Si(111).

Surface morphology parameters and electrical resistivity values of aluminum films grown by magnetron sputtering (M) and thermal evaporation (E)

Designation of sample	Roughness RMS, nm	Size of the grain, nm	Resistivity, $\Omega\cdot$nm		Critical temperature T_c, K
			at 300 K	at 77 K	
M-800/300/Si(111)	4.2 ± 0.5	170	33	12	
M-700/300/Si(111)	4.1 ± 0.5	160	27	9	
M-600/300/Si(111)	4.7 ± 0.5	150	45	10	
M-500/300/Si(111)	5.4 ± 0.5	230	45	10	1.22 ± 0.05
M-400/300/Si(111)	5.0 ± 0.5	250	45	10	
M-300/300/Si(111)	4.3 ± 0.5	280	37	9	1.20 ± 0.05
E-77/Si(111)	1.7 ± 0.3	15	96	74	2.30 ± 0.05
E-77/SiO_2	2.6 ± 0.3	45	260	230	2.20 ± 0.05
E-300/Si(111)	1.5 ± 0.3	20	96	63	1.25 ± 0.05
E-300/SiO_2	1.3 ± 0.3	54	220	180	

Note. In sample designations, numbers indicate deposition temperatures (in K): for M-series samples, the first number corresponds to the temperature of forming seed homobuffer layers, the second number — the substrate temperature during sputtering of the main film, for E-series samples, the substrate temperature during film evaporation is given.

and absorption coefficients of electromagnetic waves was studied in [11].

The surface morphology of the film samples was investigated by scanning electron microscopy (SEM) and atomic force microscopy (AFM). The averaged values of grain sizes according to the applied measurement techniques and resistances per square of the 150 nm thick films are given in the table. Temperature dependences of resistivity and values of the critical temperature of the superconducting transition were measured in a closed-cycle cryostat HELIOX-AC-V (Oxford Instruments) of the sorption type on ^{3}He with a minimum temperature of 280 mK.

The table shows that the largest grain $(250 \pm 30\,\text{nm})$ and avearage grain roughness $(\text{RMS} = 5.0 \pm 0.5\,\text{nm})$ M-series films were obtained by sputtering on a homobuffer layer formed at substrate temperatures of 300−500 K. The smoothest E-series films with grain size 20 ± 5 nm and $\text{RMS} = 1.7 \pm 0.3$ nm were obtained by evaporation on a liquid nitrogen-cooled Si(111) substrate. The highest superconducting transition temperature was obtained for the smallest-grain E-77/Si(111) film. A similar increase in the critical temperature of aluminum was obtained in [12] evaporation in an oxygen atmosphere and in co-evaporation with addition of silicon. It is known that the properties of Al films are strongly affected by the presence of oxygen and carbon impurities in them. It should be noted that films with higher resistivity will have higher kinetic inductance, which can be described by a simple formula for kinetic inductance per square of film

$$L_k = \hbar R_n/(\pi \Delta_0),$$

where R_n — resistivity per square of film, Δ_0 — energy gap. In this case, a significant increase in the kinetic inductance L_k can be obtained without the need to form very thin films (less than 5 nm). In our case, an increase in resistance by almost an order of magnitude and T_c by a factor of 2 would be equivalent to a 20-fold decrease in thickness, i.e., a 100 nm film would have the same kinetic inductance as a 5 nm film. The trend of increasing disorder in the amorphous film can be continued up to the critical temperature of 5 K, but further increase will lead to a decrease in T_c for very disordered films [13–15]. Another important property of fine-grained films may be the reduction of the inelastic relaxation length, which affects the cooling efficiency of the superconductor and improves the performance of electron coolers based on the superconductor-insulator-normal-metal [16] tunnel junctions.

The studied aluminum films evaporated on liquid nitrogen-cooled substrates can be used to create more compact superconductor microwave resonant cavities, highly sensitive detectors based on superconductor-insulator-superconductor and superconductor-insulator-normal metal-insulator-superconductor tunnel junctions with a wider temperature range and dynamic range, as well as more efficient electronic coolers based on superconductor-insulator-normal metal tunnel junctions.

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Conflict of interest

The authors declare that they have no conflict of interest.

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