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Thin aluminum films deposited on liquid nitrogen cooled substrates

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The microstructure of aluminum films thermally deposited on monocrystalline silicon substrates at liquid nitrogen temperature and at room temperature has been studied. The results of the study of the morphology and microstructure of the film by X-ray diffraction, atomic force and electron microscopy are presented. It is shown that the RMS roughness decreases from 0.4−1.2 to 0.19−0.34 nm when the substrate is cooled from room temperature to liquid nitrogen temperature.

Keywords: thin films, surface morphology, film structure, superconductivity, SIS junction, crystal structure.

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1. Introduction

The problem of quality of aluminum superconducting tunnel junctions is caused by the film surface morphology [1]. A problem arises to develop available film growth methods to control the morphology of their surface. Method of molecular-beam epitaxy (MBE) allows providing the growth of aluminum epitaxial films with atomically smooth surface, however this technology is poorly applicable for the mass production. There are two alternative approaches to dealing with the problem of atomically smooth films: sputtering by method of quasi-epitaxy films with substrate temperatures $T \geq 0.5T_{\text{mp}}$ (T_{mp} — melting temperature of deposited metal), as in [2], or film sputtering on the cooled substrate as in [3].

This paper outlines the studies of the microstructure and morphology of aluminum films deposited on the silicon substrates cooled with liquid nitrogen.

2. Experiment

Single-crystal Al films were obtained by the method of thermal evaporation using Leybold Heraeus Z-400 device. Conventional Si(111) substrates with a four-degree disorientation were used for evaporation on the single-crystal substrates. The films grew at a room temperature of 293 K and liquid nitrogen temperature of 77 K. To prevent moisture deposition on the substrate under cryogenic temperatures a nitrogen trap was placed in the chamber. The cooling effect was provided due to passing the liquid nitrogen through a tube where the substrate holder was soldered to. The holder started to be cooled a minute before the film sputtering initiated and continued during the film growth process. The film was formed with a rate of 0.4−0.6 nm/s. Designation of samples and conditions of growth of Al films are given in Table 1.

The morphology of the surface, homogeneity and thickness of aluminum films were studied by SEM method using electronic microscope Carl Zeiss Ultra 55. Composition of films was controlled using an X-ray energy-dispersive spectrometer (EDS) — Oxford Instrument INCA X-act. The energy of electronic beam was 5 and 20 keV in the modes of microanalysis and chip surface images, respectively.

Phase analysis (XRD) of Al/Si(111) film samples was carried out using an X-ray diffractometer SmartLab (Rigaku, Japan) with 9 kW rotating Cu-anode. Diffraction patterns in the scanning system $2\theta/\omega$ and 2θ were recorded using a parallel X-ray beam with a 0.00087 rad divergence. The films thickness and density profile were analyzed using X-ray reflectometry method (XRR). For registration of XRR curves a double slit monochromator Ge(220) was additionally installed on the diffractometer. XRR curves were simulated in GlobalFit (Rigaku) program.

For checking the produced seed layers and final films growth we used an atomic-force microscopy (AFM). AFM-measurements were carried out using Dimension Icon (Bruker) microscope fitted with commercial cantilevers RTESPA-300, in QNM mode, in the air. The cantilever tip is made of silicone with a nominal radius of 8 nm, resonance frequency 300 kHz and pre-calibrated (Bruker) hardness of 40 N/m. The scanning frequency was 1 Hz, while specified peak force was ~ 3 nN. Straight linear scanning was used. Scanning area was $1 \times 1\,\mu\text{m}^2$. AFM images were processed in Gwyddion software. Filtering

Table 1. Conditions of Al films thermal sputtering on Si(111)

Number of sample	Substrate	Temperature of substrate, K	Rate of of evaporation, nm/s	Pressure of evaporation, Torr
TA8-10	Si(111)	77–100	0.6	0.54 × 1E-5
TA8-11	Si(111)	293	0.46	0.53 × 1E-5

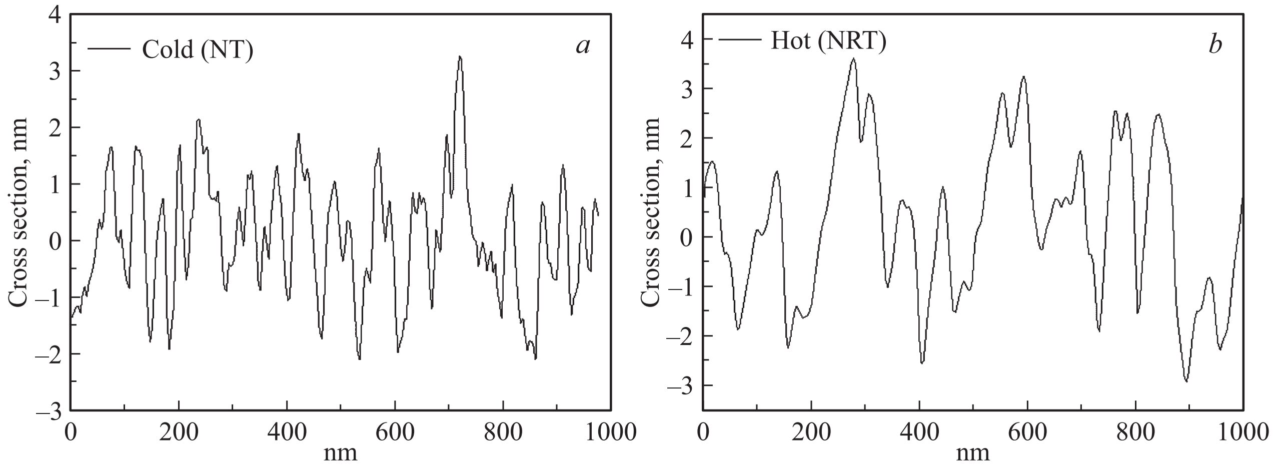

Figure 1. AFM-profiles of surfaces of Al/Si(111) films evaporated on the nitrogen cooled substrate — NT (*a*) and without cooling — RT (*b*).

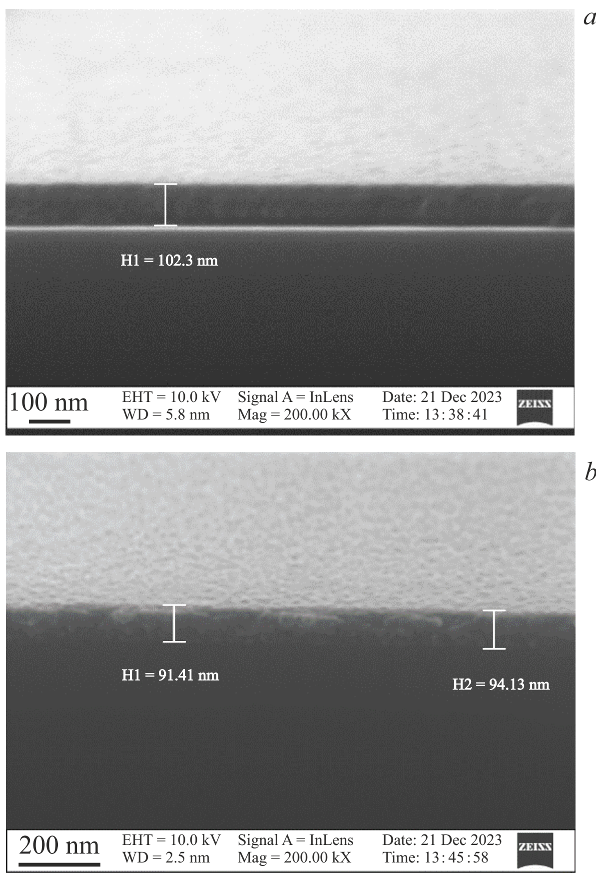

Figure 2. Images of the film chipping surfaces for the films evaporated at 77 K (*a*) and 293 K (*b*).

operations included subtraction of the 3d order surfaces, removal of steps in X direction, and removal of scratches.

Rms parameter was used to assess the surface roughness for the entire scan area. An autocorrelation function was used to characterize the lateral size of surfaces inhomogeneities in quantity. The function was plotted based on processing of films surfaces images using wsxm.eu (WSxM v5.0 Develop 10.2) program.

The films microstructure and morphology of their surfaces were studied using X-ray reflectometry, diffractometry, scanning electronic and atomic power microscopy methods. The results of experiments demonstrate that 100 nm thick aluminum films evaporated on a cooled surface compared to films evaporated at room temperature are featuring lower roughness. The most smooth film was obtained on a cooled silicon substrate with $\langle 111 \rangle$ orientation. Such films are designed for manufacture of SIS and SIN tunnel junctions. The primary analysis of surface was carried out using Atomic Force Microscopy (AFM). Cross-sections of films evaporated at various substrate temperatures are given in Figure 1.

Table 2 gives averaged rms roughness values σ_{rms} and maximal σ_{max} for various scans and samples for 100 nm thick films.

Electron microscopy of the films edges is given in Figure 2. Here, we can see that the surface of film evaporated on cold substrate is more smooth.

Figure 3 shows X-ray diffraction patterns of Al films in geometry GIXRD (*a*) and XRD (*b*). According to diffraction pattern analysis we may conclude that RT and

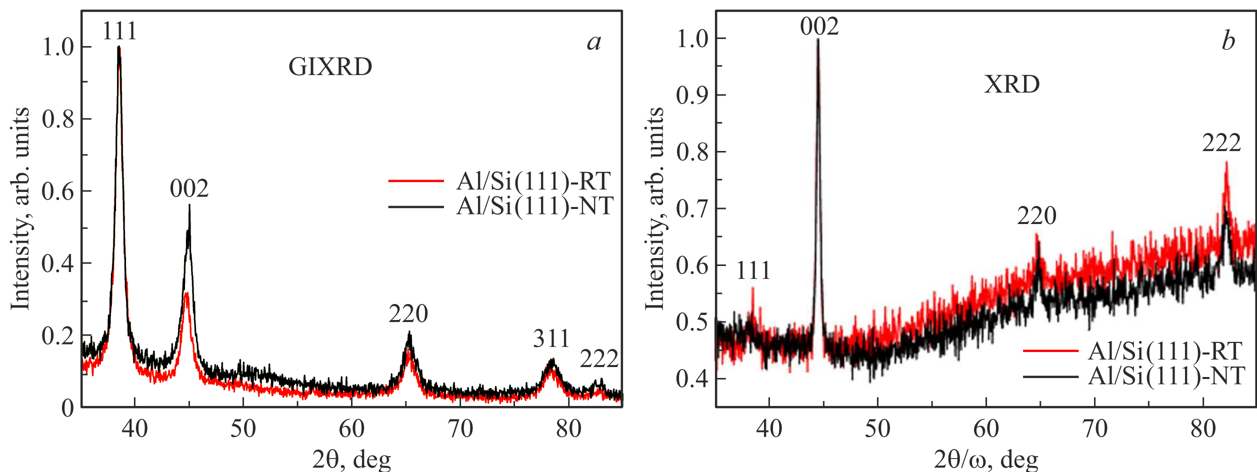

Figure 3. Diffraction patterns of Al films evaporated on Si(111) substrate (*a, b*) and on SiO_2/Si(001), in XRD geometry (*a*) and — GIXRD at grazing angle $\varphi = 0.5^\circ$) (*b*).

Table 2. Film surface roughness versus substrate temperature during sputtering

Temperature	σ_{rms} (nm)	σ_{max} (nm)
77 K	0.19–0.34	0.8–2.5
293 K	0.42–1.2	1.5–4.7

NT aluminum films on Si(111) are polycrystalline. The films are featuring a clearly distinct texture (001), only one reflection is actually observed 002 (Figure 3, *b*). The half-widths of corresponding diffraction peaks (crystallite size) for the studied films are practically equal. The recorded diffraction patterns greatly differ from diffraction patterns for Al films with texture (111) obtained by magnetron sputtering. It shall be emphasized that our films mainly contain crystallites 001 with their surface parallel to the surface of Si(111) substrates. Large amount of Al{001} crystallites correlates with the lower surface roughness, e.g. up to rms ~ 0.25 nm for Al (NT = 77°K) films.

A lot of small „horns“ (Figure 1, *b*) are observed on coarse particles– these „horns“ are individual particles with sizes of 15–20 nm. Coarse particles are 50 nm in size. In NT-films the particles average size is ~ 10–15 nm. It shall be emphasized that in AFM this image represents only a 5 nm thick surface layer. Diffraction takes place only on individual crystallites since we can neglect the effect of particles size in our case. Crystallites have lateral diffraction sizes of 9–11 nm which is a good correlation with AFM data. In our case the sizes of crystallites are largely defined by the energy of deposited atoms rather than the substrate temperature. While the substrate being at room temperature adds energy to the aluminum atoms contributing to the crystallites coagulation and transforming into larger size particles (pieces). This, apparently, leads to higher roughness of RT-films.

3. Conclusion

Due to evaporation of aluminum films on the single-crystal nitrogen-cooled Si(111) substrates the film roughness can be reduced more than twice compared to the film evaporated at room temperature; the surface roughness may reach even rms ~ 0.25 nm.

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Conflict of interest

The authors declare that they have no conflict of interest.

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