Nb₃Al thin film deposition for low-noise terahertz electronics

D Dochev, A B Pavolotsky, V Belitsky and H Olofsson

Group for Advanced Receiver Development and Onsala Space Observatory, Department of Radio- and Space Science, Chalmers University of Technology, SE 412 96 Gothenburg, Sweden

E-mail: dimitar.dochev@chalmers.se

Abstract. Higher energy gap superconducting materials were always interesting for low-noise mixer applications such as superconductor-insulator-superconductor tunnel junctions (SIS) and hot-electron bolometer (HEB) used in sub-millimeter and terahertz parts of electro-magnetic spectrum. Here, we report a novel approach for producing Nb₃Al thin film by co-sputtering from two confocally arranged Nb and Al dc-magnetrons onto substrate heated up to 830°C. Characterization of the deposited films revealed presence of the A15 phase and measured critical temperature was up to 15.7 K with the transition width 0.2-0.3 K for a 300 nm thick film. We measured the film critical magnetic field and studied influence of annealing on the film properties. We have investigated compositional depth profile of the deposited films by spectroscopy of reflected electrons.

1. Introduction

Nb material dominating Superconductor-Insulator-Superconductor (SIS) tunnel junction mixer technology, widely used in radio astronomy receiver applications, constrains the upper frequency to about 1250 GHz due to resistive losses above the gap frequency. Use of a high-gap material eliminates the loss immediately after the gap frequency and extends the frequency band of SIS mixers, with Nb₂Al films expected 30% up, as compared to currently used Nb and even further improved for NbTiN [1] material.

The compound superconductor Nb₃Al, which has the β-W structure (A15) [2], has critical temperature \( T_c \) of about 18.8 K for bulk material [3]. Ever since its discovery, the material has received substantial attention due to its possibility to carry very large current densities and its high energy gap. Consequently, Nb₃Al material was considered as a candidate for superconducting tunnel junctions and even hot-electron bolometers (HEB).

However, absence of established Nb₃Al thin film deposition technique required for SIS or HEB mixer fabrication hinders practical applications. Certain requirements in terms of film thickness and substrate choice have to be considered prior to fabrication of SIS and HEB structures. Deposition of too thick superconducting films in SIS structure is unwanted due to possible developing of high surface roughness and thus complications in the insulation interlayer fabrication. Substrates with relatively low dielectric constant and low dielectric losses, such as crystal quartz, are usually preferred for THz devices whereas, to our knowledge, the most frequently used substrates for Nb₃Al deposition are oxidized silicon, (110) and (0001) sapphire.

There are several approaches to deposit Nb₃Al thin films. The most common technique for depositing this superconducting material is by magnetron sputtering, DC [4] or RF [5], from a
stoichiometric target. Disadvantage of this method is sensitivity of the deposited film critical temperature to argon pressure together with necessity of using a high dielectric constant substrate, e.g., $(1\overline{1}02)$ sapphire. Resulting film thickness $\geq 0.4$ µm are needed to provide good film quality with $T_c$ of 17.7 K [6]. Nb$_3$Al thin films can be synthesized from sputter-deposited or evaporated Nb/Al multilayers [7], followed by annealing diffusion reaction process. Using the latter technique it was possible to reach critical temperature of about 16.2 K for a 0.4 µm thick film deposited on a sapphire substrate. Producing Nb$_3$Al films has also been done by means of e-beam co-evaporation [8] by employing two separate sources, Nb and Al targets. Even though, the reported transition temperature was on the onset of 16.7 K, the film was 0.5 µm thick and deposited on sapphire. Interestingly, most of the sited above attempts, including current work, resulted in Nb$_3$Al films with $T_c$ comparable or higher than that of the best NbTiN films.

Co-sputtering technique is a well established method for composite materials’ deposition. However, to our knowledge no other work on dc magnetron co-sputtered Nb$_3$Al has been reported. Nevertheless, there were attempts to synthesize Nb$_3$Ge by dc co-sputtering from a single cathode [9] as well as Nb$_3$Si by dual-target magnetron sputtering [10]. In this paper, we report on thin Nb$_3$Al film deposition technique by co-sputtering from two confocally arranged Nb and Al dc-magnetrons, thus avoiding the need in costly alloy target. The results of further study of the deposited film properties are presented in terms of material, magnetic and electrical characterization and involve X-ray diffraction analysis (XRD), spectroscopy of reflected electrons (SRE) [11], critical magnetic fields and resistance vs. temperature measurements.

2. Experiment

2.1. Film Deposition

Nb$_3$Al films were deposited by dc magnetron co-sputtering from two confocally arranged targets, 99.95% Nb and 99.99% Al in a high-vacuum sputtering system. The substrate was rotated during deposition to ensure the uniformity of the films. In the present experiments, we used oxidized silicon wafers coated with a 150 nm buffer layer of dc magnetron sputtered AlN in order to avoid interaction with the substrate material via diffusion for the duration of the high temperature deposition of Nb$_3$Al. During the deposition, the substrate temperature was set in the range 700 - 830°C. The ultimate pressure in the chamber was typically about $2 \times 10^{-7}$ Torr, achieved by turbomolecular pump. The deposition rate of Nb was kept at the level of about 120 Å/min, while Al was varied between 17 and 39 Å/min whereas keeping the final film thickness constant around 300 nm. The atomic concentration of Al was estimated based on the ratios of Nb and Al deposition rates and their atomic volumes resulted in Al content in the deposited films within the range between 12 at% Al to 24 at% Al.

The films were exposed to annealing carried out after deposition in rapid thermal processing (RTP) oven in argon atmosphere with the temperature of 1000°C achieved in ~20 s. After 30 s seconds of annealing the temperature decreased to 700°C in less than 45 s under Ar flow followed by a constant nitrogen flow supplied during the cooling from 300°C to room temperature.

2.2. Film characterization

The crystal structure and lattice constants of the films were determined by X-ray diffractometry (XRD) using Ni filtered CuKα radiation (40 mA, 40 kV). The superconducting transition temperature ($T_c$), defined as the middle of the superconducting transition, was measured using a four-probe resistance measurement technique and performed in a He dewar using a dip-stick and in a close-cycle 12 K cryo-cooler, both with an accuracy of $\pm 0.1$ K using calibrated temperature sensors from Lakeshore. Spectroscopy of reflected electron (SRE), with experimental details described in [11], was used to identify the film compositional depth-profile. Low magnetic field measurements (near $T_c$) were conducted using a 5 T Nb-Ti superconducting magnet.
3. Results and discussion
The \( T_c \) dependence vs. the substrate temperature during the film deposition was studied in the range 700º-830ºC for both annealed and as-deposited films. No significant improvement has been found above 750ºC, in agreement with Tanabe [4, 6]. We have observed that magnetron sputtered AlN buffer layer enhances the critical temperature of the as-deposited films up to about 1 K as compared to using of a bare oxidized Si-wafer. The as-deposited samples exhibited depressed \( T_c \) achieving maximum \( T_c \) of \( \sim 12 \) K for 16 at % Al for 300 nm thick film, Figure 1. The residual resistance ratio (RRR) for these films indicated values lower than 1.1. The X-ray patterns showed very broad and weak peaks. This is a clear indication that the as-deposited sputtered films have strongly disordered structure. SRE measurements point towards a strong segregation of Al from the film surface side and Al depletion from the substrate side even though the averaged Al concentration across the film depth was as expected, see Figure 2. We believe, this effect of the elemental redistribution is due to the problems of nucleation on the chosen substrate.

![Figure 1. Superconducting transition temperature vs. alloy composition for as-deposited and annealed samples.](image1)

![Figure 2. Depth profiles of Nb3Al/AlN/SiO2/Si multilayered samples with 12 at % Al (dashed) and 24 at % Al (solid).](image2)

For further improvement of the film properties, in terms of making a film with less disordered internal structure, a rapid heating to temperatures up to 1000ºC was applied. It was reported in [12] that the transformation from bcc to A15 phase will occur at fast heating as soon as the temperature reaches 900-1000ºC and this short time heat treatment should be followed by a rapid cooling to retain the bcc phase in metastable state. The rapid thermal annealing (RTA) resulted in films with significantly increased \( T_c \) having the maximum value of 15.7 K for sample with 24 at % Al, Figure 3. It can be seen from Figure 1 that for small amounts of aluminum the transition temperature remained nearly the same and then increased rapidly and finally leveled off over the range of aluminum content 20 to 24 %, similar to reported in [13]. We also observed that annealing of Nb3Al thin films results in increased lattice parameter together with growth of \( \sigma \)-phase, which is in agreement with Khan [14]. Still, X-ray diffraction patterns possessed dispersed X-ray pattern with broad peaks.
Figure 3. Superconducting transition of 24 at % Al Nb-Al film after annealing. The thickness of the film is approximately 300 nm.

In order to improve nucleation conditions for Nb$_3$Al at the substrate surface, we deposited first a thin Nb film (dc-magnetron sputtered at 10 Å/s), which is known from conducted XRD measurements to have well ordered and textured (110) columnar structure, where dimensions of Nb (110) \( \sqrt{\frac{11}{2}} \times \sqrt{\frac{6}{2}} \) \( R \arccos\left(\frac{\sqrt{6}}{2}\right) \) are close to that of (210) Nb$_3$Al. Nb$_3$Al film grown on such seed layer demonstrates strong and relatively narrow XRD peak corresponding to (210) plane, Figure 4. This fact together with higher RRR value (~20 % higher than for the film without seed layer) points out to a higher crystallographic order of the film.

Figure 4. X-ray diffraction patterns of as-deposited (solid) and annealed (dashed) Nb-Al film with 100 nm thick Nb seed layer. The thickness of the Nb$_3$Al film is approximately 300 nm. The \( T_c \) values of as-deposited and annealed films were 9 K and 15.3 K respectively.

We estimate magnetic field penetration depth for our Nb$_3$Al films out of the measured values of their critical temperature \( T_c \) and specific resistance before transition \( \rho_0 \). Reference [15] gives expression for penetration depth at 0 K, in for weak-coupled BCS superconductor; in dirty limit \( \lambda_{GL}(0)[cm] = 6.42 \times 10^{-6} \left( \frac{\rho_0(\mu\Omega\cdot cm)}{T_c(K)} \right)^{1/2} \). For measured values of \( T_c = 15.3 \) K and specific resistance \( \rho_0 = 163 \mu\Omega\cdot cm \), we get the estimation of penetration depth for our Nb$_3$Al film, \( \lambda_{GL}(0) \approx 210 \) nm.

We recorded \( H_{c2} \) dependence on the temperature, by varying temperature at fixed value of magnetic field and measuring resistance of the film biased by current much smaller than critical one at the experiment conditions (Figure 5a). Extrapolating this data to \( T = 0 \) K, we obtain the value \( H_{c2}(0) = 17.8 \) T, from which we estimate coherence length \( \xi_0 = 4.3 \) nm given by the expression [16]
\[ \xi_0 = \left( \frac{\Phi_0}{2\pi H_c(0)} \right)^{1/2}, \]

where \( \Phi_0 = \hbar c/2e \) is the magnetic flux quantum. From the slope of the critical field temperature dependence near \( T_c \) (Figure 5b): \( (dH_c/dT)_{T_c} = -2.38 \text{ T K}^{-1} \) we estimate electron diffusion coefficient \( D \approx 0.5 \text{ cm}^2\cdot\text{s}^{-1} \) following the expression

\[ D \approx \frac{1}{\gamma} \frac{\pi}{\xi_0^2} \]

[17].

**Figure 5.** Temperature dependence of the critical field measured for annealed 300 nm thick Nb\(_3\)Al thin film with 22 at \% Al, deposited on 100 nm thick Nb seed layer. The sample orientation was kept parallel to the magnetic field.

### 4. Conclusions

We presented results of development of Nb\(_3\)Al thin film deposition via DC-magnetron co-sputtering from pure Nb and Al targets. The reported studies’ goal was to obtain thin Nb\(_3\)Al films, up to 300 nm, for use in Superconductor-Insulator-Superconductor (SIS) tunnel junction as well as in hot-electron bolometers (HEB). The deposited films have measured critical temperature up to 15.7 K with transition width of 0.2-0.3 K after rapid thermal annealing. Relatively low measured critical temperature in as-deposited Nb\(_3\)Al films is attributed to film nucleation conditions including substrate interface and crystallographic order of the film. Spectroscopy of reflected electrons measurements of as-deposited films indicated the presence of non-homogeneous aluminum and niobium composition across the deposited film thickness with depletion of the aluminum around the substrate-to-film interface. The estimated values of magnetic field penetration depth, coherence length and electron diffusion constants were close to the values, reported earlier for NbN and NbTiN films.

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