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The role of the substrate temperature on superconducting properties of sputtered Nb films

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ABSTRACT

The influence of the substrate temperature T_S on the superconducting properties of 100 nm sputtered Nb films, prepared directly onto orientated Si (100) substrate, was systematically investigated by measuring their structural, morphological and magnetic properties. Within the T_S interval 293–373 K no significant change is observed either in the Nb lattice parameter or in the superconducting transition temperature T_C. For $T_S > 373$ K, a degradation of the Nb superconducting properties was observed concomitantly with an increase of the Nb lattice parameter. This effect was attributed to an interdiffusion at the Si/Nb interface and/or an enhancement of the internal stress caused by high temperature deposition. The temperature dependence of the critical current density $J_C(T)$ was estimated from magnetization measurements and its behavior is explained based on the granular morphology of Nb films. This work provides some insights on the optimum T_S responsible for the highest J_C value. It also brings information on how T_S affect the superconducting properties of Nb films sputtered directly on Si (100) by DC magnetron sputtering.

1. Introduction

The production of niobium (Nb) superconducting thin films is important both from the fundamental viewpoint and for the development of superconducting-based technological devices, i.e., nano-squids [1]; cryogenic circuits for switches operating at high frequency [2]; single-photon detectors [3] and so on. Furthermore, the development of these superconducting systems allows studying different superconducting properties, which are crucial for a better understanding of the entire superconducting phenomenon [4–8]. As an example, it has recently been reported that many hetero-structures are built with the niobium being the active superconducting element [9–12]. In this sense, these hybrid systems provide a unique opportunity to explore interesting phenomena in nanoscale regime; for instance, the correlation between superconductivity and ferromagnetism.

Nb films, with superconducting properties, can be obtained with different preparation procedures [10–21]. According to results reported in the literature [8,10–21], it can be inferred that sputtering seems to be the most suitable method to produce superconducting Nb films, i.e., a nanostructured material with optimal critical transition temperature T_C and other appropriate physical properties. However, during a sputtering process, there are several deposition parameters that may determine the final physical properties of the sputtered films. The most relevant deposition parameters are, in general, the substrate material, the substrate temperature T_S, the residual chamber pressure (the value and its constituent) and, obviously, the deposition rate. As a general rule, finding out the optimal parameters is a challenging task, since they will depend on the particularities of the experimental setup used in the deposition process. The necessary knowhow to obtain good Nb films comes from a robust and clear comprehension about the correlation between superconducting (T_C, critical current density J_C, etc.) and structural (lattice parameters, grain sizes, roughness, etc.) properties.

It has been shown that the T_C value of sputtered Nb films does not only depend on the lattice parameter [17,19] and the grain size [17, 22], but it can also be modified by the material used as substrate [17, 23]. Moreover, the T_C value can either decrease when the Nb film is annealed ex-situ to temperatures close to 573 K [16] or it may enhance when hydrostatic pressure is applied [13]. It has been reported that Nb films deposited on Si substrate show a larger lattice parameter and reduction of both, thickness and grain size [20]. These two effects were discussed in terms of the O_2 content at Nb grain boundaries [20]. In thin films, the T_C value can also be affected by other mechanisms as, for example, the proximity effect [8,15,18]. The references cited above show that the deposition by DC sputtering allows to prepare Nb films with different structural parameters, where the superconducting properties can clearly be correlated with the structural disorder effect [15]. Optimal superconducting properties have been reported for 50 nm thick Nb films grown on sapphire substrates [15].

In spite of the efforts devoted to understand this problem, some particular questions are still open, deserving a particular attention. One point that has not extensively explored in the literature is how the substrate temperature T_S can modified the superconducting

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properties of sputtered Nb films grown directly on the Si substrates [17]. So, in the present work, it was done a systematic study to describe the influence of T_S on the quality of superconducting properties of Nb films prepared by DC sputtering onto Si (100) substrates. To reach this goal, several depositions were performed at different T_S values, while the other parameters [deposition rate, gas pressure, residual gas atmosphere (measured and "controlled" by residual gas analyzer - RGA), Si substrate wafer, etc.] were fixed during each process. Our results clearly demonstrate a dependence of the superconducting properties of Nb films on their morphologies; the latter governed by the T_S value. In addition, the results are reproducible, since we have prepared three similar batches of samples and they have shown quite similar T_C and J_C values. For example, the T_C value changes roughly 0.1 K in the reproducibly tests.

2. Experimental

100 nm thick Nb films were prepared onto Si (100) substrates, kept at different T_S, using the AJA Orion-8 DC magnetron sputtering setup. The T_S values were measured using a K-type thermocouple sensor coupled to the rotating sample holder. T_S was stabilized (during 5 min) before starting the Nb film depositions. Thus, we can assume that the film is in thermal equilibrium with the sample holder (film temperature $T_F = T_S$). A 5 in. silicon wafer was cut in squared shapes, with typical areas covering about $10^{-2} \times 10^{-2}$ m^2. These Si pieces were preliminary cleaned in a bath with Extran neutral detergent by an ultrasonic machine. Subsequently, they were washed in consecutives and repeated baths of water and acetone. Finally, they were dried with a flux of inert gas. 2 in. Nb disk, with 99.995% purity, was used as the target material. The residual chamber pressure was better than 5×10^{-6} Pa before filling the deposition chamber with 3×10^{-1} Pa argon gas (99.999% purity). The residual gas atmosphere was measured by a Residual Gas Analyzer (RGA) device. The film thickness was calibrated by measuring X-ray reflectivity of a single film. The deposition rate of about 0.72 Å/s (maximum rate of Nb obtained with our power supply), obtained by X-ray, was monitored using the quartz crystal balance, which is positioned close to the sample holder position. A pre-sputtering process was performed before the film depositions in order to remove 10 nm from the Nb target and reducing possible natural oxide layers during the deposition. The 100 nm films were prepared at T_S equal to 293, 323, 373, 393 and 423 K. No buffer and capping layers of other materials were used, which means that the Nb films were directly deposited onto the Si (100) substrate. In principle, since the sample characterizations were done ex-situ, a natural oxide Nb phase is formed before the sample measurements. This natural Nb oxide seems to passivate the metallic core Nb and keeps the properties of the samples for a long time interval. Measurements performed two weeks after the sample preparation, keeping the film in vacuum setup, show similar physical features and absence of aging effect (no significant change in the T_C value $- \Delta T_C \sim 0.1$ K).

The grazing incidence X-ray diffraction (GIXRD) patterns were recorded, using a Cu K_α-radiation ($\lambda = 1.5418$ Å), in an X-ray diffractometer, *Rigaku Ultima IV*, where the incident angle was kept fix (the sample holder is fix with $\alpha_i = 1°$), while the detector scanned the film surface in steps of 0.05° and count rate of 3 s/pass. The GIXRD patterns were fitted by the Maud software [24], which already considers the small 2θ displacements that occur due to the refraction effects [25]. The relevant extracted parameters were: the lattice parameter (a) and the crystalline grain size (τ). The morphology of the films surface was studied using an atomic force microscopy (*SPM 9600 Shimadzu*). The data were acquired in a non-contact method, where parameters like the root-mean-square roughness (σ), the lateral correlation length (ξ), the fractal dimension (δ) of the surface and the effective area A of the particles, were measured on typical areas of 1×1 μm^2.

The magnetic properties of the Nb films were obtained by using a Physical Property Measurement System (PPMS) from Quantum Design that operates with an evercool type-II system. The critical temperature T_C of the Nb superconducting films was obtained from the onset of the diamagnetic signal in the M(T) curve recorded for a probe field of 3979 A/m. Magnetic loops [M(H)] were recorded with the scan field applied perpendicular to the sample plane for different temperatures, after cooling the sample from $T > T_C$ in zero-field mode (ZFC), for each M(H) case.

3. Results and discussions

Room temperature GIXRD patterns of the Nb films, prepared at different T_S, are plotted in Fig. 1. The most relevant peaks (those clearly observable in the figure) of these GIXRD profiles were indexed, using the *Maud* software [24], to the body centered cubic (bcc) structure of bulk Nb (space group: I m–3 m, 229). Furthermore, three low intense broad Bragg lines [100 times lower than the most intense peak, (110)] are peaked between $2\theta = 40°$ and 52°. The intensity is too low that the peaks are diluted in the background, being not detected in two of the five films investigated here (investigations done in the same samples after 14 days still show similar features). These additional peaks are related to the thin natural Nb oxide formed at the film surface during ex-situ experiments. The very low intensities of these lines (relatively to the main peaks of the metallic Nb) inform that only few monolayers were naturally oxidized and according to above results, this process was enough to passivate and protect the metallic core of Nb films against deeper oxidation of Nb layer.

Another aspect to be noted is the fact that in the GIXRD method (using a fix sample holder), the diffraction lines [different (h k l)] can only appear if the film is not textured and the crystallographic grains have random orientations, like in a polycrystalline powder [26]. In other words, from the structural viewpoint, all these Nb films are polycrystalline, have a bcc structure and keep their pure metal properties, since it is previewed that the small amount of oxide does not influence the magnetization results, as will be shown later. Table 1 and Fig. 2 display the dependence of the a- and τ-parameters on the T_S; values that were obtained by fitting the GIXRD patterns with the Maud software [24]. The uncertainties were calculated, from the fitting procedure, to be equal to 0.002 Å and 2 nm for the a- and τ-quantities, respectively. As can be noted, for $T_S \leq 373$ K, the a-quantity remains nearly constant. Further increase of T_S markedly shift the lattice parameter of the film to a value well above the one found in the bulk phase (3.303 Å) [17], and according to the literature, this would lead to a considerable decrease of the T_C value [17,19]. This increase of the a-parameter for the films prepared at $T_S > 373$ K may be attributed to two main effects: Si interdiffusion to Nb layer and/or an enhancement of the internal stress caused by high temperature deposition. Thus, these two effects would degrade the superconducting properties.

The AFM images of the Nb films are shown in Fig. 3. The σ [1.4(2)–1.6(2) nm] and δ [2.03(2)–2.05(2)] parameters are roughly constant for all films, when their uncertainties are considered. The δ parameter describes (by a single parameter) a scale-independent measure of the surface roughness (for lengths lower than ξ), i.e., it represents the roughness irregularities taken in many scales of length (<ξ). Here, the value around 2 for the δ quantity informs that the film roughness is relatively smooth (<ξ), pictured as sand dunes. On the other hand, it should be mentioned that the ξ quantity is a statistical parameter that gives a measure of the roughness lateral length scale (it can be roughly understood as an average of roughness wavelengths taken over all directions). Therefore, the ξ parameter works as an effective cut-off length, where the height correlations vanish, i.e., if the distance between two points is within of the ξ value, their heights are correlated and the roughness can be described by the δ parameter [27,28]. Thus, our results suggest that the ξ parameter varies from 2(1) nm ($T_S = 293$ K, 323 K) to 3(1) nm ($T_S = 373$ K, 393 K), which indicates that the roughness oscillations have predominantly short wavelengths. Consequently, it can be said that the films roughness is similar and it is not dependent