STRAIN EFFECTS IN THE SUPERCONDUCTING PROPERTIES OF NIOBIUM THIN FILMS GROWN ON SAPPHIRE*

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Abstract

Fundamental aspects of epitaxial thin film growth such as strain due to lattice mismatch can drastically affect the superconducting properties of thin films. In our studies, we show a clear correlation between the surface morphology and microstructure with the superconducting properties of single crystal Nb(110) thin films sputter deposited on *a*-plane sapphire substrates. We found that the lattice mismatch between Nb and sapphire induces the formation of a hexagonal surface structure during the first 3 atomic layers followed by a strained *bcc* Nb(110) phase whose lattice parameter progressively relaxes reaching bulk value after 14 Nb atomic layers. The influence of the properties of such initial layers on the superconducting transition process is analyzed in detail. The results reported here indicate that interfacial strain effects must also be considered when evaluating the feasibility of multilayers for SRF cavity applications

INTRODUCTION

Superconducting thin films and multilayers have attracted the attention of the SRF scientific community in the last years due to the promise of overcoming the maximum field gradients that current bulk Nb SRF cavities can withstand, pushing them above 100 MeV/m [1]. Nevertheless, in order to achieve the desired properties, special attention needs to be devoted to the epitaxy and growth mode of such thin films. In particular, high quality films have been obtained on *a*-plane sapphire, where Nb grows (110) with the Nb[$\bar{1}11$] direction parallel to Al_2O_3[0001] leading to a lattice mismatch of 10.7% along the Nb[001] direction and of 8.3 % along the perpendicular Nb[110] direction[2, 3]. Oderno et al.[4] showed a transition from an initial hexagonal surface structure for thicknesses lower than 1.5 nm to *bcc* Nb(110) for thicker films, nevertheless they did not report on whether the *bcc* phase is already relaxed or if there exists a coherent strain relaxation. The study of the early stages of Nb thin films growth is of paramount importance since the presence of additional phases, coherent relaxation of the strain, misfit dislocations or other epitaxial growth defects can strongly affect the superconducting properties of the entire system. Nevertheless, a clear connection between interfacial growth effects and superconducting properties for Nb thin films grown on *a*-plane sapphire has not been established yet. Here, we present a complete correlation between morphology and structure of Nb thin films and their superconducting properties, emphasizing the influence of the first strained atomic layers on the global superconducting properties of the films.

RESULTS AND DISCUSSION

Nb thin films were prepared by DC magnetron sputtering deposition in an ultra-high vacuum (UHV) system with base pressure in the low 10^{-10} Torr range on *a*-plane sapphire substrates with miscut angles lower than 0.05 degrees. The substrates were ultrasonically cleaned in baths of acetone and methanol and subsequently annealed at 600 ºC for one hour in UHV conditions prior to growth. Sputtering deposition was carried out from a high purity (99.95 %) Nb target at 1 x 10^{-3} Torr Ar pressure, resulting in a growth rate of 0.35 Å/s. Nb films of thickness up to 600 nm were deposited at a substrate temperature of 600ºC which was determined to favor crystalline ordering.

The surface morphology of superconducting thin films is of great importance for SRF applications since it can affect their surface impedance and hence their SRF performance In Figure 1, Atomic Force Microscopy (AFM) topography images clearly show the evolution of the morphology of the Nb films with thickness ranging from 30 nm to 600 nm.

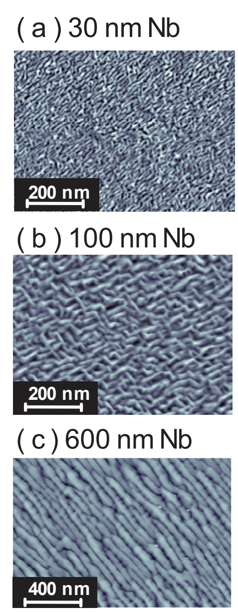

Figure 1: AFM topography images for the (a) 30 nm (b) 100 nm and (c) 600 nm Nb films grown on a-plane sapphire.

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Elongated surface features along the two principal directions of the Nb(110) surface, *i.e.* Nb[001] and Nb[110], are observed for thinner films [Figure 1 (a) and (b)]. For thicker films, only one of the anisotropy directions prevails and grooves along such direction are observed [Figure 1 (c)].

The microstructure of the films was investigated *in-situ* with reflection high energy electron diffraction (RHEED) during various stages of growth. Initially, two different streaks patterns that repeat every 60 degrees are observed when the electron beam is directed along the in-plane [0001] and [$\bar{1}$100] sapphire directions, *i.e.* separated by 30 degrees on the surface. In addition, the interline distances along those two directions have a √3 ratio, indicative of a hexagonal surface structure as previously observed by Oderno et al. [4]. The lattice parameter for such hexagonal structure was extracted from the pattern found along the Nb[11$\bar{2}$0] direction parallel to the Al_2O_3[0001] direction, as shown in Figure 2. An initial value of 0.294 nm for 1 Nb AL decreases and stabilizes at 0.289 nm for 2 and 3 AL. The hexagonal diffraction patterns are progressively replaced by a new set of streaks corresponding to *bcc* Nb(001) orientated Nb[$\bar{1}$11]||Al_2O_3[0001]. Nevertheless, coexistence of both phases is found for Nb thickness ranging from 3 to 5 AL (0.69 to 1.15 nm). An initial 3.93 % expansion of the lattice parameter compared to bulk is observed for films 5 AL thick due to strain in the lattice. Such strain is progressively relaxed, achieving Nb bulk lattice parameter (0.33 nm) for 14 atomic layers (3.22 nm) thick films. Our RHEED analysis clearly reveals that the Nb films grown on *a*-plane sapphire evolve via two main mechanisms to overcome the initial lattice mismatch: an initial hexagonal phase energetically more favorable for thicknesses up to 3 AL, followed by a strained *bcc* Nb(110) phase which progressively relaxes reaching equilibrium after 14 Nb atomic layers.

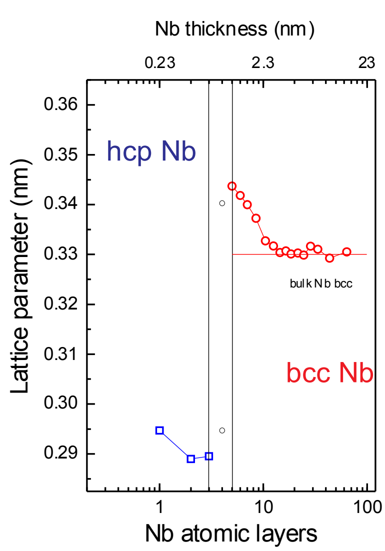

Figure 2: Evolution of the Nb structure and lattice parameter for films with thickness ranging from 1 to 63 atomic layers (0.23 nm/AL). An initial hexagonal growth mode is observed for the first 3 atomic layers followed by a strained bcc phase, which lattice parameters relaxes after 14 atomic layers (3.22 nm).

The superconducting properties of the Nb thin films were investigated using Superconducting Quantum Interference Device (SQUID) magnetometry applying DC and AC magnetic fields in order to explore the static and dynamic responses of the system. SQUID magnetometry is a well-established method to investigate the superconducting properties of metals and high temperature superconductors. When AC magnetic fields are applied to the sample, the frequency dependent complex susceptibility has two components $\chi(\omega)=\chi'(\omega)+i\chi''(\omega)$, where the real part $\chi'(\omega)$ describes the behavior of the system in phase with the incoming AC field and the imaginary part $\chi''(\omega)$ accounts for the energy losses in the system. Figure 3 shows χ' (left column) and χ'' (right column) for the 30, 100 and 600 nm thick Nb films measured with an AC field of 3.5 Oe and 1 Hz superimposed to a continuous 100 Oe field, both along the plane of the Nb film. The field used to probe the samples was chosen much smaller than the lower critical field H_{c1} for bulk Nb, i.e. 1700 Oe, in order to minimize its effects on the superconducting state of the films. For the case of the 100 and 600 nm thick Nb film a type II superconductor transition is observed, with critical temperatures T_c of 8.75 and 8.7 K respectively, which differs from the 9.2 K bulk value due to their thin film nature. Nevertheless, for the case of the thinner 30 nm thick Nb film, a χ' susceptibility transition with two steps can be clearly observed, accompanied by two peaks in the χ'' susceptibility at 7.64 and 8.08 K. Even though the critical temperature is expected to decrease with thickness in Nb films, the observed reduction exceeds previous observations[5]. The presence of two or more steps in the χ'' *vs.* temperature dependence has been associated in previous studies with the presence of grains and transport through grain boundaries in superconductors[6, 7]. Such behavior has been explained as the consequence of the onset of intra-grain and inter-grain currents during the superconducting transition. When cooling down the superconductor bellow its critical temperature, an onset of superconductivity is initially observed inside the grains giving rise to a first inflection of χ' and a first peak in χ''. In this regime, intra-grain currents give rise to the field shielding and thus to the first inflection of χ' and the first peak in χ''. As the temperature is further lowered, inter-grain currents appear thus completing the transition to superconductor state and giving rise to the second inflection of χ' and the second peak in χ''. We note that for the Nb on *a*-plane sapphire system we observed similar grain sizes using x-ray diffraction (XRD) for all the films, ranging from 30 nm for the 30 nm thick Nb film to only 46 nm for the 600 nm film, thus we do not expect variations in inter-grain contact *vs.* Nb thickness. Nevertheless, we have previously shown with our

RHEED analysis that the Nb films are strained during the early stages of growth. Such effects are dominant in the first 3 nm and therefore can strongly affect the superconducting response of the thinner films and be less relevant in thicker films where the dominant contribution is due to the bulk-like phase with optimum superconducting properties. Thus, the observed response for the 30 nm thick Nb film can be attributed to the presence of two predominant phases in the sample: *(i)* a first one with poor superconducting properties due to strain in the lattice and positioned closer to the interface with the substrate and *(ii)* a second one corresponding to the relaxed Nb layers that exhibit similar behavior to bulk Nb.

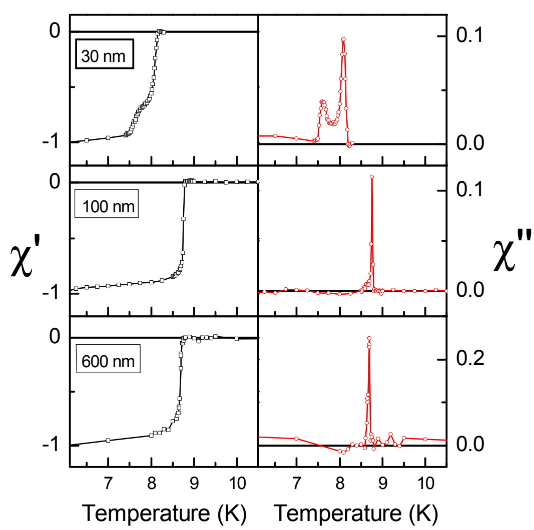

Figure 3: (a) Real χ' and (b) imaginary χ'' parts of the susceptibility for 30, 100 and 600 nm Nb films grown on a-plane sapphire. The two steps structure observed in the 30 nm Nb film is associated to the strain in the first atomic layers.

CONCLUSIONS

A complete correlation between morphology and structure with superconducting properties such as critical temperature and complex susceptibility has been presented for epitaxial single phase Nb(110) thin films sputter deposited on *a*-plane sapphire substrates. A two-fold surface anisotropy is found for films up to approximately 100 nm thick turning into uniaxial anisotropy consisting of very elongated features on the surface in thicker films. RHEED characterization of the films demonstrated the presence of an initial hexagonal phase energetically more favorable for thicknesses up to 3 AL, followed by a strained *bcc* Nb(110) phase which progressively relaxes reaching equilibrium after 14 Nb atomic layers. The superconducting properties of such initial strained layer are found to be poorer than the remainder of the film after strain relaxation is achieved, exhibiting lower critical temperatures and fields. Thus, our studies provide insight into the identification of dissipative effects present in the film lattice associated with the presence of strain and associated defects during the early stages of growth of Nb on *a*-plane sapphire. These studies will help to elucidate the choice of appropriate ceramic surfaces leading to enhanced SRF performance in multilayers for next generation cavities.

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REFERENCES

[1] A. Gurevich, Applied Physics Letters **88** (1), 012511 (2006).
[2] A. M. Valente-Feliciano, C. E. Reece, J. Spradlin and X. Zhao, Proceedings of IPAC'10, Kyoto, Japan (2010).
[3] G. L. Zhou and C. P. Flynn, Physical Review B **59** (12), 7860 (1999).
[4] V. Oderno, C. Dufour, K. Dumesnil, A. M. P. Mangin and G. Marchal, Philosophical Magazine Letters **78** (5), 419 - 426 (1998).
[5] A. Gubin, K. Il'in, S. Vitusevich, M. Siegel and N. Klein, Physical Review B **72** (6), 064503 (2005).
[6] F. Gömöry, Superconductor Science and Technology **10** (8), 523 (1997).
[7] R. Singh, R. Lal, U. C. Upreti, D. K. Suri, A. V. Narlikar, V. P. S. Awana, J. Albino Aguiar and M. Shahabuddin, Physical Review B **55** (2), 1216 (1997).