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Sputtered niobium: Epitaxial thin film growth on a-plane sapphire

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This application note describes the growth of epitaxial Nb thin films by sputtering onto single crystal substrates at high temperature in the Royce Deposition System. For sputter deposition onto a-plane sapphire substrates at 800°C, a 65 nm Nb film shows strong bcc 110 structural ordering and a residual resistivity ratio of 30, consistent with an epitaxial microstructure. The sample has a superconducting T_c of 8.986 K. Epitaxial Nb has important applications for superconducting electronics, quantum technologies, and as a template for the epitaxial growth of further overlayers.

1 Introduction

Nb is an elemental superconductor. In pure bulk samples, Nb has a superconducting T_c of 9.25 K [1]. The T_c is relatively high for an elemental superconductor with s-wave pairing, which makes it the material of choice for many applications. Recently, we have demonstrated a supercurrent diode effect in single layer epitaxial Nb devices [2] and a superconducting memory effect in epitaxial Nb–Er multilayers [3]. Additionally, epitaxial Nb is commonly used as a template for the epitaxial growth of further metallic and ceramic overlayers [4], such as rare-earths [5, 6].

In this application note, a 65 nm thick Nb thin film grown in the Royce Deposition System is shown to have strong bcc 110 structural ordering and a residual resistivity ratio of 30, consistent with an epitaxial microstructure. For application considerations, the films have low surface roughness and a T_c close to bulk.

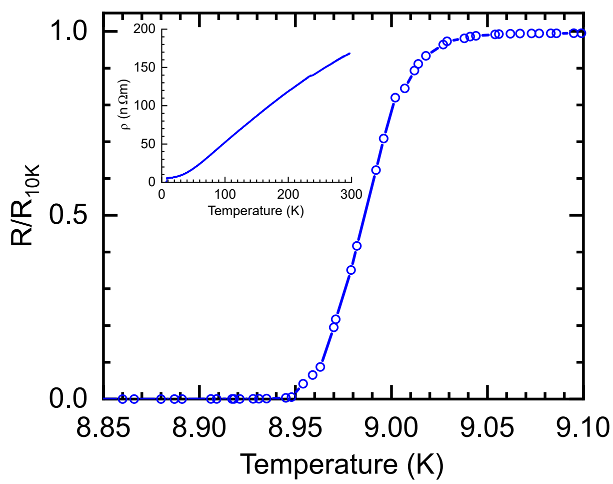

Figure 1: Normalised resistance versus temperature of 65 nm epitaxial Nb film. The measured T_c at 50% the normal state resistance is 8.986 K and the transition width is 48 mK. Inset: Resistivity for the film over the full range of temperature.

2 Growth

Nb films were deposited by dc magnetron sputtering in the Royce Deposition System. The magnetrons were mounted in a ring below the substrate with a source-substrate distances of 134 mm. A liquid nitrogen shroud was used to bring the base pressure of the system down to 2.9 x10^{-9} mbar. A 20x20mm a-plane Al_2O_3 substrate was placed in a Ta holder and loaded at room temperature, then the substrate heater was ramped to a set temperature of 800°C for the growth. For growth, Ar (6N purity) gas was flowed through the chamber at 10 sccm with the gate valve to the turbo-molecular pump throttled to 46% to give an equilibrium process pressure of 3.5x10^{-3} mbar. Nb was sputtered from 5N purity targets at a dc power of 80 W for a deposition rate of 0.62 Å/s per Nb source. To increase the growth rate, two Nb sources were co-sputtered.

In this application note, a-plane sapphire substrates were chosen for epitaxial growths. The epitaxial relationship between sapphire and Nb is well characterised and the growth method described here should be applicable to other substrate orientations [4]. Note that the growth temperature may need to be further optimised for alternative substrates.

3 Properties

We measure resistance as a function of temperature using a ^{4}He cryostat and a conventional four-point-probe measurement configuration. The sample used here has been patterned into a 6 μm track. The T_c is determined as the temperature where the resistance reaches 50% the normal state (10 K) value. Shown in Figure 1, the measured T_c is 8.986 K. The transition width can be determined from the decrease

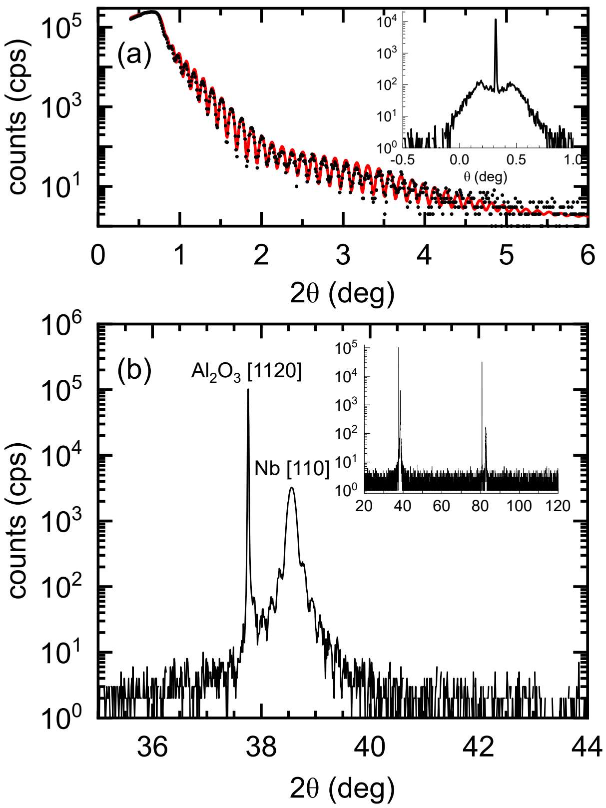

Figure 2: X-ray characterisation of the epitaxial 65 nm Nb film. (a) X-ray reflectivity with best fit to the data. The parameters used to determine the best fits are given in Table 1. The insets show the rocking curves at 2$\theta = 1°$. (b) X-ray diffraction aligned to the main substrate peak. The Nb 110 and Al_2O_3 1120 Bragg diffraction peaks can be observed. The inset shows the full angular range of the data.

in resistance from 90% to 10% the normal state value and is 48 mK.

Considering the decrease in resistance between room temperature and 10 K (Figure 1 inset), we determine the residual-resistivity ratio (RRR) for our film to be 30. The RRR can be used to estimate the mean free path (ℓ) of a Nb film according to the relations,

$$RRR = \frac{\rho_{\text{thermal}} + \rho_{\text{residual}}}{\rho_{\text{residual}}}, \quad (1)$$

and

$$\rho_{\text{residual}}\ell = 460 \text{ n}\Omega\text{m.nm}, \quad (2)$$

taking ρ_{thermal} to be approximately the bulk resistivity of 139.5 nΩm, or by directly determining ρ_{residual} from the van der Pauw method. For our film, we determine an estimate for ℓ of 96 nm. By comparison, a polycrystalline film grown at room temperature of a similar thickness has a RRR of 2.8, giving an estimate for ℓ of 6 nm. The increased RRR and ℓ is expected for the epitaxial sample due to the decrease in crystallographic defects such as grain boundaries.

Fitting the Bloch-Grüneisen formula[7, 8] to the full R(T) data set gives a Debye temperature ($\Theta_D = 200 \pm 5\,\mathrm{K}$, compared to a reported value for bulk niobium of $270 \pm 10\,\mathrm{K}$[9]

X-ray diffraction (XRD) and reflectivity (XRR) of the film is measured using a Bruker D8 diffractometer at room temperature. An additional four-bounce Ge(022) incident beam monochromator is added to the x-ray optics to remove all incident wavelengths except Cu K_α (1.54056 Å).

Figure 2 (a) shows the XRR on the epitaxial Nb film with the best fit line through the data. The Kiessig fringes are present out to about 2θ= 5° and the observed enveloping at 2θ= 2° suggests the presence of an oxide surface layer. Fitting to the XRR data provides the thickness, roughness, and density of the films. The best fit parameters corresponding to the fits shown in Figure 2 (a) are provided in Table 1. To fully model the reflectivity, a thin interface layer is added to the Al_2O_3/Nb interface and a niobium oxide layer is added to the top surface, both with density as a free fitting parameter. The fitted roughness at the Al_2O_3/Nb interface and need for an additional interface fitting layer may correspond to an alloying or chemical reaction between the Nb and the Al_2O_3 substrate, such as the formation of NbAl. These additional fitting layers are a common feature of thin film Nb [10]. The insets to Figure 2 show the rocking curves at 2$\theta = 1°$. The Yoneda wings are suggestive of surface roughness.

Figure 2 (b) shows the XRD for the epitaxial Nb film. We fit a Gaussian to the bcc Nb 110 peak to determine the peak position and FWHM. From the peak position (2$\theta = 38.559°$), we calculate the Nb 110 c-plane spacing to be 2.333 Å. This value is identical to the bulk value, indicating that the epitaxial growth has resulted in a film with very little c-plane strain.

To estimate the crystallite size, we use the Scherrer formula and the fitted FWHM of 0.14206° to yield an estimated c-plane grain size of 59 nm. This estimated grain size is close to the fitted thickness of the Nb of 64.4±0.3 nm indicating good structural coherence (92%).

Layer	Thickness (nm)	Roughness (nm)	SLD r_eÅ^{-3}
(oxide)	2.2 ± 0.4	0.8 ± 0.1	1.853
Niobium	64.4 ± 0.3	0.5 ± 0.2	2.083
(interface)	1.9 ± 0.5	1.0 ± 0.5	1.877
Al_2O_3	–	1.0 ± 0.2	2.427

Table 1: Structural parameters of the 65 nm thick epitaxial Nb film corresponding to the best fit shown in Figure 2 (a).

4 Further Information

The Royce Deposition System is a multichamber, multitechnique thin film deposition tool based at the University of Leeds as part of the Henry Royce Institute. Materials from the Royce Deposition System are available as a facility service and for collaborations. Sample growth ID: MET20211008_03.

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