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DC MAGNETISM OF NIOBIUM THIN FILMS*

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Abstract

Niobium thin films were deposited onto a-plane sapphire with varying kinetic energy and varying substrate temperature. There were no consistent trends which related the particle energy or substrate temperature to RRR. The sample which displayed the largest RRR of 229 was then compared to both a thin film deposited with similar conditions onto copper substrate and to bulk niobium. DC magnetometry measurements suggest that the mechanism of flux entry into thin film niobium and bulk niobium may vary due to differences in the volumes of both defects and impurities located within the grains. Results also suggest that magnetic flux may penetrate thin films at small fields due to the sample geometry.

INTRODUCTION

The RF cavities made of copper and coated with a thin layer of superconducting material are already widely used as an alternative to bulk Nb cavities. They are less expensive, copper has better thermal conductivity, and in theory they could perform even better than the bulk Nb cavities. However, in present their cavities do not provide the same quality of $Q_0(E)$ function yet. ASTeC continues its superconducting thin film programme of systematic study on correlation between the surface preparations, the deposition parameters, morphology, structure and chemistry, and superconducting properties of the films. This paper was devoted to deposit the film with the highest quality and compare its properties to bulk Nb and to check how significant could be the edge effect in SQUID magnetometer measurements in magnetic field.

FILM DEPOSITION

18 niobium thin film samples were deposited in ASTeC by either high impulse magnetron sputtering (HiPIMS), DC or Pulsed DC magnetron sputtering onto single crystal a-plane sapphire. Each substrate was cleaned in ultrasonic baths of acetone, then isopropanol, then deionised water before being inserted into the deposition chamber. The vacuum system was baked prior to deposition at 150 °C for 3 days to achieve a base pressure of 2×10^{-10} mbar. A three-inch planar magnetron was used to deposit the sample with a 99.95% purity niobium target. The magnetron was 150 mm from the substrate surface at an angle of 45°. The substrate was rotated at 4 rpm for the duration of the

deposition. Krypton sputter gas was used during the deposition at a constant pressure of 7×10^{-3} mbar. For samples deposited by HiPIMS, the power supply was set to pulse at 200 Hz with a pulse length of 100 μ s. The average plasma current was 600 mA whilst the peak current at the target surface was 40 A. When depositing with pulsed DC, the power supply was set to a power of 400 W with repetition rate of 350 kHz and 50 % duty cycle. DC sputtering was performed with a power of 400 W. Films were deposited with a substrate temperature ranging 500 to 1000 °C with either a grounded substrate or a DC bias voltage of -80 V. The deposition was monitored throughout by sampling small volumes of process gas with an RGA and by optical spectrometer. It has been assumed that HiPIMS provides the largest ionisation percentage of the sputtered material, followed by pulsed DC then finally DC [1]. The study intends to describe the changes in superconducting properties of niobium thin films which depend on substrate temperature and the energy of sputtered material. A-plane sapphire was chosen as substrate due to its high melting temperature of 2050 °C.

Samples were first measured for RRR. The sample with the largest RRR was later compared to a sample deposited with the same deposition conditions onto copper substrate which has a much lower melting temperature of 1085 °C. The comparable samples were also analysed by both SEM and DC magnetometer.

RESULTS

RRR measurements

The deposited films were tested for RRR using a four-point probe with the resistance of the sample measured from room temperature down to below T_c . The RRR results for every deposited sample are shown in table 1. The results show that RRR varies from 10 to 229 for all samples however there was no consistent trend which connected RRR with either the power supply used, the deposition temperature or the applied bias. In some cases, very similar deposition conditions resulted in very different RRR.

The resistance versus temperature curve for the sample with the largest RRR of 229 is shown in figure 1. The sample was deposited by HiPIMS at 800 °C with a -80 V DC bias. A sample deposited with the same deposition conditions onto copper substrate, and with RRR of 52, is

also shown for comparison. The sample deposited onto a-plane sapphire exhibited the smallest normal state resistance of all samples at $0.0006 \text{ m}\Omega$ just before the superconducting transition. The superconducting transition temperature of the sample was found to be $9.32 \pm 0.02 \text{ K}$. The niobium film deposited onto copper displays a larger normal state resistance than that deposited onto a-plane sapphire and a T_C of $9.40 \pm 0.03 \text{ K}$.

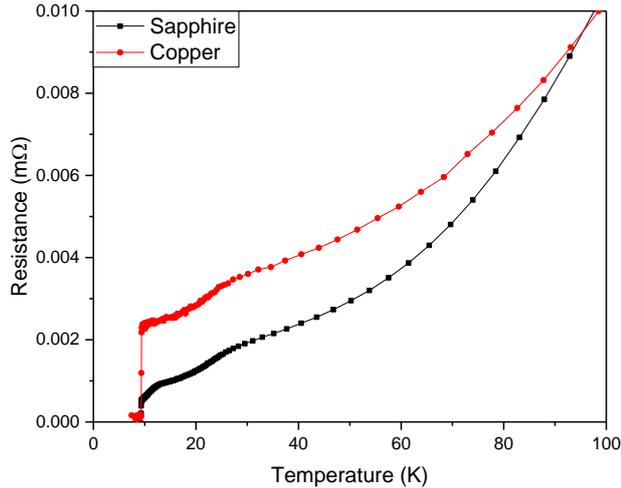


Figure 1: Resistance versus temperature curves for niobium thin films deposited with the same conditions onto either a-plane sapphire or copper.

Table 1: The RRR values for niobium thin films deposited by either DC, pulsed DC or HiPIMS at temperatures of either 500, 800 or 1000 °C with either a grounded substrate or biased at -80 V DC.

| Power Supply | Temp (°C) | Bias (-V) | RRR |
|--------------|-----------|-----------|-----|
| HiPIMS | 500 | 0 | 124 |
| Pulsed DC | 500 | 0 | 172 |
| DC | 500 | 0 | 10 |
| HiPIMS | 500 | 80 | 13 |
| Pulsed DC | 500 | 80 | 121 |
| DC | 500 | 80 | 218 |
| HiPIMS | 800 | 0 | 150 |
| Pulsed DC | 800 | 0 | 117 |
| DC | 800 | 0 | 47 |
| HiPIMS | 800 | 80 | 229 |
| Pulsed DC | 800 | 80 | 86 |
| DC | 800 | 80 | 18 |
| HiPIMS | 1000 | 0 | 145 |
| Pulsed DC | 1000 | 0 | 45 |
| DC | 1000 | 0 | 43 |
| HiPIMS | 1000 | 80 | 51 |
| Pulsed DC | 1000 | 80 | 190 |
| DC | 1000 | 80 | 146 |

Figure 2 displays a planar SEM image of the film deposited onto a-plane sapphire with the largest RRR. The niobium grains measure up to 2 microns across. Figure 3 shows a cross sectional SEM image of the comparable thin film deposited onto copper substrate with the same deposition conditions. Again, the niobium grains are the order of microns across, like those deposited onto a-plane sapphire.

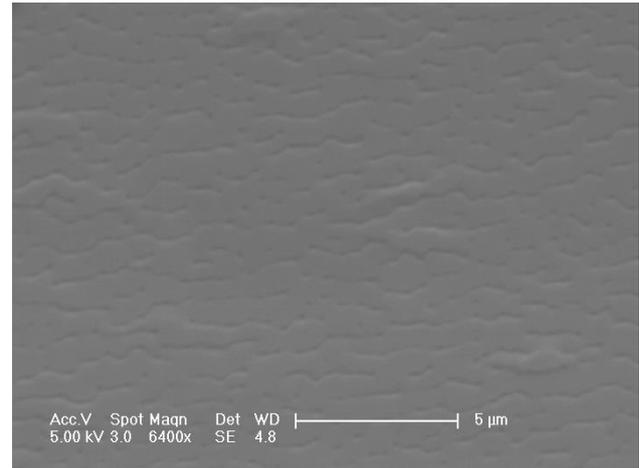


Figure 2: Surface SEM image of the niobium thin film with the largest RRR of 229, deposited by HiPIMS at 800 °C with -80 V DC bias.

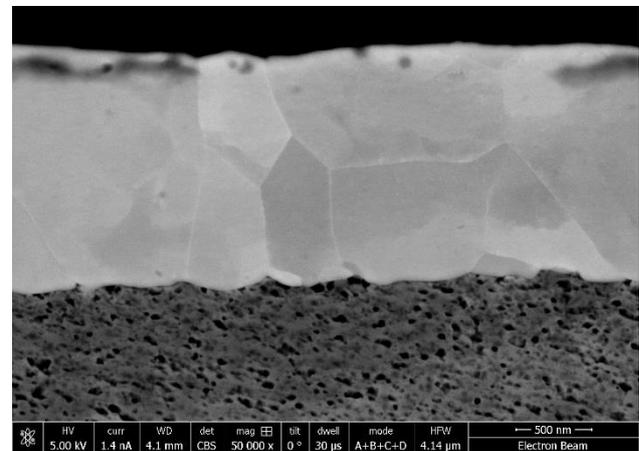


Figure 3: Cross sectional SEM image of a niobium thin film deposited by HiPIMS onto copper substrate at 800 °C with -80 V DC bias.

Magnetisation measurement

The sample with the largest RRR of 229 was measured using a DC magnetometer at 4.2 K. The magnetic moment was measured at ISIS with a Quantum Design MPMS3 by increasing magnetic field from zero to 6000 Oe. Three overlapping 5 mm x 5 mm pieces of the sample were measured. The sample was oriented so that its plane was as close as possible to parallel with the magnetic field.

The magnetic moment of a sample in the Meissner state increases linearly with an applied magnetic field. Thus, a deviation from the linear dependence should correspond to

H_{C1} for a large sample in ideal geometry (infinity long superconducting cylinder or infinity large plate). Non-ideal conditions at the edges of real samples should be considered. In a case of thin films, the edge effect could be significant. Thus, we define the point where the measured moment leaves the straight line describes the first detected flux entry into the sample as H_{dev} .

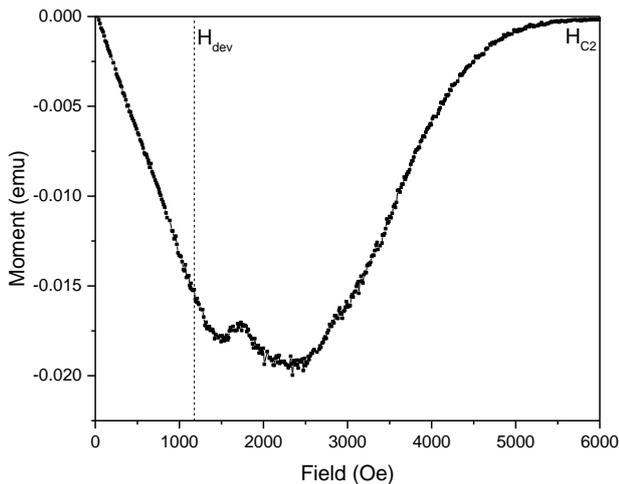


Figure 4: The changing magnetic moment of a niobium thin film deposited onto a-plane sapphire.

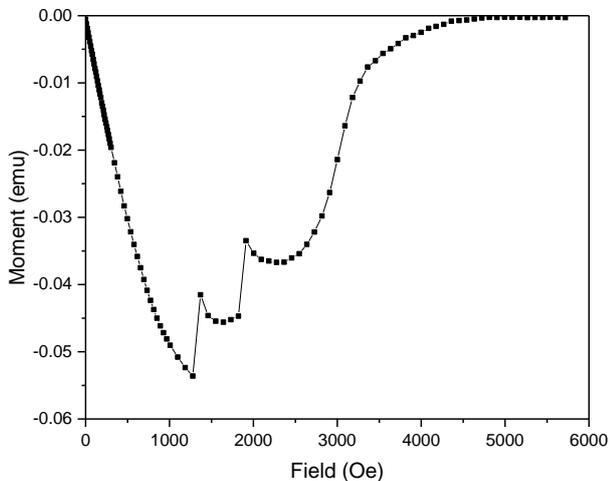


Figure 5: The changing magnetic moment of a niobium thin film deposited onto copper.

A straight fitting line was plotted for the linear section of the hysteresis curve and any deviation from the predicted moment was calculated using the root mean squares error (RMSE) method. The field at which the magnetisation curve returns to zero is equivalent to the upper critical field, H_{C2} , of the thin film.

The DC magnetic moment for the niobium film deposited by HiPIMS at 800 °C and with -80 V DC bias onto a-plane sapphire is shown in figure 4. H_{dev} for the thin film sample was measured at 1200 Oe and H_{C2} was measured at 6000 Oe.

Figure 5 displays the DC magnetic moment for the sample deposited by HiPIMS at 800 °C with -80 V DC bias onto copper. H_{dev} , at 330 Oe, is much smaller than for the film deposited onto sapphire. Alternatively, H_{C2} is smaller for the copper substrate at 5000 Oe.

Meissner state ratio

When the sample is in the full Meissner state the magnetic moment, M_i , increases linearly with the applied magnetic field, H , and can be described as.

$$M_i = \beta H \quad (1)$$

An algorithm was applied to the DC magnetisation curve so that it was displayed as a ratio of the measured moment over M_i . Coefficient β is the equation of the fitting line and was multiplied by the measured moment, M , to give M_i . Thus, a Meissner state ratio, $M/M_i = 1$, corresponds to a sample in the Meissner state. Any deviation from $M/M_i = 1$ will therefore be proportional to the fraction of the sample volume which is no longer in the Meissner state and contains magnetic flux.

The Meissner state ratio of the samples deposited onto a-plane sapphire and onto copper substrate by HiPIMS at 800 °C with -80 V DC bias are shown in figure 6. A search of the literature for DC magnetic data of a bulk niobium sample with comparable RRR to the sample on a-plane sapphire gave values of H_{dev} of 1800 Oe and H_{C2} of 2800 Oe for bulk niobium with RRR of 250 [2]. The blue line added to figure 6 gives an example of the expected M/M_i for the bulk niobium values in [2].

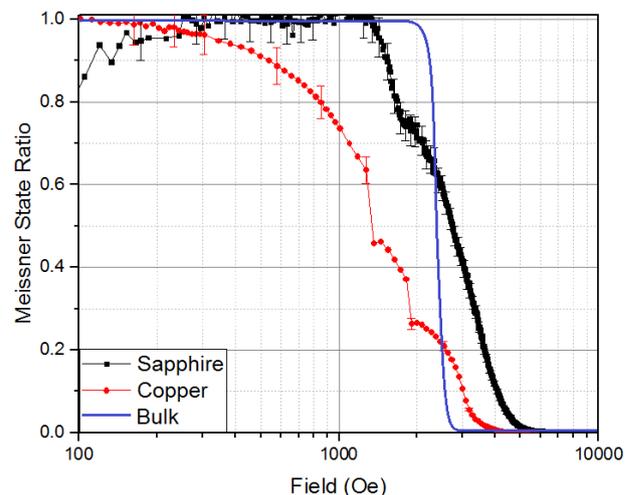


Figure 6: The Meissner state ratio of niobium thin films deposited with similar deposition conditions onto either a-plane sapphire or copper substrate and the expected value for bulk niobium.

DISCUSSION

The purpose of this study was to compare changes in the superconducting properties of niobium thin films, deposited onto a-plane sapphire, depending on the substrate temperature and energy of the arriving ionised material. Expectations were that RRR would increase with

deposition temperature and to some extent by increasing particle energy. RRR may then plateau for all particle energies once the substrate temperature became sufficiently large. The actual data showed seemingly random changes in RRR for the different conditions which were tested. No explanation can be given at present for the observed results.

The values of H_{dev} and H_{C2} dictate the sharpness of the transition from full Meissner state to normal conducting. The transition itself becomes more obvious when looking at the plot of M/M_i . The transition represents the mixed superconducting / normal conducting state, the properties of which, are dependent on the defect or impurities present within the superconductor which act as pinning centres.

As H_{dev} and H_{C2} are different for both the thin films and bulk material then the pinning properties of each must also vary.

An explanation of the way the magnetic field penetrates a sample may give further insight into the nature of the types of defect. In the case of type II superconductors, magnetic field will first penetrate as Josephson vortices at the grain boundaries followed by Abrikosov vortices within the grain [3].

The DC magnetic properties of bulk niobium can be explained relatively easily. A type II superconductor with large average grain size but few defects within the grain interior, such as the bulk niobium typically used for SRF cavity fabrication [4], would first allow Josephson vortices to penetrate and be pinned at grain boundaries. The magnetic field which is pinned at the grain boundaries then puts pressure on the grain interior. Once Abrikosov vortices begin to enter the grain interior then they move relatively easily due to the lack of pinning centres. H_{C2} in this instance is small and the transition from Meissner state to normal conducting is sharp.

The DC magnetic properties of thin films can be more complicated to explain. Two theories can be proposed and may combine to cause the observed results in figure 6.

Firstly, the niobium thin films deposited during this study are type II superconductors with both grain boundaries and many defects and impurities within the grain interior, as is typical of films deposited by magnetron sputtering [5,6]. Josephson vortices still penetrate first into the thin film at the grain boundaries but when the Abrikosov vortices enter the grain interior they become pinned and H_{C2} becomes larger than for the bulk material.

Secondly, the film alignment within the magnetic field of the DC magnetometer greatly affects the field strength at which flux can enter the sample. Magnetic field penetrates a thin film at a maximum strength when the sample plane is aligned perfectly parallel to it [7]. Thus, any small deviation from parallel can lead to early penetration at smaller fields and the transition to normal conducting becomes broader.

The comparison between the Meissner state ratio of the sample deposited onto copper and a-plane sapphire highlights how both the models of flux entry into thin films may apply. The SEM images show both films to have comparable grain size but the sapphire sample has the

largest RRR. This would suggest larger volumes of either defects or impurities within the sample deposited onto copper. It can be assumed from the discussion of how flux enters the thin film sample, that H_{C2} should be smaller for the a-plane sapphire substrate. The fact that H_{C2} was largest for the film on a-plane sapphire substrate would suggest that field could have penetrated the sample early due to the sample geometry.

CONCLUSIONS

A range of niobium films were deposited onto a-plane sapphire however the RRR values did not show consistent results. The sample with the largest RRR was then compared to a similar film deposited onto copper and to bulk niobium. The DC magnetic moment was measured and showed that although thin film niobium consistently shows smaller first penetration fields than for bulk, this may in part be due to the thin film geometry within the applied magnetic field.

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