PROOF OF CONCEPT THIN FILMS AND MULTILAYERS TOWARD ENHANCED FIELD GRADIENTS IN SRF CAVITIES

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Nb SRF cavities

- The choice of Nb for superconducting cavities has been dictated by the requirement of having a material with a high lower critical field $B_{c1}$ and a large energy gap $\Delta$ to prevent vortex dissipation and provide a low surface resistance $R_s$ caused by thermally-activated quasiparticles at $T << T_c$ and $\omega << \Delta$,
- $R_s = (A\omega^2/T) \exp(-\Delta/T) + R_i$ where $R_i$ is a small temperature independent residual resistance and $A$ depends on SC parameters and $\omega$ and $T$
Significant improvement could be achieved if a Nb cavity is coated with a multilayer consisting of alternating superconducting S layers with higher $B_{c1}$ and dielectric I layers. The S layer has thickness $d < \lambda$, and therefore can remain in the Meissner state at fields much higher than $B_{c1}$ bulk due to the increase of the parallel $B_{c1}$ in a thin film, while the insulating layer (~15 nm) is needed to prevent Josephson coupling between the SC layers. Such structure would be particularly efficient in the case of elliptical cavities where the magnetic field is concentrated well inside the cavity and is parallel to the surface.

Thin film geometry $\rightarrow B_{c1}$ enhancement $\rightarrow$ Multilayer shielding

Image: CERN Accelerator School
Our experimental approach and methods

• In order to test the Gurevich model we have investigated the effect of microstructure and morphology on the superconducting properties of Nb thin films deposited onto different ceramic surfaces. In particular we studied a-plane sapphire and (001) MgO.
• We have also investigated Nb, NbN, NbTiN and MgB$_2$ based S/I/S trilayers.
• We monitored the microstructure of the films, the morphology of the surface and the superconducting properties as well as the DC and RF transport properties.
• We explored several aspects in the thin film deposition parameters-space, such as growth rate, substrate temperature during growth, annealing treatments, etc.
Thin film growth onto various different surfaces

• Growth on sapphire, magnesium oxide and copper surfaces
**Nb growth on a-plane sapphire**

- Nb can grow epitaxially on a-plane sapphire, with Nb(110)//Al2O3(11-20)

Comparison of RRR values obtained by different groups:

<table>
<thead>
<tr>
<th>Group</th>
<th>Nb film thickness (nm)</th>
<th>RRR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lukaszew</td>
<td>600</td>
<td>97</td>
</tr>
<tr>
<td>S. A. Wolf [1]</td>
<td>600</td>
<td>82</td>
</tr>
<tr>
<td>G. Wu [2]</td>
<td>235</td>
<td>50.2*</td>
</tr>
</tbody>
</table>

*RRR values for niobium thin films is highly dependent on thickness*

Early stages of growth

• Using Reflection high energy electron diffraction (RHEED), we observed a **hexagonal Nb surface structure** for the first 3 atomic layers followed by a strained **bcc Nb(110)** structure and the lattice parameter relaxes after 3 nm.

• RHEED images for the hexagonal phase at the third atomic layer. Patterns repeat every 60 deg.
Susceptibility AC measurements

- The thinner Nb film exhibits two steps in the $\chi'$ susceptibility transition accompanied by two peaks in the $\chi''$ susceptibility due to strained Nb layers at the interface.
- Growth on a-plane sapphire initially follows a hexagonal surface structure to relax the strain and to stabilize the subsequent growth of $bcc$ Nb(110) phase.
- Such initial layers affect the superconducting properties of the films and these effects must be taken into account in the design of multilayers.

\[ \chi(\omega) = \chi'(\omega) + i \chi''(\omega) \]

Biaxial anisotropy is observed for thicknesses up to 100 nm while uniaxial anisotropy is observed. For thicker films
Nb growth on (001) MgO

• Nb can also be epitaxially grown on (001) MgO surfaces.
• *Unexpected findings:* We have found that depending on the deposition conditions it is possible to tailor different epitaxial possibilities.
RHEED images for Nb(110) on MgO
Scaling of surface features

RRR = 46.5
RMS = 6.51 nm
Nb (001) on MgO

RRR = 165    RMS = 4.06 nm

>200 RRR values!
**Nb (001) on MgO**  

MgO out of box | MgO annealed at 600 °C | 30 nm Nb | 100 nm Nb

RHEED beam along MgO [100]

MgO out of box | MgO annealed at 600 °C | 30 nm Nb | 100 nm Nb

RHEED beam along MgO [110]
SQUID characterization

Tc = 9.2 K!

Possible loss due to interfacial strain
Nb-based trilayer

- 30 nm Nb
- 15 nm MgO
- 250 nm Nb
- MgO (100)

RHEED indicates a film with a high degree of (001) texture.

XRD confirmed RHEED results:

- Nb(110)
- Nb(200)
SQUID magnetometry

• Antoine et al [1] using SQUID magnetometry as well as third harmonic analysis to validate SQUID magnetometry measured the vortex penetration field on multilayered samples and demonstrated field enhancement.

• In our work, we measured hysteresis loops as well as trapped moments that appear after application and removal of the applied field, following the work of C. Bohmer et al. [2]

SQUID characterization

Temperature (K)

$\chi'$

$\chi''$

Long Moment (emu)

Field (Oe)

SQUID characterization
Nb on Cu (111)

• Growth at room temperature and annealing at 350 °C leads to the crystallization of Nb islands in a hexagonal surface structure, even though Nb is expected to growth tetragonal (110).

Nb films on Cu (001) surfaces

Possible Nb/Cu(100) epitaxy:

(a) RHEED pattern for Nb(110)/Cu(100)/Si(100) along the Si[100] and Si[110] azimuths. (b) A representative 2 µm x 2 µm AFM scan for Nb films on the Cu template.
SC properties for different growth T

- The films grown at 150 °C have a very sharp transition from the superconducting state to the normal state that begins at ~9 K while films grown at RT have a much more gradual transition.
- Our results suggest that an increased deposition temperature of Nb onto Cu leads to films with higher crystalline quality (grain size) and thus improved superconducting properties (HC1).

Nb films on Cu

Nb films with quality comparable to high RRR bulk Nb (as used for SRF cavities) have been produced both on single crystal and polycrystalline Cu substrates with energetic condensation via ECR (electron cyclotron resonance) at Jlab. SRF measurements are in progress.

Hetero-epitaxial relationships between Nb and Cu verified.

<table>
<thead>
<tr>
<th>Substrate</th>
<th>RRR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single crystal</td>
<td></td>
</tr>
<tr>
<td>Cu (100)</td>
<td>129</td>
</tr>
<tr>
<td>Cu (110)</td>
<td>275</td>
</tr>
<tr>
<td>Cu (111)</td>
<td>242</td>
</tr>
<tr>
<td>Polycrystalline</td>
<td></td>
</tr>
<tr>
<td>Cu fine grains</td>
<td>150</td>
</tr>
<tr>
<td>Cu large grains</td>
<td>289</td>
</tr>
</tbody>
</table>
Other possible SC thin films for the SIS model

NbN, MgB$_2$, etc.
Growth Procedure for NbN Films
Partial Pressure Series

All NbN films are ~200 nm thick based on XRR/Profilometry
Compare Surface Morphology of Nb and NbN similar films

RMS Roughness for comparable film thickness:

NbN <1 nm
Nb(100) 1.21 nm
Nb(110) 2.45 nm

NbN films microstructure

Intensity (arb. units)

$\delta$-NbN(200)  MgO(200)

5.9%
11.8%
14.7%
20.6%
26.5%

$q_z$ (Å$^{-1}$)

(a) Bulk

(b) N$_2$ Partial Pressure (%)  Spacing (Å)

(c) N$_2$ Partial Pressure (%)  Misalignment ($^\circ$)

(d) N$_2$ Partial Pressure (%)  Mosaicity ($^\circ$)
Residual Resistance Ratio

Resistive behavior for NbN differs from that of metals such as Nb. RRR=1 is indication of very good quality film!
Superconducting Properties

(a) Temperature (K)

(b) Normalized Moment (arb. units)

We have initiated investigations on \textit{MgB$_2$ thin films}.

\[ B_{c1} = \frac{2\phi_0}{\pi d^2} \ln \frac{d}{\xi}, \quad d < \lambda \]

\cite{Gurevich06}

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SIS layers

• NbN-based, MgB$_2$ based and NbTiN-based trilayers
NbN-based multilayer

50 nm NbN
15 nm MgO
250 nm Nb

MgO (100)

Long Moment (emu)
Field (Oe)

$H_{c1}$-NbN-based-Multilayer $\sim 220$ mT!
$H_{c1}$-bulk Nb = 170 mT

XRD characterization MgB$_2$-based ML sample

50 nm NbN
15 nm MgO
250 nm Nb
MgO (100)
XRD scan optimized on the MgB$_2$(200) peak.
The scan indicates that there are multiple MgB$_2$ phases present, all strained.

<table>
<thead>
<tr>
<th>Phase</th>
<th>Lattice Constant (Å)</th>
<th>Strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk</td>
<td>3.523</td>
<td></td>
</tr>
<tr>
<td>(2θ = 51.863°)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1$^{st}$ phase</td>
<td>3.4275</td>
<td>2.7108%</td>
</tr>
<tr>
<td>2$^{nd}$ phase</td>
<td>3.4223</td>
<td>2.8584%</td>
</tr>
<tr>
<td>3$^{rd}$ phase</td>
<td>3.4155</td>
<td>3.0514%</td>
</tr>
</tbody>
</table>

![XRD detail](image-url)
SQUID characterization

\( T_c \sim 30.2 \text{ K} \) (recall that bulk \( T_c = 39 \text{K} \))
SQUID measurements

Reference Nb penetration field ~ 1300 Oe; MgB$_2$-based ML penetration field ~ 1700 Oe
Surface impedance characterizations (SIC)

• The ultimate performance test of the films and multilayers for this application is a measurement of their surface impedance, $R_s$.

• We note that $R_s$ can be written as:

$$R_s = R_{BCS}(T) + R_i,$$

where $R_{BCS} = (A\omega^2/T) \exp[-\Delta/(k_BT)]$

**Note:** $R_s$ decreases strongly for higher-Tc materials with larger superconducting gap $\Delta = 1.86T_c$, implying that materials with $T_c > 20K$, could have the $R_{BCS}$ at 4.2 K comparable to $R_{BCS}$ of Nb at 2K. However, small $R_s$ also implies small residual resistance $R_i$ and no nodes in the superconducting gap, which rules out the d-wave high-Tc cuprates for which $R_s \propto T^2$

• JLab’s SIC system is uniquely capable of making temperature-dependent RF surface impedance measurements on 2inch-sized thin film samples.

• Summary of 2 inch samples studied:

  o Nb thin films grown under various conditions
  o Epitaxial MgB$_2$ films
  o NbN-based trilayer on copper
  o NbTiN-based multilayers deposited on sapphire
Nb films were grown at Jlab at different T, and bias conditions, etc. using ECR.
MgB$_2$ films epitaxially grown on sapphire

MgB$_2$ films produced by Temple Univ. were measured calorimetrically at 7.5 GHz

Effective surface resistance of 200 nm and 350 nm MgB$_2$ on sapphire substrates, together with surface resistance of large grain Nb sample. (7.5 GHz)

At the SRF 2011 the surface impedance of a MgB$_2$-based multilayer was reported, and the residual resistance was found $\sim 181 \, \mu\Omega$
Samples grown on Cu substrates contained large grains (on the order of millimeters) that were visible to the naked eye. The surfaces of these samples are dominated by rough features as seen with SEM and optical microscopy. After annealing 10 nm of MgO and 60 nm of NbN were deposited (W&M).
We note that the residual resistance ($R_i$) around 2K is approximately 35 $\mu$Ohm.
We notice that the residual resistance of these thin film samples is one order larger than that of bulk Nb around 2K but in general lower than that of MgB$_2$-based ML samples.
NbTiN-based SIS structures on bulk Nb and Nb/Cu substrates

<table>
<thead>
<tr>
<th></th>
<th>AlN</th>
<th>NbTiN</th>
</tr>
</thead>
<tbody>
<tr>
<td>N₂/Ar</td>
<td>0.33</td>
<td>0.23</td>
</tr>
<tr>
<td>Total pressure [Torr]</td>
<td>2x10⁻³</td>
<td>2x10⁻³</td>
</tr>
<tr>
<td>Sputtering Power [W]</td>
<td>100</td>
<td>300</td>
</tr>
<tr>
<td>Deposition rate [nm/min]</td>
<td>~ 5</td>
<td>~ 18</td>
</tr>
<tr>
<td>Thickness [nm]</td>
<td>10</td>
<td>100</td>
</tr>
<tr>
<td>Tc [K]</td>
<td>N/A</td>
<td>14</td>
</tr>
</tbody>
</table>

NbN-based multilayers have been very promising regarding magnetic shielding, but we note that this material suffers from a higher resistivity due to the presence of both metallic and gaseous vacancies randomly distributed while the ternary nitride NbTiN presents all the advantages of NbN and also exhibits increased metallic electrical conduction properties with higher titanium (Ti) percentage.
We note that the residual resistance ($R_i$) around 2K is approximately 30 $\mu$Ohm. We also distinguish two temperature regimes with transitions around 8.5 K and 14.5K, related to Nb and NbTiN respectively.
Conclusions

• Trilayers incorporating NbN and following the “Gurevich model” were shown to shield niobium in the pioneer work by Antoine et al. using SQUID magnetometry as well as third harmonic analysis.

• By tailoring thin film growth parameters, and also using SQUID magnetometry we were able to demonstrate shielding beyond the critical field of Nb also using NbN-based trilayers.

• We have also demonstrated that other suitable superconductors show promise for SRF applications, but further studies to optimize thin film deposition conditions must be undertaken in this case.

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