ALD-Based NbTiN studies for SIS R&D

Isabel González Díaz-Palacio on behalf of the SRF R&D Team

TESLA Technology Collaboration Meeting (Virtual)

19th-21st January 2021
Superconductor- Insulator- Superconductor multistructures motivation and possible materials

Atomic Layer Deposition: Thermal and Plasma

Plasma Enhanced Atomic Layer Deposition: NbTiN thin films

Process Optimization of PEALD NbTiN

Effect of thermal treatment on NbTiN thin films
New structure proposed by A. Gurevich [1]
The idea is to coat the SRF cavities with alternating superconducting and insulating layers.
Requirements for the superconductor:
- Thin film thickness $<< \lambda_L$
- Higher Tc and $\Delta$
- Lower $\rho_n$
These multilayers provide magnetic screening of the bulk cavity and lower surface resistance which allows to increase the accelerating field and reduce the losses.

Motivation: S-I-S Multilayers

SUPERCONDUCTOR
Nb compounds: Nb$_3$Sn, Nb nitrides (NbN and NbTiN)
Others: MgB$_2$

INSULATOR
Al$_2$O$_3$
AlN
Nb$_2$O$_5$
MgO

Tc [K] : Nb=9.23 ; NbN=16.2 ; NbTiN=17.23
excellent superconducting properties of NbN with the good metallic and structural properties of TiN

AIN as buffer layer improves the NbTiN superconducting properties

Deposition Techniques
High power impulse magnetron sputtering
High Temperature Chemical Vapor Deposition
Atomic Layer deposition (ALD)

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Atomic layer deposition (ALD)

ALD is a sequential technique based on the self-limiting reactions between gases and solids

- **Exceptional conformality** (also on high-aspect ratio structures)
- **Precise thickness control** (constant growth per cycle (GPC))
- **Homogeneity** (pinhole-free)
- **Small film roughness**
  - Slow process
  - Not all stoichiometries are possible – correct precursors necessary
Thermal ALD

- **Metal chloride precursors** (NbCl$_5$ and TiCl$_4$) can *contaminate* the deposited film with chlorine
- NH$_3$ as nitrogen source is often *insufficient* as reductant power to obtain stoichiometric metal nitrides
- Requires *high ALD temperatures* (>400 °C)

**Plasma Enhanced ALD**

- **Highly reactive radicals** produced by a plasma source act as a coreactant
- **Low ALD deposition temperatures** (<400 °C)
- Metallorganic precursors
**Supercycle ALD Approach for AlN Nb\textsubscript{x}Ti\textsubscript{1-x}N**

**System**
- Arradiance GEMStar XT-P
- Dry pump (Ebara A10s)
- $P_{\text{base}} = 4 \times 10^{-2}$ mbar
- Ar (6.0) purge gas

**Precursor**
- TMA
- TDMAT
- TBTDEN

**Plasma**
- $N_2/H_2$-Plasma

**Aim**
- Supercycle ALD
- AlN
- TiN
- Nb\textsubscript{x}Ti\textsubscript{1-x}N

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Tuning the superconducting properties by varying ratio of NbN to TiN

EDX Composition Analysis

<table>
<thead>
<tr>
<th>TiN:NbN</th>
<th>At% Ti</th>
<th>At% Nb</th>
</tr>
</thead>
<tbody>
<tr>
<td>5:1</td>
<td>0.69</td>
<td>0.15</td>
</tr>
<tr>
<td>4:1</td>
<td>0.64</td>
<td>0.15</td>
</tr>
<tr>
<td>3:1</td>
<td>0.68</td>
<td>0.2</td>
</tr>
<tr>
<td>2:1</td>
<td>0.58</td>
<td>0.23</td>
</tr>
<tr>
<td>1:1</td>
<td>0.34</td>
<td>0.33</td>
</tr>
<tr>
<td>1:2</td>
<td>0.22</td>
<td>0.41</td>
</tr>
</tbody>
</table>

Increasing Nb-content in thin films → increase in $T_c$ and $H_c$
The deposition process has been optimized in order to improve the superconducting properties of NbTiN thin films.

- **Deposition Temperature**
  - @250°C Limitation of process

- **Plasma Parameters**
  1) **RF Forward Power**
  - @300W Limitation of setup

Luisa Ehmcke, UHH

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Process Optimization of individual binary processes

- Plasma Parameters
  2) Plasma Dose Time

![Resistivity vs Plasma Time plots for TiN and NbN](image)

Luisa Ehmcke, UHH

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**Plasma Parameters**

3) Gases Flow: $\text{H}_2 \& \text{N}_2 \rightarrow \text{H}_2=45 \; ; \; \text{N}_2=12$

- **Pressure:** decrease the base pressure improves the film quality

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Slight improvement in the superconducting properties optimizing $\text{Nb}_{0.66}\text{Ti}_{0.33}\text{N}$ deposition

↑ $T_c$ 10% **Before:** $T_c = 7.03 \; [K]$; $\rho = 1268 \; \mu\Omega\cdot\text{cm}$

↓ $\rho$ 30% **After:** $T_c = 7.78 \; [K]$; $\rho = 898 \; \mu\Omega\cdot\text{cm}$
Influence of temperature and time of the annealing process in the transport properties of the Nb$_{0.66}$Ti$_{0.33}$N films

Tc and resistivity were improved by RTA.
Influence of the gas atmosphere in the transport properties the Nb$_{0.66}$Ti$_{0.33}$N films

- Different annealing atmospheres:
  1. Ar/H$_2$ mixture (95% of Ar and 5% of H$_2$)
  2. N$_2$
  3. N$_2$/H$_2$ mixture (85% of N$_2$ and 15% of H$_2$)

- Investigation different RTA atmospheres

<table>
<thead>
<tr>
<th>Atmosphere</th>
<th>Resistivity / $\mu\Omega \cdot cm$</th>
<th>Tc</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar/H$_2$</td>
<td>152</td>
<td>13.05</td>
</tr>
<tr>
<td>N$_2$</td>
<td>198</td>
<td>12.7</td>
</tr>
<tr>
<td>H$_2$(15%)/N$_2$(85%)</td>
<td>186</td>
<td>12.03</td>
</tr>
<tr>
<td>H$_2$(33.3%)/N$_2$(66.6%)</td>
<td>302</td>
<td>11</td>
</tr>
</tbody>
</table>

1. After the first RTA, a 2nd thermal treatment (without vacuum break) in pure N$_2$ atmosphere has been performed at 1000°C. The results showed that Tc has been increased.

   The best result is $Tc = 13.93 \text{ K}$; $\rho = 132 \mu\Omega \cdot cm$

Further studies are needed to establish best recipe

Maximum Tc reached $Tc = 13.5 \text{ K}$
WORK UNTIL NOW

✓ PE-ALD NbTiN deposition process has been optimized for $T_c$ and resistivity, still insufficient.

✓ Post-deposition thermal annealing has been performed to investigate the effect on the films and different temperatures, annealing times and gas atmospheres have been studied.

As deposited $T_c=7.78$ K → After RTA $T_c=13.93$K

NEXT STEPS

- Lattice characterization, using XRR/XRD/EBSD/PALS. The aim is to fully understand the effect of the RTA
- Analyze the $H_2$ concentration using EMGA
- SRF measurements to obtain $H_{c1}$ and the superconducting gap $\Delta$


THANK YOU FOR YOUR ATTENTION!

Feel free to contact via: igonzale@physnet.uni-hamburg.de
### Backup

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_c$ [K]</th>
<th>$\rho_n$ ($\mu$Ω cm)</th>
<th>$H_c2$ (0) [mT]</th>
<th>$H_{c1}$ (0) [T]</th>
<th>$H_{c2}$ (0) [T]</th>
<th>$H_{c1}$ [T]</th>
<th>$\lambda$(0) [nm]</th>
<th>$\Delta$ [meV]</th>
<th>$\xi$ [nm]</th>
<th>Type</th>
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<tbody>
<tr>
<td>Nb</td>
<td>9.23</td>
<td>2</td>
<td>200</td>
<td>0.17</td>
<td>0.28</td>
<td>0.219</td>
<td>40</td>
<td>1.5</td>
<td>28</td>
<td>II</td>
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<tr>
<td>Pb</td>
<td>7.2</td>
<td>80</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
<td></td>
<td>48</td>
<td></td>
<td></td>
<td>I</td>
</tr>
<tr>
<td>NbN</td>
<td>16.2</td>
<td>70</td>
<td>230</td>
<td>0.02</td>
<td>15</td>
<td>0.214</td>
<td>200-350</td>
<td>2.6</td>
<td>&lt;5</td>
<td>II, B1 comp.</td>
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<tr>
<td>NbTiN</td>
<td>17.3</td>
<td>35</td>
<td></td>
<td>0.03</td>
<td></td>
<td></td>
<td>150-200</td>
<td></td>
<td>&lt;5</td>
<td>II, B1 comp.</td>
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<tr>
<td>Nb$_2$Sn</td>
<td>18</td>
<td>20</td>
<td>540</td>
<td>0.05</td>
<td>30</td>
<td>0.425</td>
<td>80-100</td>
<td>3.1</td>
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<td>V$_3$Si</td>
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<td>4</td>
<td>720</td>
<td>0.072</td>
<td>24.5</td>
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<td>Mo$_3$Re</td>
<td>15</td>
<td>10-30</td>
<td>430</td>
<td>0.03</td>
<td>3.5</td>
<td>0.17</td>
<td>140</td>
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<td></td>
<td>II, A15</td>
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<tr>
<td>MgB$_2$</td>
<td>40</td>
<td>0.1-10</td>
<td>430</td>
<td>0.03</td>
<td>3.5-60</td>
<td>0.17</td>
<td>140</td>
<td>2.3/7.2</td>
<td>2.3/7.2</td>
<td>II- 2 gaps</td>
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<tr>
<td>2H-NbSe$_2$</td>
<td>7.1</td>
<td>68</td>
<td>120</td>
<td>0.013</td>
<td>2.7-15</td>
<td>0.095</td>
<td>100-160</td>
<td>8-10</td>
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<td>II-2gaps</td>
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<tr>
<td>YBCO</td>
<td>93</td>
<td>1400</td>
<td></td>
<td>0.01</td>
<td>100</td>
<td>1.05</td>
<td>150</td>
<td>20</td>
<td>0.03/2</td>
<td>d-wave</td>
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<tr>
<td>pnictides</td>
<td>30-55</td>
<td>500-900</td>
<td></td>
<td>0.03</td>
<td>&gt;100</td>
<td>0.756</td>
<td>200</td>
<td>10.20</td>
<td>2</td>
<td>s/d wave</td>
</tr>
</tbody>
</table>

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Supercycle ALD Approach for Nb$_x$Ti$_{1-x}$N

Aim: Tuning the superconducting properties of the deposited thin by varying ratio of NbN to TiN