Performance Boundaries in Nb$_3$Sn Superconductors

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Acknowledgments

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Outline

- Critical current density and critical current

- Composition variation in Nb₃Sn wires

- Composition and $H_{c2}(T)$

- Pinning capacity, grain boundary pinning, grain size

- Composition and $J_c$

- Strain dependence ($time allowing$)

- Present status and future prospects
Wire $J_c$ progress versus time

![Graph showing $J_c$ non-Cu [kA/mm$^2$] versus year. The graph plots data points for years 1992 to 2004, showing an increasing trend with $\mu_0H = 12T$ and $T = 4.2K$.]

*Parrell, ACE 2004*
What determines $J_c$?

**Pinning capacity**

- Average grain size

**Effective $H - T$ phase boundary**

- Composition
- Strain state

$J_c \rightarrow I_c$?
What determines $I_c$?

- Powder-in-tube wire (SMI)

- 50% Non – Cu fraction

- Only 20% of the wire carries $J_c$
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Composition: $\text{Nb}_3\text{Sn} \rightarrow \text{Nb}_{1-\beta} \beta \text{Sn}_\beta$

- Binary phase diagram $\rightarrow$ 18 to 25 at.% Sn $\rightarrow$ ‘A15’

$\text{Charlesworth, JMS 1970, Flükiger, ACE 1982}$
Nb$_3$Sn diffusion reaction in wires

- Reaction at 675°C vs time in Powder-in-Tube wire (SMI)

![Image showing diffusion reaction process and products](image_url)
Composition variation in wires

- Composition analysis on SMI Powder-in-Tube wire

- \(0.3 \text{ at.\% Sn/}\mu\text{m}\)
- \(J_c(12\text{T},4.2) = 2250 \text{ A/mm}^2\)
Composition variation in wires

- Bronze process wire
  Univ. of Geneva

- 4 at.% Sn/μm
- $J_c(12T, 4.2) = 720$ A/mm$^2$

Abächerli, TAS 2005
Composition variation in wires

- OST Internal-Tin wire
- Flat Sn content at 24 at.%
- $J_c(12\text{T}, 4.2) = 3000 \text{ A/mm}^2$

Uglietti, MT19 2005
### Increasing $J_c$ with increasing Sn

<table>
<thead>
<tr>
<th>Process</th>
<th>Sn Concentration</th>
<th>Gradient</th>
<th>$J_c(12T,4.2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Geneva Bronze Process</td>
<td>25 at.% Sn @ source</td>
<td>4 at.% Sn/µm gradient</td>
<td>$J_c(12T,4.2) = 720$ A/mm²</td>
</tr>
<tr>
<td>SMI Powder-In-Tube</td>
<td>25 at.% Sn @ source</td>
<td>0.3 at.% Sn/µm gradient</td>
<td>$J_c(12T,4.2) = 2250$ A/mm²</td>
</tr>
<tr>
<td>OST Internal Tin</td>
<td>24 at.% Sn no gradient</td>
<td></td>
<td>$J_c(12T,4.2) = 3000$ A/mm²</td>
</tr>
</tbody>
</table>

Sn richer
Higher $J_c$
Why?
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- Composition and $H_{c2}(T)$
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What happens with changing Sn content?

- **Pure Nb**
  - *bcc* Nb spacing 0.286 nm
  - $T_c = 9.2$ K

- **Nb$_3$Sn → A15 unit cell**
  - *bcc* Sn, orthogonal Nb chains
  - Nb spacing 0.265 nm
  - High peaks in d-band DOS
  - Increased $T_c = 18$ K

- **Off-stoichiometry**
  - Sn vacancies unstable
  - Excess Nb on Sn sites
    - Additional d-band
    - Less electrons for chains
    - Rounded off DOS peaks
    - Reduced $T_c$

**Dew-Hughes, Cryogenics 1975**
Nb chain continuity, $N(E_F)$, $\lambda_{ep}$, $T_c$, $H_{c2}$

In general

- Sn deficiency
- Tetragonal distortion
  - 24.5 – 25 at.% Sn
- Strain
- Alloying (Ti, Ta, …)
- Dislocations
- Anti-site disorder

All affect Nb chain integrity (‘Long Range Order’)

- And thus $N(E_F)$ and $\lambda_{ep}$
- And thus $T_c$ and $H_{c2}$
$T_c$ and $H_{c2}$ versus Sn content

Single crystal, bulk and thin film samples

$$T_c(\beta) = \frac{-12.3}{1 + \exp\left(\frac{\beta - 0.22}{0.009}\right)} + 18.3$$

$$\mu_0 H_{c2}(\beta) = -10^{-30} \exp\left(\frac{\beta}{0.00348}\right) + 577 \beta - 107$$
\( H_{c2}(T) \) versus Sn content

- Jewell, ACE 2004, bulk samples

- Sn richer A15 has higher \( H_{c2}(T) \) (until \( \sim 24.5 \) at.% Sn)
$H_{c2}(T)$ in wires

- $H_{c2}(T)$ from small current, resistive transitions

Graphs showing the upper critical field $H_{c2}$ as a function of temperature for different materials:

- SMI Binary PIT
  - 10-90%: $H_{c2}(T_{c}) = 0.8T$
  - $T_{c}(0) = 26.6-27.4T$

- SMI Reinforced ternary PIT
  - 10-90%: $H_{c2}(T_{c}) = 0.9T$
  - $T_{c}(0) = 28.2-29.1T$

- Funakawa Ternary bronze
  - 10-90%: $H_{c2}(T_{c}) = 0.9T$
  - $T_{c}(0) = 28.0-28.9T$

- UW-ASC Binary bulk
  - 10-90%: $H_{c2}(T_{c}) = 1.8T$
  - $T_{c}(0) = 26.5-28.3T$

Inset diagram showing the resistivity $\rho(H)$ with 99% and 1% normal state at $\mu_0H$.
Normalized $H_{c2}(T)$ all available results

- **Shape $H_{c2}(T)$ independent of**
  - Composition
  - Morphology
  - Strain state
  - Applied critical state criterion

\[ \ln \left( \frac{T}{T_c(0)} \right) = \psi \left( \frac{1}{2} \right) - \psi \left( \frac{1}{2} + \frac{hD\mu_0H_{c2}(T)}{2\phi_0k_BT} \right) \]

Approximation:
\[ \frac{H_{c2}(t)}{H_{c2}(0)} \approx 1 - t^{1.52}, \quad t = \frac{T}{T_c(0)} \]
Highest $H_{c2}(T)$ in wires

$\mu_0 H_{c2}(0) = 30 \, \text{T}, \ T_c(0) = 18 \, \text{K}$ is upper limit
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Pinning: Why does Nb$_3$Sn need it?

- Nb$_3$Sn slab in $H_{c1} < H < H_{c2}$
- Field quanta $\phi_0 = h / 2e$ (flux-lines) penetrate slab

Transport current ($\nabla \times B = \mu_0 J$) causes gradient $B_x$

Flux-lines repel → move ($\nabla \times E = -dB/dt$) → $E_y$ → Loss
  - Need to be ‘pinned’ at ‘pinning centers’ by ‘pinning force’ $F_p$

Optimal pinning at 1 pinning center / flux-line
What determines pinning capacity?

Pinning centers

- Positions with minima in SC wave function
  - Normal regions
  - Grain boundaries
  - Lattice imperfections
  - …

- $\text{Nb}_3\text{Sn}$
  - Grain boundaries
    - Main pinning centers
  - Grain size determines $F_{\text{Pmax}}$
What determines grain size?

- Presence of grain nucleation points
- Reaction time and temperature
What is an optimal grain size?

Ideal: One pinning center per flux-line → $a_\Delta \approx d_{av}$

- Flux-line spacing → field dependent
  - E.g. at 12 T $a_\Delta = (4/3)^{1/4}(\phi_0/\mu_0 H)^{1/2} = 14$ nm
  - Grain size in Nb₃Sn wires → 100 – 200 nm
  - Order of magnitude from optimal

- For any practical field $a_\Delta < d_{av}$
  - Collective pinning (‘shearing’ of FLL)
  - $a_\Delta \rightarrow d_{av}$ only for $\mu_0 H << 1$ T

- NbTi in contrast
  - Nano-scale distribution of $\alpha$-Ti precipitates
  - $a_\Delta \approx \alpha$-Ti distribution for application fields
  - NbTi is fully optimized
What does $a_\Delta \ll d_{av}$ mean in practice?

- **De-pinning → Synchronous shearing of FLL**
- $F_{P_{\text{max}}}$ at $H/H_{c2} = 0.2$
  - About 6 T for Nb$_3$Sn
  - Far below application fields
- **Grain refinement / APC**
  - $F_{P_{\text{max}}}$ to higher field
  - $F_{P_{\text{max}}} \rightarrow H/H_{c2} > 0.4$ shown by Cooley, ACE 2002
  - Higher fields accessible with Nb$_3$Sn
- **Much room for improvement!**

- **Example: Bronze processed ITER wire (Furukawa)**

![Graph showing reduced pinning force vs. reduced magnetic field](image)
Alternative presentation $a_\Delta << d_{av}$

- Flux shear model
  - Kramer JAP 1973

\[
F_p(H) = 12.8 \frac{(\mu_0 H_{c2})^{2.5}}{\kappa_1^2} \frac{h^{0.5} (1-h)^2}{(1-a_\Delta(H)/d_{av})^2}, \quad h = \frac{H}{H_{c2}} \quad \text{[GN/m}^3]\]

\[
J_c^{0.5} (\mu_0 H)^{0.25} = \frac{1.1 \times 10^5}{\kappa_1} \frac{\mu_0 (H_{c2} - H)}{(1-a_\Delta(H)/d_{av})}
\]

- $a_\Delta << d_{av}$: Kramer plot

\[
f_K(H) \equiv J_c^{0.5} (\mu_0 H)^{0.25} \approx \frac{1.1 \times 10^5}{\kappa_1} \frac{\mu_0 (H_{c2} - H)}{\kappa_1} \mu_0 (H_{c2} - H) \quad \therefore \quad f_K(H) \propto H
\]

- Linear in $H$
‘Kramer’ plot

Plot of $f_K(H)$ at various temperatures
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Are Kramer plots linear?

\[ F_p(h) = 12.8 \left( \frac{\mu_0 H_{c2}}{\kappa_1^2} \right)^{2.5} h^{0.5} (1-h)^2 \quad a_\Delta \ll d_{av} \]

\[ F_p(h) \approx F_{pmax} h^p (1-h)^q \quad p = 0.5, \quad q = 2 \]

- **Linearity from** \( h \approx 0.03 \) to 0.8
  - Confirmed by measurements
- \( a_\Delta \approx d_{av} \) only below \( h \approx 0.03 \)
- Different pinning mechanism?
  - only below \( h \approx 0.03 \)

- **Non-linearity below** \( h \approx 0.03 \)
  - Different pinning mechanism
- **Non-linearity above** \( h \approx 0.8 \)
  - Inhomogeneity artifacts
  - Averaging over \( H_{c2} \) distribution
Effective $H_{c2}(T)^*$ for $J_c$

$J_c$ scales with ‘some’ average $H_{c2}(T)^*$

- $J_c$ gain if all A15 is stoichiometric?

$J_c(12T,4.2K)$
- From 2250 A/mm² to 2900 A/mm²
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Strain sensitivity of $H_{c2}(T)$

- Longitudinal strain effects on effective $H_{c2}(T)^*$

Strain and composition have similar effects
- Need for a separation of parameters
Strain sensitivity of $J_c(H, T)$

- $J_c(10 \text{T}, T, \varepsilon_{\text{axial}})$
- $J_c(H, 8 \text{K}, \varepsilon_{\text{axial}})$

Why is strain sensitivity increased at higher $H$ and $T$?
Strain sensitivity versus composition

At higher $H$ and $T$

- Low Sn A15 sections “die out”
  - Benefit PIT and IT vs Bronze:
    - Larger volume fraction high Sn
  - High Sn sections determine SC properties
- Increased strain sensitivity
  - Is Sn rich A15 more strain sensitive than Sn poor A15?

Does wire optimization through Sn enrichment cause higher strain sensitivity?
Strain sensitivity versus LRO

- $S \rightarrow$ Bragg-Williams order parameter

- Higher LRO ($\wedge$ more Sn) $\rightarrow$ larger strain sensitivity

Flükiger, ACE 1984
Strain in ternary and binary wires

- Alloyed → more disorder → reduced strain sensitivity?
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Prospects for critical current density

Simulations on SMI-PIT Nb(Ta)\textsubscript{3}Sn assuming all have same pinning

- **Pinning?**
  - SMI-PIT grains ~ 140 nm
  - OST-IT grains ~ 170 nm
  - 12 T $\rightarrow a_\Delta = 14$ nm
- **Large gains possible**

- 5000 A/mm\textsuperscript{2} (+65%) physical limit with present wire designs?
  - Unless pinning is improved
- 4000 A/mm\textsuperscript{2} realistic optimization goal?
Summary

Wire optimizations past decade

- Sn enrichment
- A15 fraction in non-Cu optimization
- Physical limit 5 kA/mm², realistic limit 4 kA/mm²

Grain refinement / APC

- The next big step?
- Grain size one order above optimal
- Grain 10 – 20 nm desired → nano technology

Strain

- Strain and composition parameter separation needed
- Sn enrichment = more strain sensitivity?
- Much work to be done (3D, theory, bulk, film, …)
More information

Available on request → agodeke@lbl.gov
Optional theory section
\[ N(E_F) \text{ and } \lambda_{ep} \rightarrow T_c \text{ and } H_{c2} \]

- **Weak coupling (BCS based)**
  
  \[ T_c(0) \approx \frac{2e^{\gamma_E}}{k_B} \hbar \omega_c \exp \left[ -\frac{1}{V_0 N(E_F)} \right] \quad \therefore \quad T_c(0) \approx 1.134 \Theta_D \exp \left[ -\frac{1}{\lambda_{ep}} \right] \]

  \[ \mu_0 H_{c2}(0) \approx k_B e N(E_F) \rho_n T_c(0) = \frac{3e}{\pi^2 k_B} \gamma \rho_n T_c(0) \]

- **Interaction strength independent (Eliashberg based)**

  \[ \lambda_{ep} = 2 \int \frac{\alpha^2(\omega)F(\omega)}{\omega} d\omega \quad \lambda_{eff} = \frac{\left( \lambda_{ep} - \mu^* \right)}{\left( 1 + 2\mu^* + 1.5\lambda_{ep}\mu^* e^{-0.28\lambda_{ep}} \right)} \]

  \[ T_c = \frac{0.25 \langle \omega^2 \rangle^{\frac{1}{2}}}{\left( e^{2/\lambda_{eff}} - 1 \right)^{\frac{1}{2}}} \quad \mu_0 H_{c2} = \ldots \]
Is Nb₃Sn weak or strong coupling?

- Moore, PRB 1979, thin film samples

Weak coupling below 23 – 24 at.% Sn
Strong coupling approaching stoichiometry
Applicable theory

\[ N(E_F) \text{ and } \lambda_{ep} \rightarrow T_c \text{ and } H_{c2} \]

- Wires \( \rightarrow 18 \text{ – } 25 \text{ at.\% Sn, polycrystalline} \)
- Interaction strength independent theory
- Not done for entire composition range
- \( N(E_F) \text{ and } \lambda_{ep} \rightarrow T_c \text{ and } H_{c2} \) remains empirical

Promising recent work

- Eliashberg-based description of \( T_c(\varepsilon) \text{ and } H_{c2}(\varepsilon) \)
  - Markiewicz, Cryogenics 2004
  - Oh, JAP 2006