Essential Features of LARP Strands and Prospects for Still Better High-Field Superconductors

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Outline

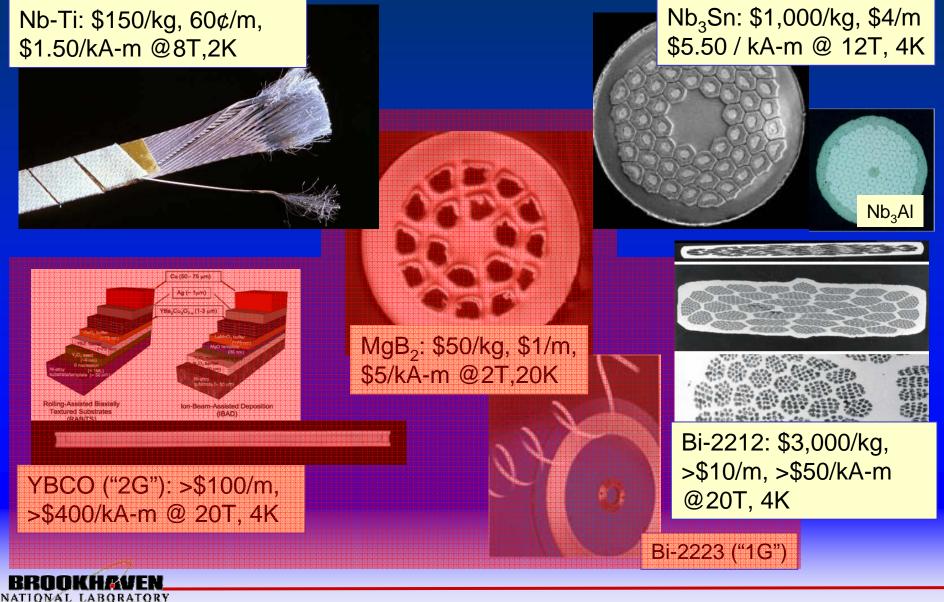
- Factors that limit performance of superconductors
- Nb₃Sn: Why has RRP* emerged over bronze-route, MJR[†], and PIT[‡] as the LARP strand process?
 - Maximizing current density
 - Overcoming limitations imposed by gradients of the tin content
 - Managing stability
- Is further improvement of Nb₃Sn possible?
- If not, what's next? Clues from emerging areas in superconducting materials for HEP
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‡ Powder-in-tube process

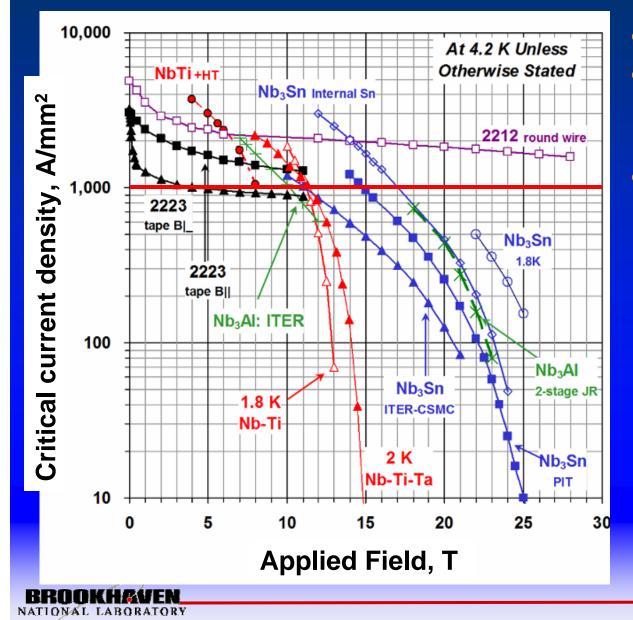


Our portfolio of long-length superconductors



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Long-length Superconductor Performance



- J_c, J_c, J_c ...
- Plot shows J_c over wire area that is not stabilizer or support
- Highest non-copper area fractions:
 - Nb-Ti ~65%
 - Nb₃Sn ~55%*
 - Bi-2212 ~35%
 - Bi-2223 ~35%
 - MgB₂ ~35%
 - YBCO ~1%
 - Includes Sn and Cu used to convert Nb to Nb₃Sn. Nb₃Sn itself is ~35% of wire area

Plot by Peter Lee http://magnet.fsu.edu/~lee/

Performance has limits at multiple levels

- >100,000 A/mm² : Depairing current limit (T_c and λ_L) Cumulative losses that limit current density in practical forms
- Material & physics factors
 - Flux pinning at high fields: 90 to >99% lost
 - Material anisotropy: large losses in \perp applied fields and at high T
- Factors related to forming the superconductor
 - Diffusion barriers: 2 to 10%
 - Materials needed to form the superconducting phase: 0 to >90%
 - Obstacles and grain connections: 0 to >90%
- Engineering factors
 - Strain, mechanical degradation: 0 to 20% lost
 - Stabilization, mechanical support: 30 to 60% lost



Example: Effects of cumulative limitations on LHC magnet strands

Nb-Ti LHC strand

- $J_D \approx 150,000 \text{ A/mm}^2$
- Flux pinning: 97% lost @ 8T, 2K
- Diffusion barrier: 4% lost
- Copper: 50% lost
- Manufacturing: 25% lost
- Total current density in strand: 150,000 x 0.03 x 0.96 x 0.5 x 0.75 ≈ 1,600 A/mm²

- Nb₃Sn LARP strand
- $J_D \approx 400,000 \text{ A/mm}^2$
- Flux pinning: 98% lost 12T, 4.2K
- Nb-Sn reaction: 30% lost
- Diffusion barrier: 10%
- Bronze (or copper + tin) & manufacturing: 40%
- Copper: 50%
- Total current density in strand: 400,000 x 0.02 x 0.7 x 0.9 x 0.6 x 0.5 ≈ 1,500 A/mm²



Lessons from these analyses

- 1. The superconductor area must be maximized.
 - The superconductor often is formed during a later reaction.
- 2. Manufacturing losses should be minimized.
 - Must make long continuous pieces.
 - Must also control shape distortions and other geometric factors.
 - Pure metals have the most ductility!
 - Except for Nb-Ti, high-field superconductors are brittle.
- 3. All processing must serve the optimization of flux pinning.
 - a. The superconducting properties must be as good as possible.
 - b. The defect nanostructure must be preserved.

Note: (a) and (b) often produce conflicting processing requirements!



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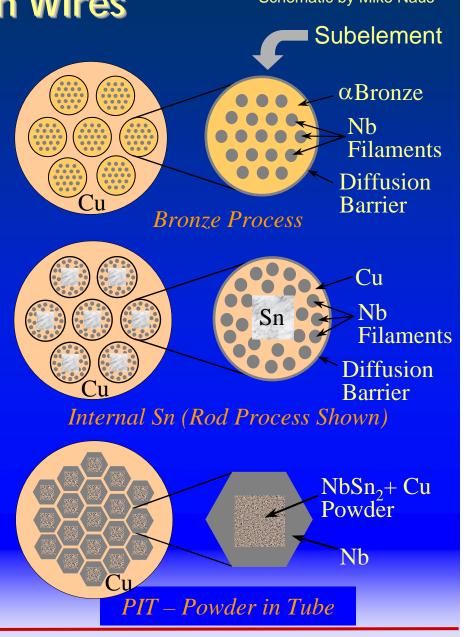


Solid State Routes to Nb-Sn Wires

Schematic by Mike Naus

Bronze route

- Alpha bronze is ductile, but must be annealed often
- 1-10 µm Nb filaments possible
- Internal Sn conductors
 - Start from Cu, Sn, Nb alloy
 - Components are more ductile than bronze, but pure Sn limits processing temperature
- Powder in Tube (PIT)
 NbSn₂ inside Nb tubes





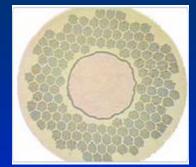
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Nb₃Sn state of the art, c.1998 VLHC workshop

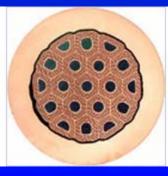
- Bronze-route: ~1,000 A/mm² available in long length at >\$15 per kA-m
- Modified Jelly-Roll (MJR) internal-tin composite: >1,500 A/mm² available in long length at ~\$10 / kA-m
- ITER-style internal tin composite modified for >1,500 A/mm²
- New player: Powder-in-tube

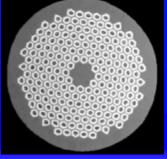
Photos courtesy of Jeff Parrell and Chad Fischer











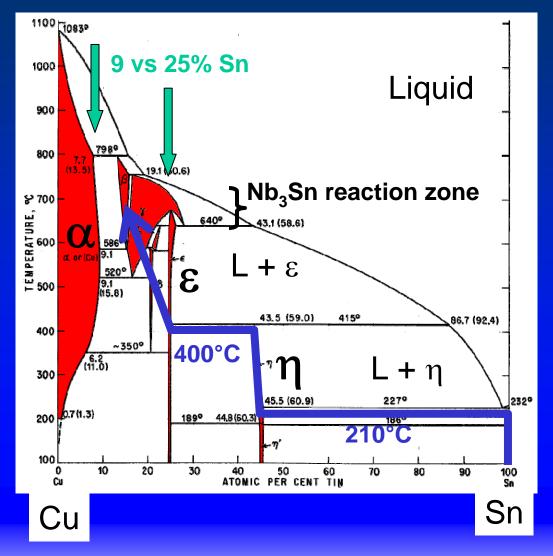
HEP specifications for VLHC (and LARP) strand

- Nb₃Sn
- J_c = 3,000 A/mm² (non-Cu) @ 12 T, 4.2 K
- Total HT time < 200 hrs</p>
- Effective filament diameter (d_{eff}) < 40 μ m
- Average piece length > 10 km at 0.3 to 1.0 mm diameter
- Cost less than \$1.50 / kA-m @ 12 T, 4.2 K



Consequences of Cu-Sn Phase Diagram

- Internal tin routes provide more tin than bronze route
 - Improved tin activity
 - Faster reactions
 - Less wire area required to supply tin atoms
- Reaction strategy avoids melting



Initial response fell short ...

- Subelement redesigned: less Cu, more Nb
- More tin added
- Filaments allowed to merge (so d_{eff} = d_{sub})
- Diffusion barrier reacted about halfway through
- Result: 2,000 to 2,200
 A/mm², slightly higher cost (but same \$/kA-m)
- Redesign needed \Rightarrow RRP

The Modified Jelly Roll Process

A. Copper shield.
Expanded niobium mesh.
Solid tin core rod.
B. Jelly roll formed above is inserted in outer copper tube.
Billet is formed ready for drawing.

C. Billet has been drawn to less than 1/2 inch in diameter.

D. Rods, approximately 1/2 inch in diameter, are cut to shorter lengths and rebundled in another copper tube.

E. Up to 50 additional drawing steps reduce material to wire 1/25 inch to 1/100 inch in diameter.

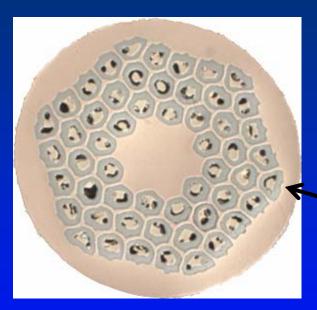
Nb diffusion barrier.



Original niobium expanded mesh becomes extremely long filaments in the finished wire. A single diamond shaped segment will be reduced from 1/4 to 1/3000 inch in cross section and elongated from one inch to one mile.



The LARP strand: Re-stacked Rod Process RRP™



Nb_{0.73}Ta_{0.02}Sn_{0.25} RRP 8220 54 subelements of 61 Cu ~ 48% $d_{\rm s}$ ~ 69 μ m @ 0.7 mm Ø Subelement combines materials with like surfaces (Cu), results in good bonding and long piece length

ot extruded ell-bond<mark>ed</mark> Cu filament Nb diffusio Sn rod in Cu tube \vdash 1 mm -



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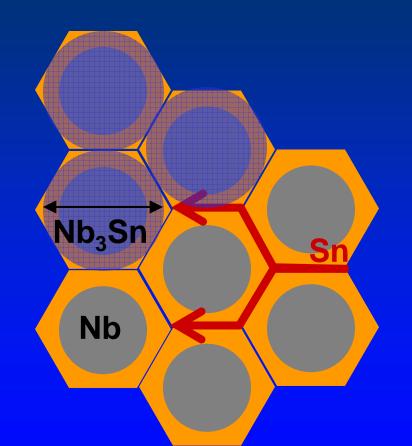
RRP strand gets closer to HEP goals

- Max $J_c \sim 3100 \text{ A/mm}^2$ at 12 T, 4.2 K
- Piece lengths are many kilometers
- Cost \$5.50 / kA-m at 12 T, 4.2 K
- Total reaction time typically 160 hrs 48 h @210°C + 48 h @400°C + 48 h @665°C, with ramp @50°C/h
- Effective filament diameter d_{eff} ~ 70 µm for 54/61 stack Further progress discussed shortly

RRP is the engineering strand we need for LARP R&D!!

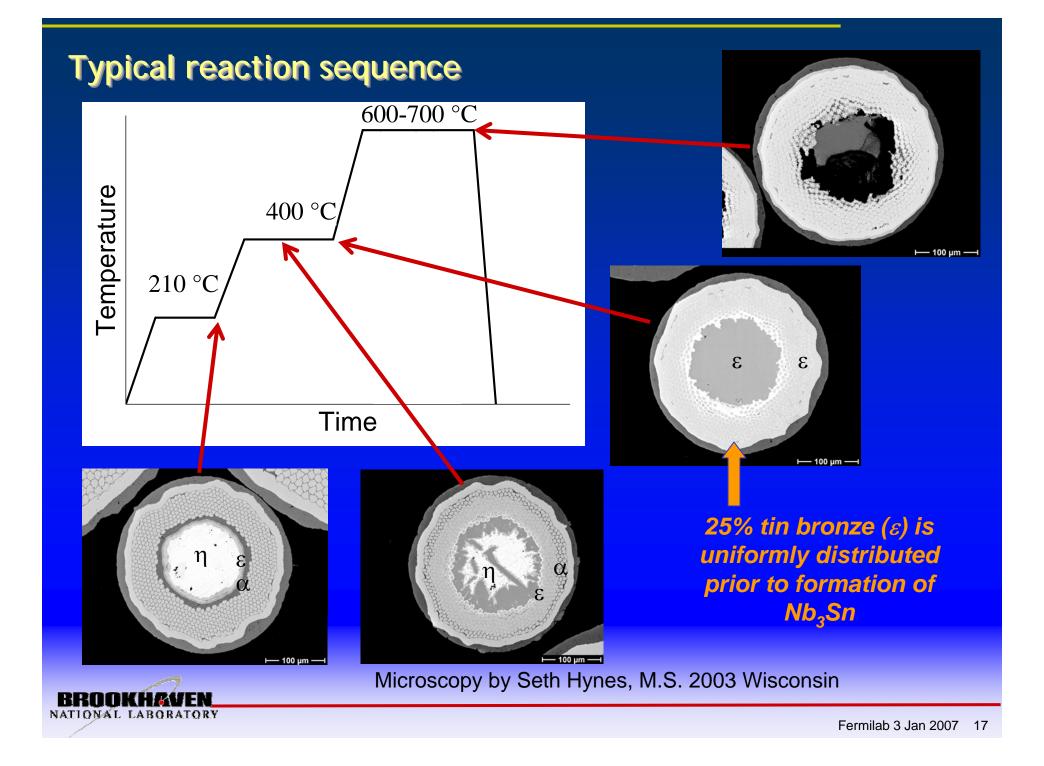


RRP provides optimum control over Cu:Nb Local Area Ratio (LAR)



- Lower LAR: more Nb₃Sn is possible if tin can get to Nb
- Higher LAR: wider diffusion pathways for tin
- Shape control reduces pinch-off as filaments grow and gives better balance of opposed trends above
 - Nb increases area by 37% upon conversion to Nb₃Sn





RRP addressed critical current lessons

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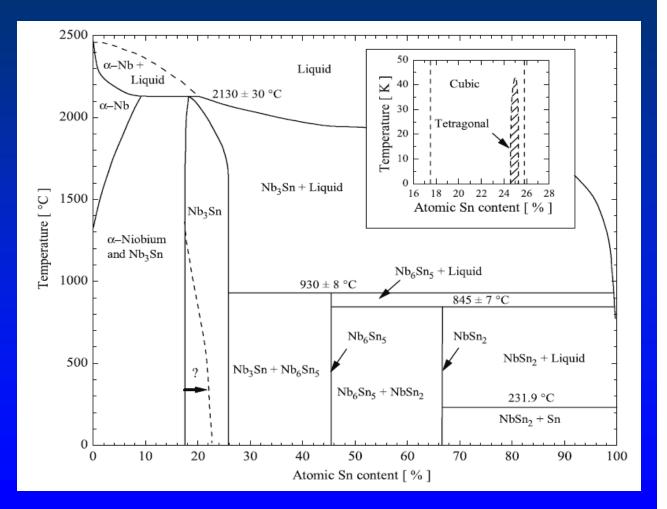
‡ Powder-in-tube process



Consequences of Nb-Sn phase diagram

- The desired Nb₃Sn phase exists from 18 to 25.5% Sn
- Thermo: Any solidstate reaction MUST produce a variation of %Sn across the SC layer
- Must have high kinetics: Variations in %Sn then occur steeply across small regions

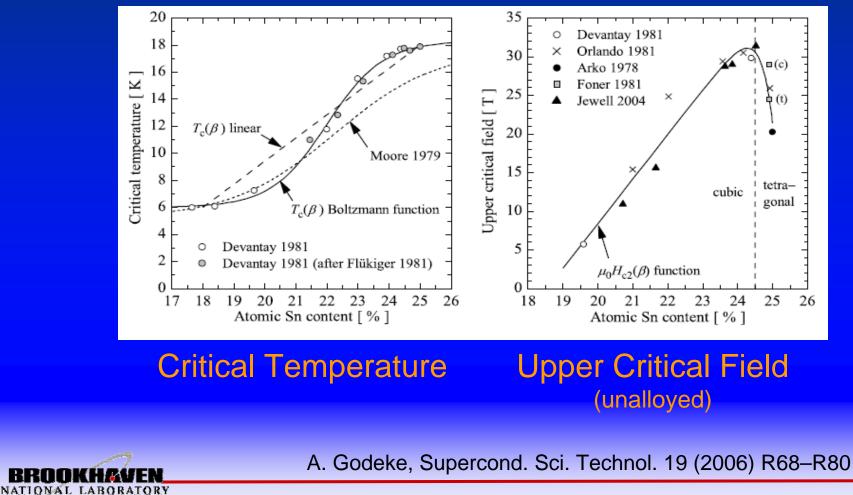
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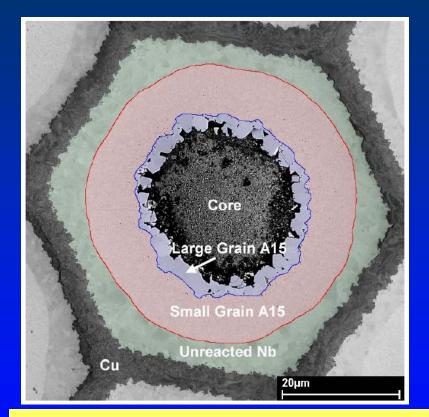
A. Godeke, Supercond. Sci. Technol. 19 (2006) R68-R80

Composition variation leads to property variation

- More tin = better superconductor
- What are the consequences for strands?



PIT Nb₃Sn wires are a model system to study Sn gradients



Lowest %Sn is on outside ⇒ transparent to magnetic probes! PIT is RRP in the limit of no copper (LAR = 0)

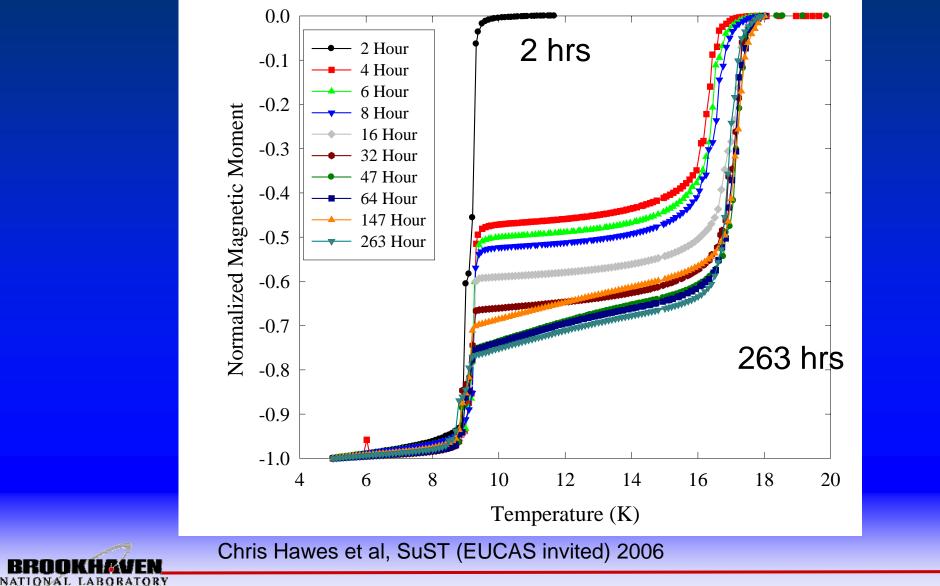
- Tin diffuses radially outward
- A new morphology forms: large grains near core
 - Large grains have poor flux pinning and do not contribute to current-carrying area

Strands courtesy Jan Lindenhoevius, SMI Photos courtesy Chad Fischer (M.S. Thesis, Wisconsin 2002) and Peter Lee (see magnet.fsu.edu/~lee)



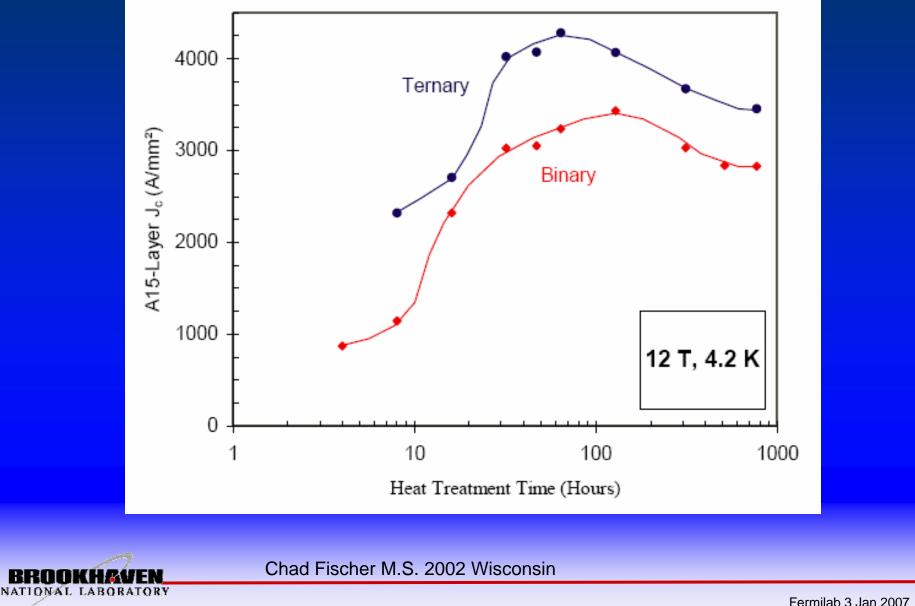
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Longer time @ 675°C: more Nb₃Sn, higher Tc



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Longer time @ 675°C: flux pinning peaks, drops



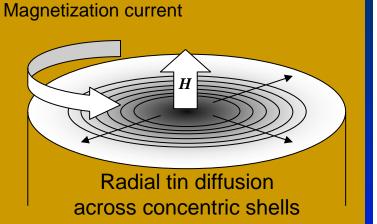
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PIT Lesson 1: Optimization of performance is complex!

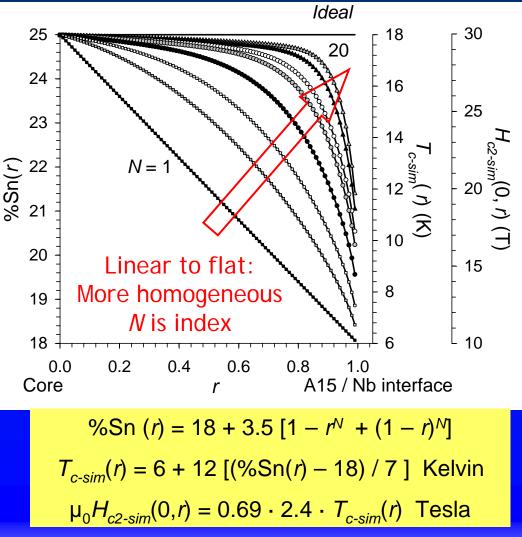
- Longer time @ temperature, or higher temperature at fixed time...
 - ... Produces more superconductor
 - ... Produces higher T_c and B_{c2} , which increase flux pinning
 - ... Eventually produces larger grains, which reduces flux pinning and the $\rm J_c$ of the $\rm Nb_3Sn$ layer
- Overall $J_c(T,H) = J_c$ layer \times amount of Nb₃Sn
- Mapping of optimum time / temperature matrix can be different for each wire
 - Tight geometry control is needed for reproducible HT recipe



Simulated tin gradients in PIT Nb₃Sn wires



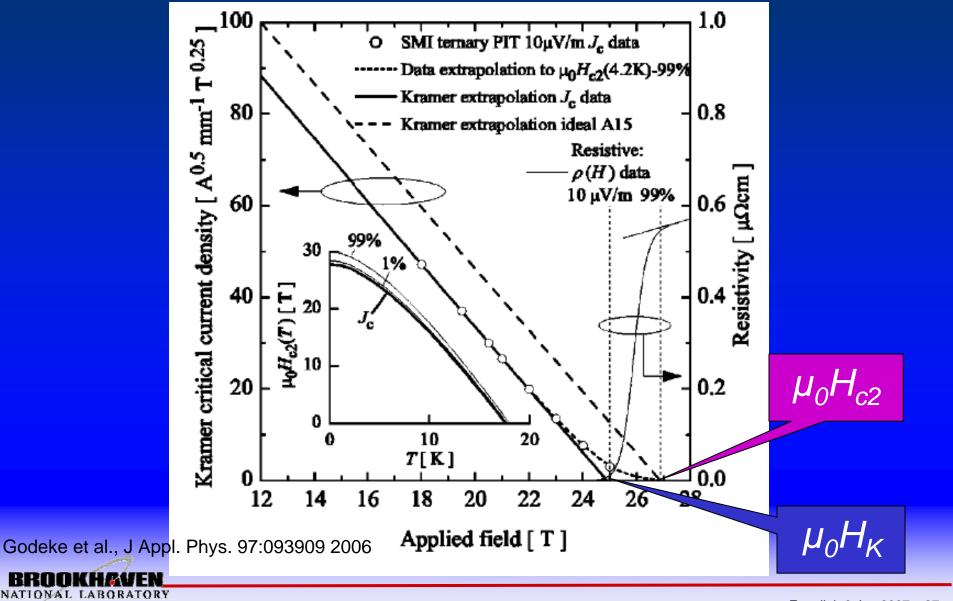
- Divide %Sn(r) profile into 100 points
 - Concentric shells of magnetization current
- From %Sn(r) get T_c(r), H_{c2}(0,r)



L. D. Cooley, C. M. Fischer, P. J. Lee, and D. C. Larbalestier, J. Appl. Phys. 2004.



Kramer plot extrapolation field $\mu_0 H_K$



Kramer extrapolation represents average of H_{c2} (%Sn)

Flatter profiles	Gradient index N	Weighted mean of H _{c2-sim} (r) at 4.2 K (T)	<i>H_K</i> at 4.2 К (Т)	Weighted mean of H _{c2-sim} (r) at 12 K (T)	<i>H_K</i> at 12 К (Т)
	1	15.7	19.1	7.1	9.6
	5	23.0	23.3	10.5	11.4
U U U	10	24.6	24.6	11.7	12.1
7	Ideal	26.5	26.4	13.6	13.6
This is the average H_{c2} (%Sn), which is always less than the ideal value (H_{c2} @ 25% Sn). This is the Kramer plot extrapolation that fixes Jc(H)					
DOONNERSON L. D. Cooley, C. M. Fischer, P. J. Lee, and D. C. Larbalestier, J. Appl. Phys. 2004.					
NATIONAL LABORATORY					

PIT Lesson 2: %Sn profile is averaged by critical current

- To improve performance at high fields, it is absolutely necessary to minimize the number of tin-poor regions
- $J_c(H)$ scales with $H_K = average H_{c2}(\%Sn)$
- RRP advantage over PIT: Cu provides rapid pathways for radial diffusion of Sn, eliminating radial Sn gradients
 - Because Cu in RRP diffuses toward the tin core, practically no tinpoor Nb₃Sn area results. Thus, RRP produces better Nb₃Sn AND does it more efficiently.
 - By contrast, the large-grain region wastes ~20% of the Nb₃Sn layer in PIT, even though it is rich in Sn.



Summary of RRP advantages

- Well bonded materials give long pieces
- Control and flexibility of LAR permits systematic adjustment of tin diffusion
- High tin activity produces rapid reactions and minimizes extent of tin-poor regions
- Radial dispersion of tin prior to formation of Nb₃Sn reduces gradients



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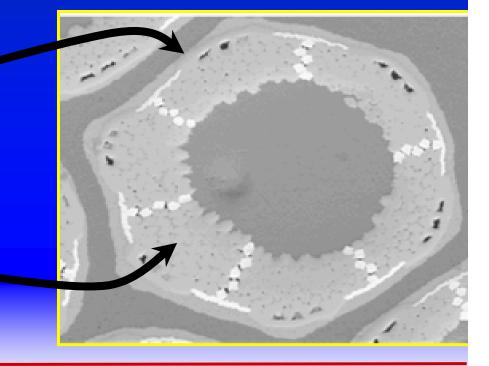
Origins of strand instability

- Large d_{eff} makes wires prone to flux jumps
- If heat released by flux jump cannot escape to helium in time (e.g. due to dirty copper), strand will quench
- Many magnets can operate with unstable strands!

Tin may breach the barrier and contaminate copper

Filaments merge into a solid superconducting mass after reaction

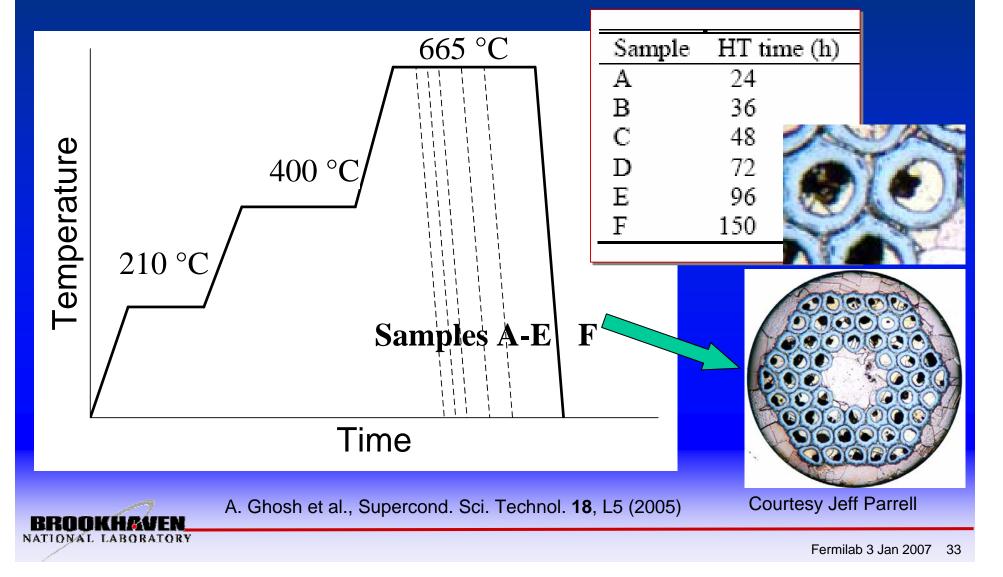
(Sometimes separators are incorporated to break up the filament ring, as shown here)



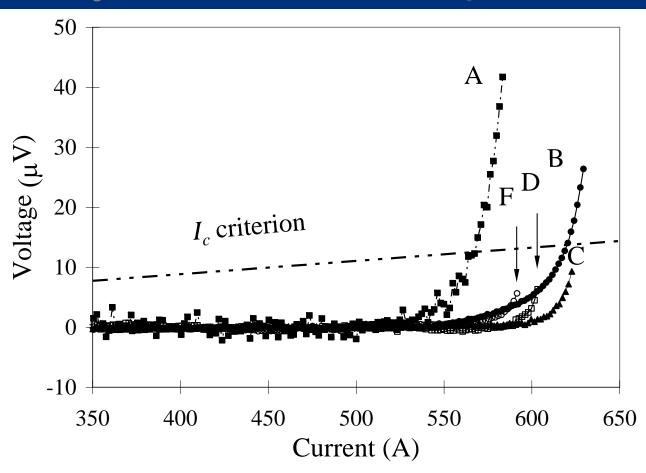


Experiment - RRP 7054 (LARP predecessor)

Limit reaction to keep tin from leaking through Nb barrier



High tin activity helps J_c reach 90% of the maximum J_c for a reaction of only 24 hours

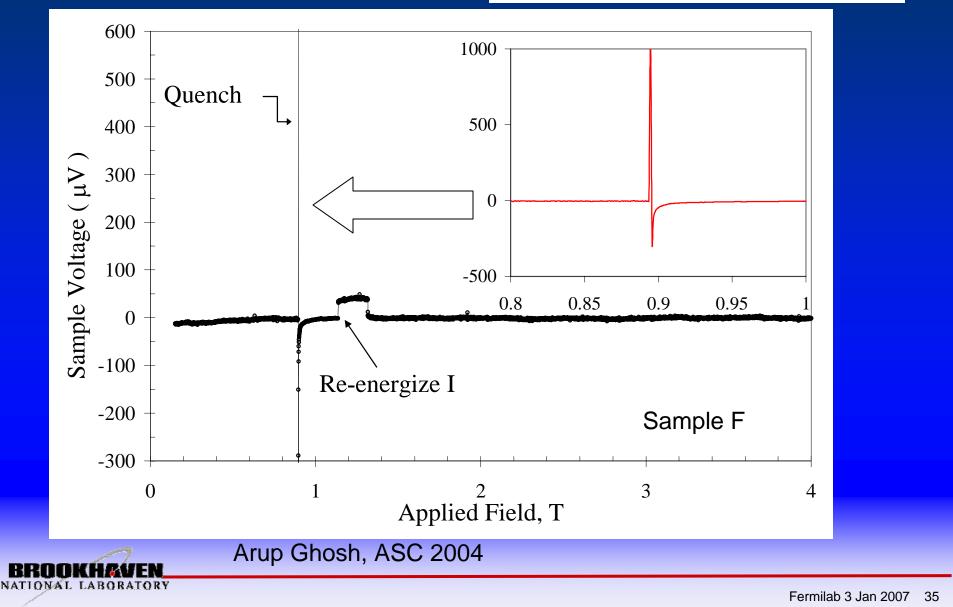


Voltage-current transitions acquired at 11.5 T for samples A-F. The resistivity criterion used to determine I_c is also shown.

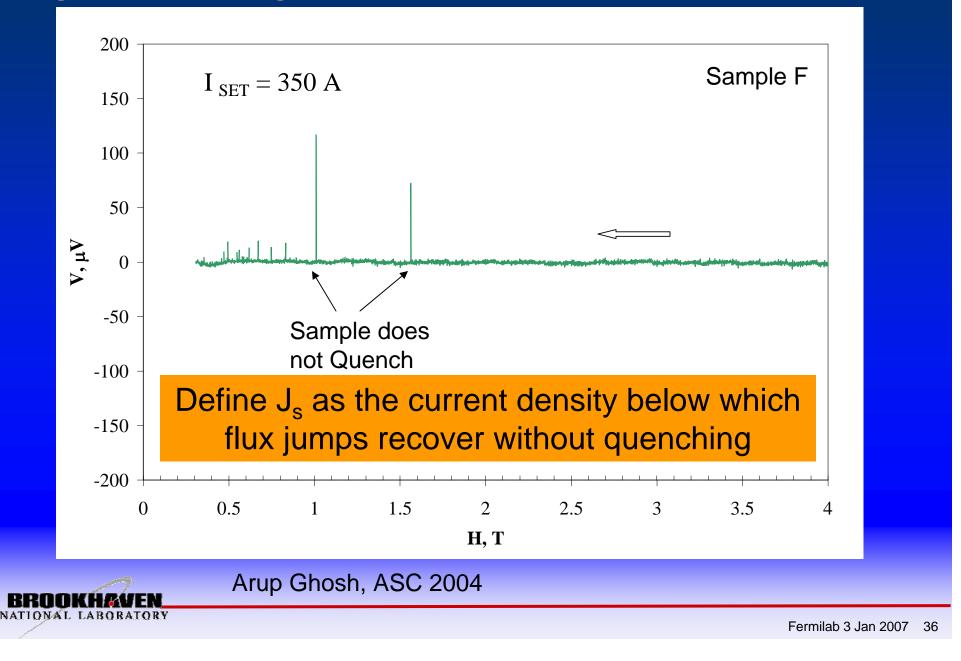
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Non-Traditional V-H plot

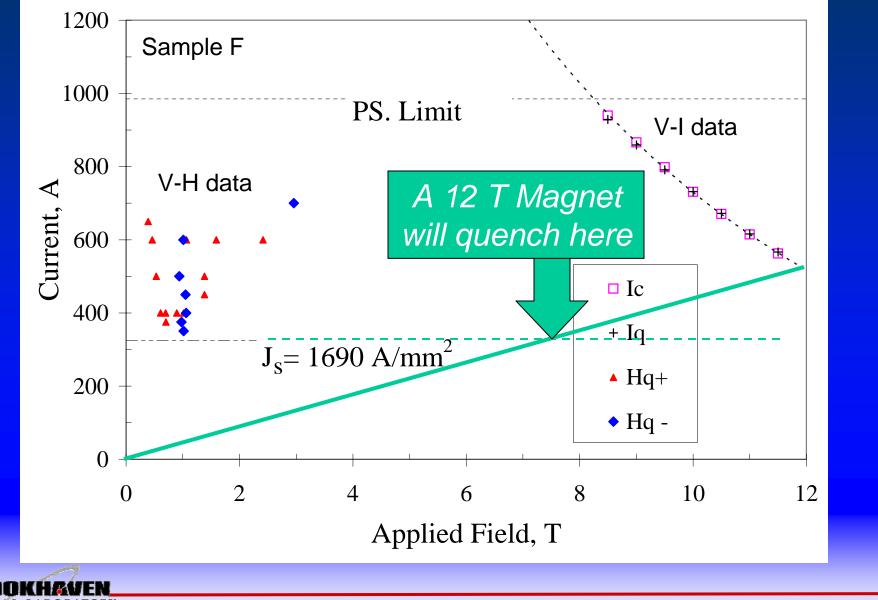
I_{SET} =400 A d*B*/d*t* = 5 mT/s



Dynamic stability threshold

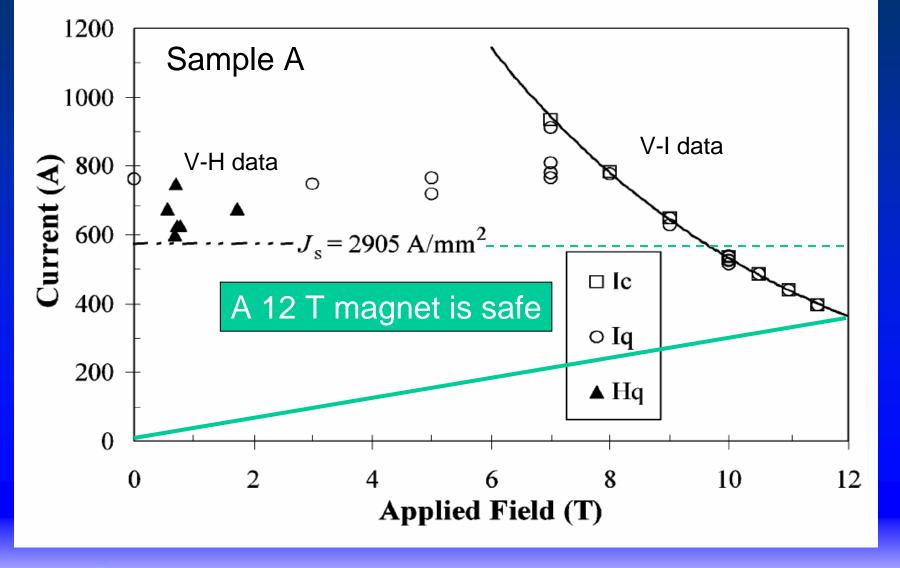


J_s is far below $J_c(12 T)$ when RRR is low



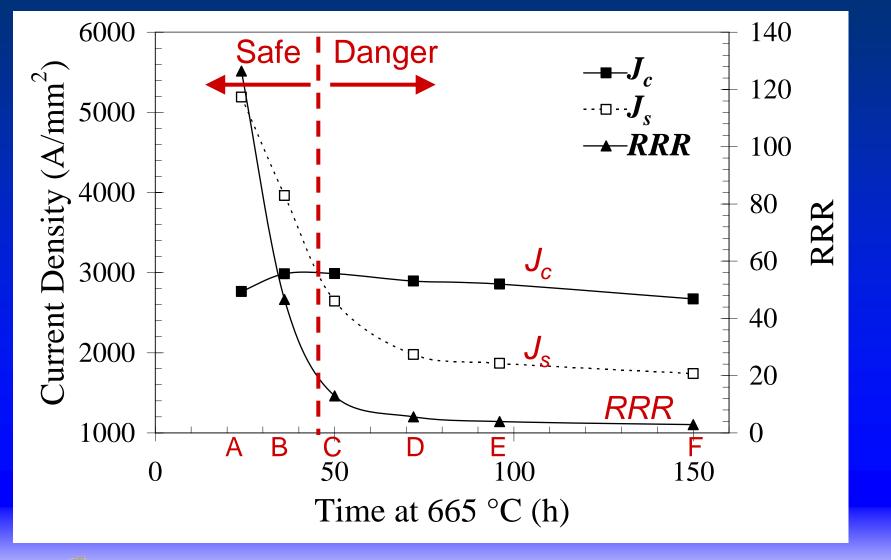
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J_s is above $J_c(12 \text{ T})$ when RRR is high





Experiment Summary: Keep J_s > J_c



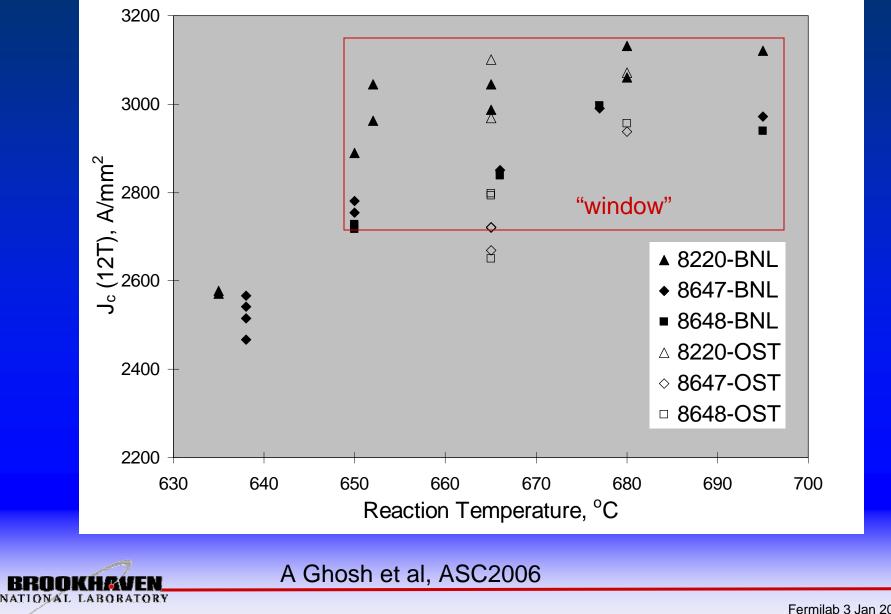


Lessons from stability work

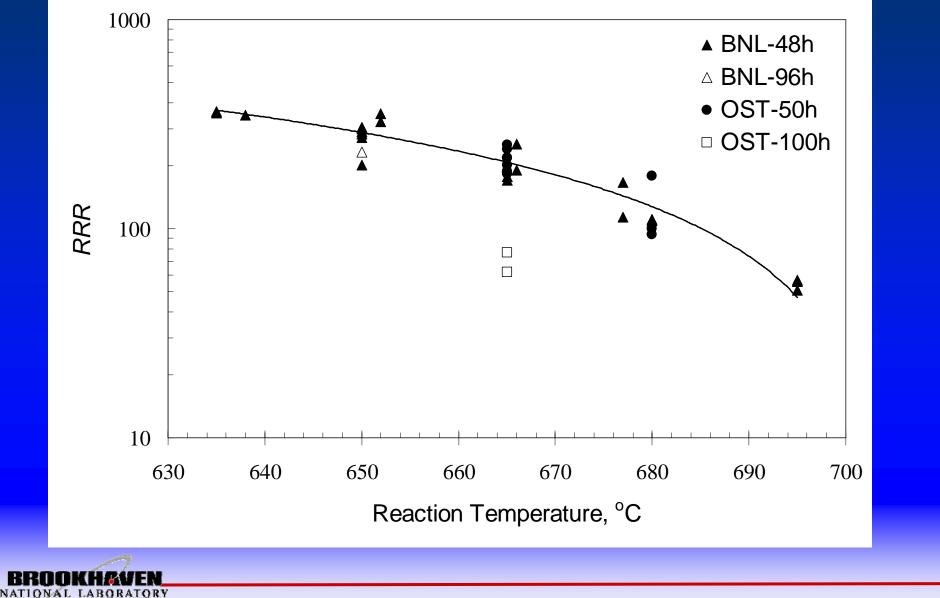
- Two parameters (RRR and J_c) must be optimized.
- Fortunately, RRP provides so much tin activity that J_c optimizes rapidly, opening a fairly wide window to tune RRR



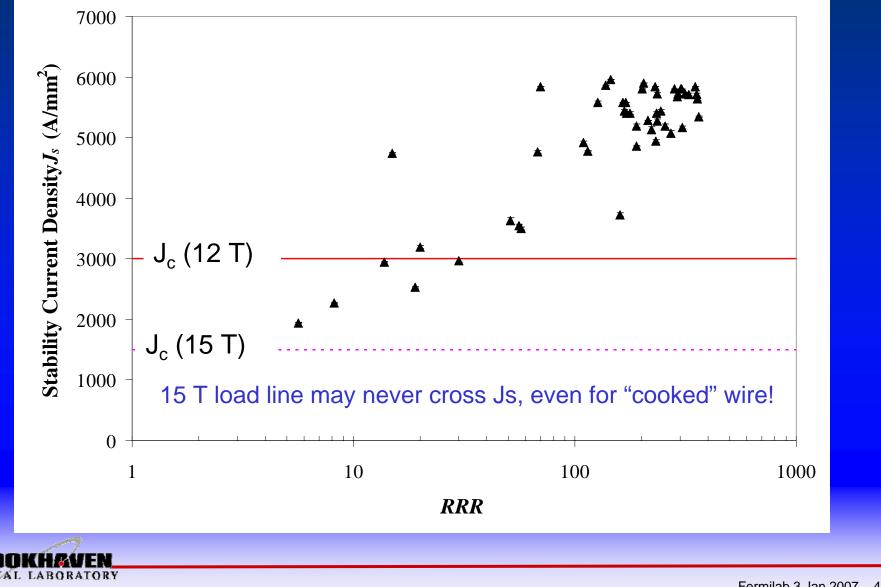
Jc (12T) BNL and OST 48-50h data



RRR as a function of reaction time and temperature



Stability current density of RRP 54/61 wire



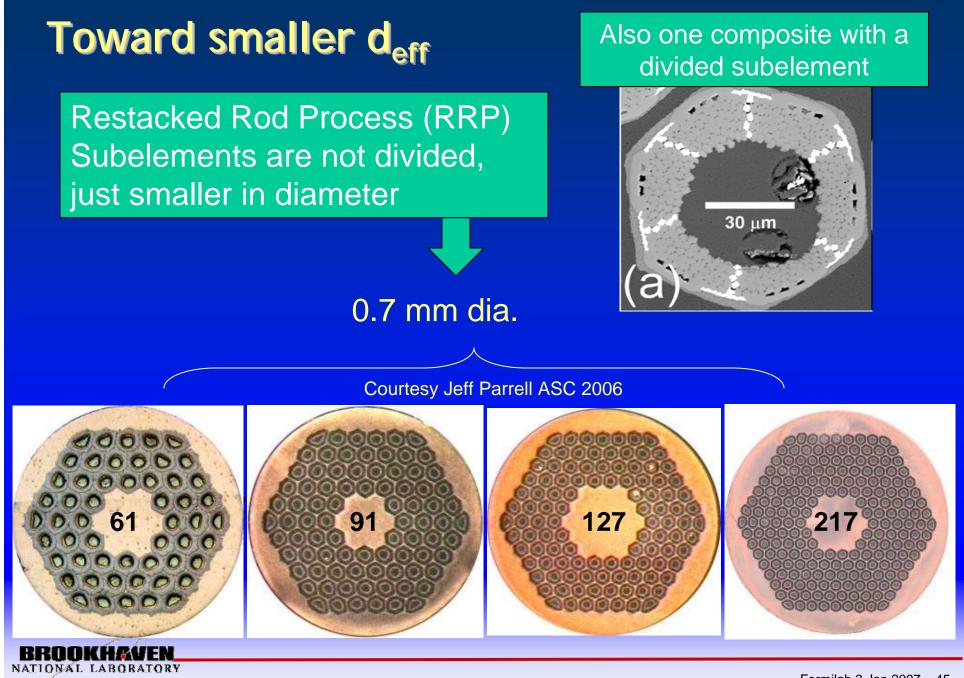
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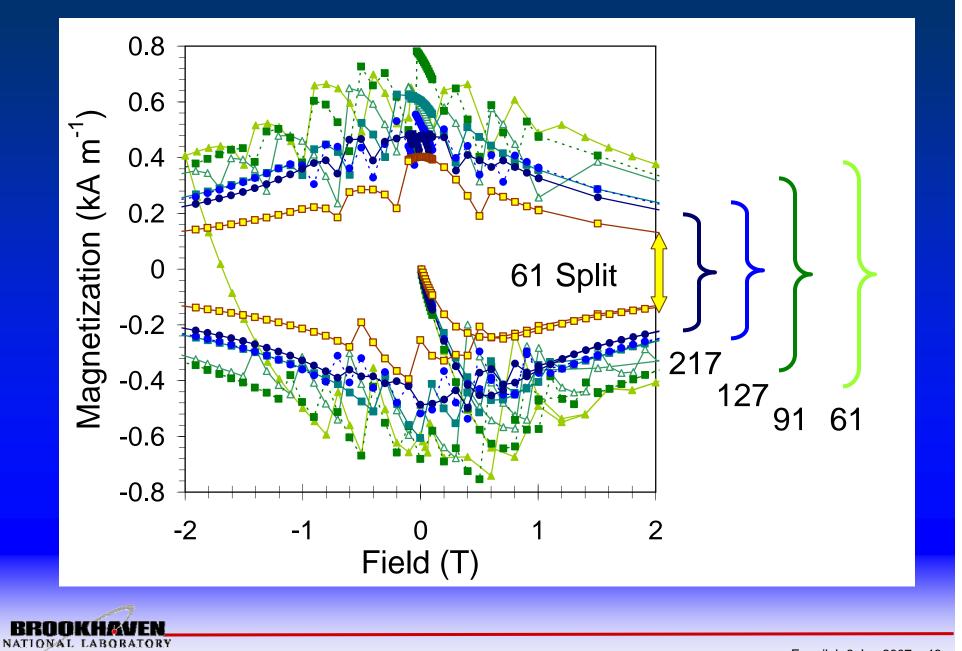
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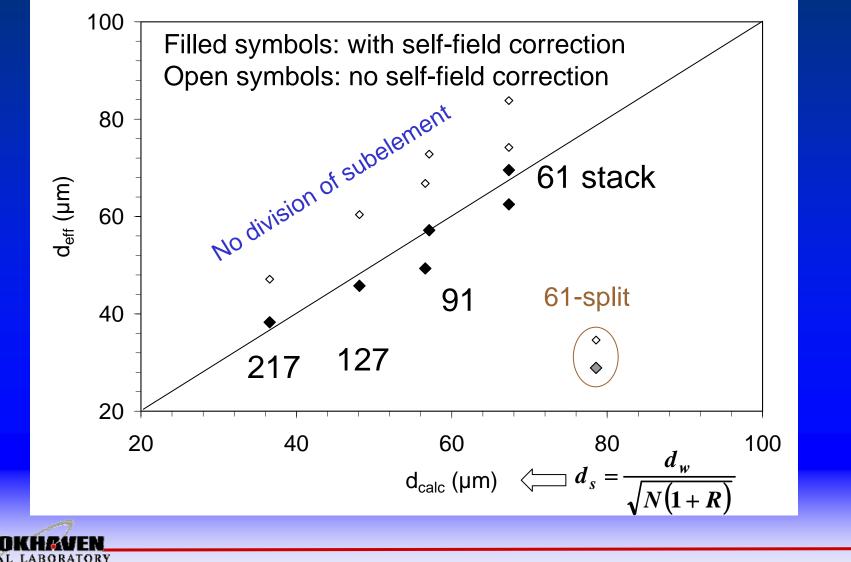
‡ Powder-in-tube process







Effective filament diameter and subelement diameter are equal; 40 µm has been achieved!



Smaller d_{eff} comes with performance sacrifice

N	d _s µm	HT type	HT	J _c (12) A mm ⁻²	RRR		
54 (61)	68	А	72h@675°C 2900		6		
		В	36@660 2985		47		
		С	24@650	2760	127		
84 (91)	57	В	48@650	3050	165		
90 (91)	57	А	48@695	2680	20		
		С	36@635	2330	344		
108 (127)	49	В	48@665	2830	127		
126 (127)	48	А	72@665	2260	4		
		В	72@635	2040	9		
		С	36@635	1910	114		
198 (217)	37	В	48@650	2460	12		
54 split	79	А	100@665	2290	6		
		В	50@665	2120	96		



Where to look for more layer Jc

- Grain size as small as 30 to 50 nm have been achieved (proof of principle)
- 50% gain in pinning might be realized by reducing grain size to ~80 nm
 - ~150 nm obtained for 650 to 665 °C right now
- IN PROGRESS: LARP 54/61
 - 2 x HT 96 h @ 620 °C
 - HT 150 h @ 605 °C

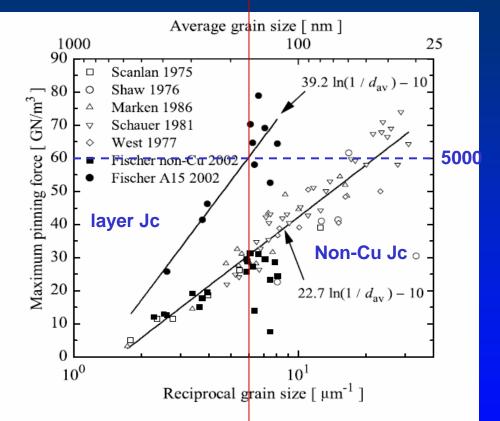


Figure 10. Maximum pinning force as function of reciprocal grain size, after Fischer [89]. (*Adapted with kind permission of C M Fischer*).

Can this line be moved to the right, e.g. by a long reaction at 550 to 600 °C?



Can a wire design yield still higher Nb₃Sn %?

- Ultimate limit: 2 volume units Nb fully react with 1 unit Sn to provide 2.67 units Nb3Sn
 - i.e., not more than 89% of noncopper area can be converted to Nb₃Sn when starting from elements
 - (2:1 volume = 3:1 mol Nb:Sn)
- Must let tin escape from core, else large grains form
 - PIT, RIT cannot therefore achieve the ultimate limit above!

- Lower LAR: increases Nb content, but pinches off tin diffusion
- Smaller Nb filaments at same LAR provides more access for Sn, but sacrifice shape control
- Better roundness of Nb and smaller Nb grain size will improve geometry stability, but this requires changes in Nb supply
- ACTIVITIES IN PROGRESS UNDER CONDUCTOR DEVELOPMENT GROUP (OI-ST)

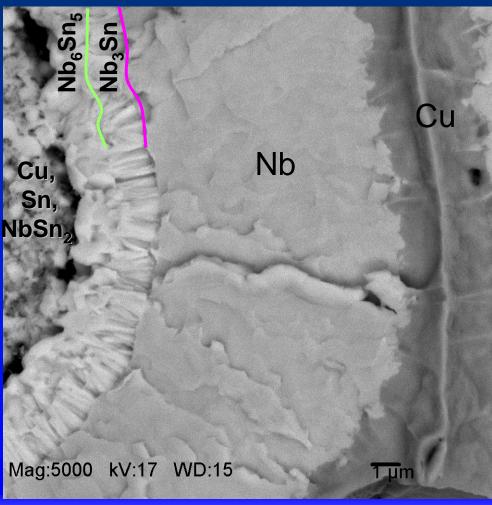


Regarding trapped tin in PIT...

SMI billet 34 24 h @ 575°C

Lesson: It was not possible to form Nb_3Sn without first forming Nb_6Sn_5 , no matter how low the reaction temp.

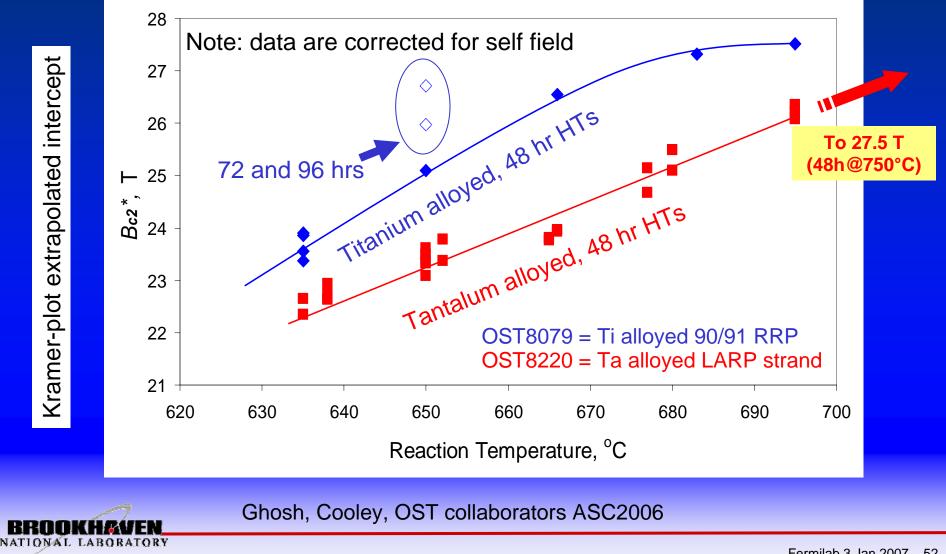
Can a thick layer of finegrained Nb_6Sn_5 be formed first, and then converted to Nb_3Sn without coarsening? (We were not successful)



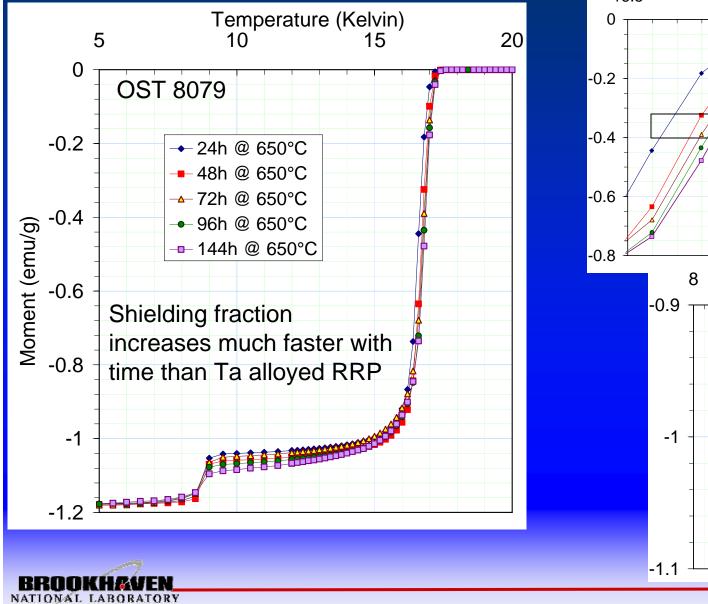
SEM and prep by Jesse Wright & Bob Sabatini Wire samples from FNAL

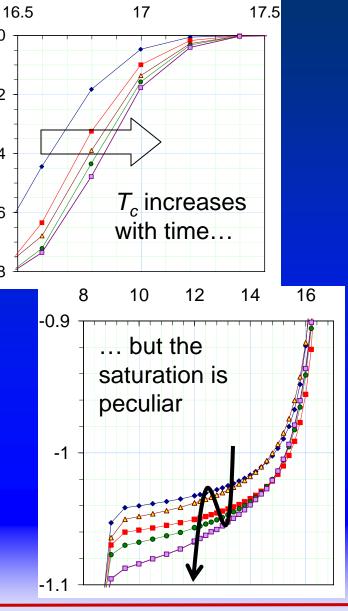


Ti alloying -Faster way to get high-field performance?

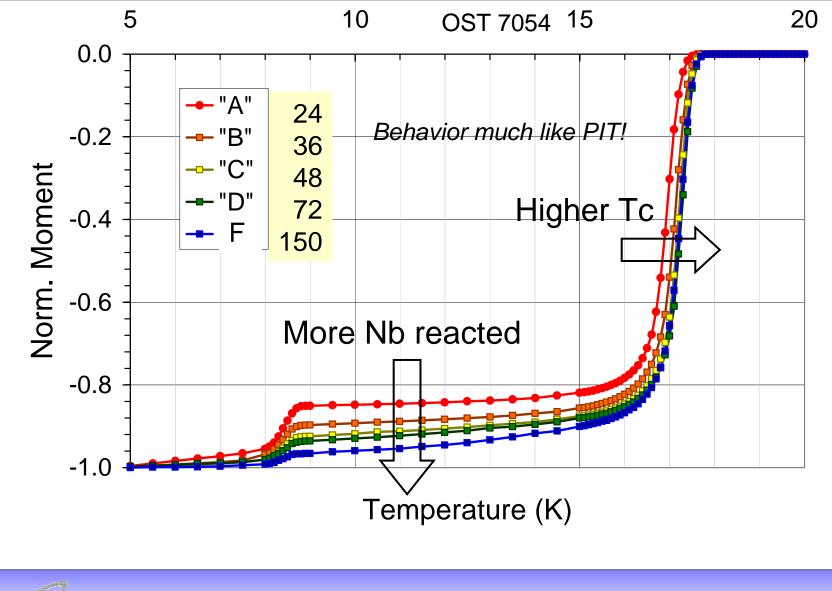


Ti gradients as well as Sn gradients?





(Ta alloyed RRP for comparison)



BROOKHAVEN

What's going on with Ti?

- Possibility 1: Ti is a more effective alloying agent than Ta (contrary to past results...).
- Must adjust LAR to accommodate Ti diffusion
- IN PROGRESS: RRP 8079 given intermediate 575°C step to diffuse Ti uniformly prior to Nb₃Sn reaction

- Possibility 2: Better Ti results are an ARTIFACT of faster tin diffusion
 - It is known that Ti penetrates quickly along grain boundaries, which would widen the "pipes" through which tin flows.
- If so, then does Ti really alloy?
 - Past work: 2% Ti at GBs, 1.3% in Nb₃Sn region

Billet	Design %Ti	LAR	Max B_{c2}^{*}	Comment
RRP-8079	2.4	0.26	27.5	
HER-7981	2.5	0.28	26.5	Tin starved?
RRP-8720	2.0	0.20	25.8	B _{c2} [*] decreases with HT time



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 - Managing stability
- Is further improvement of Nb₃Sn possible?
- If not, what's next? Clues from emerging areas in superconducting materials for HEP
 - * Restacked Rod Process™

† Modified jelly-roll process

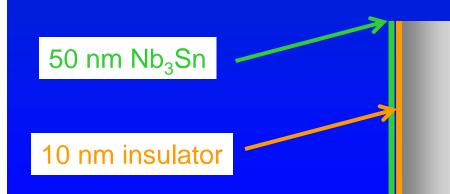
‡ Powder-in-tube process



SRF flux entry barrier: Breaking the Nb monopoly

- A Gurevich: a thin layer of a high-field superconductor over Nb will keep out fluxons and their normal electrons
- $H_{S} \sim (\lambda_{L} / d) H_{S}^{bulk}$

Several materials challenges!



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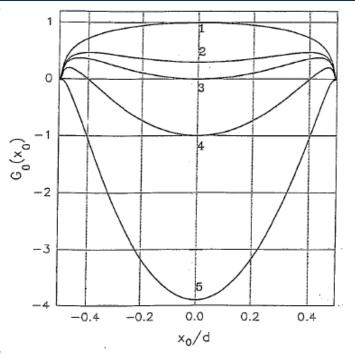


FIG. 7. Single-vortex thermodynamic potential (15) for different H at J=0: $H < H_m(1)$, $H_m < H < H_{c1}$ (2), $H=H_{c1}$ (3), $H_{c1} < H < H_s$ (4), and $H > H_s$ (5).

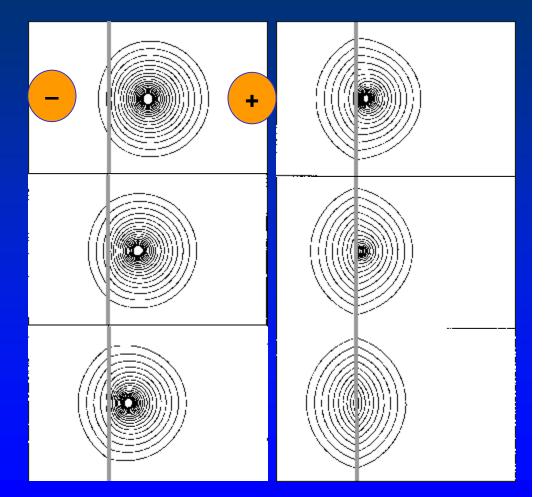
Nb

 $H_s = \phi_0 / 2\pi \, d\xi \; .$

Gurevich APL88-012511; Stejic et al PRB 49:1274 (1994)

AJ vortices at Nb grain boundaries?

 "Hybrid" Abrikosov-Josephson vortices will penetrate boundaries even though the boundaries themselves do not obstruct current



A Gurevich PRB 2003-2005



50 T solenoid for muon cooling

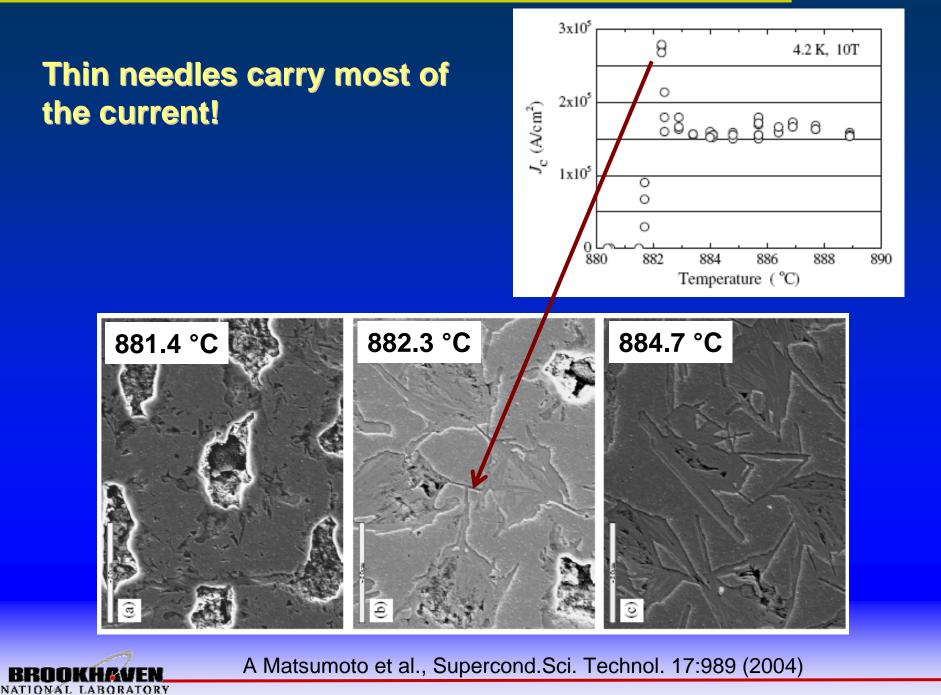
A Bi-2212 + stainless steel monster?



<u>Challenges</u>

- Open the reaction window to enable wind and react
 - Reaction temperature depends on melting point of Bi_{2+x}Sr_{2+y}CaCu₂O₈ ceramic, which is sensitive to Bi:Sr ratio and must be controlled to ±2°C at > 880°C
 - ±15 °C possible but at expense of homogeneity and therefore current density (i.e. highest current density for smallest window)
 - How to heat a coil so uniformly?
- Explore react and wind
- Increase the current density and fill factor

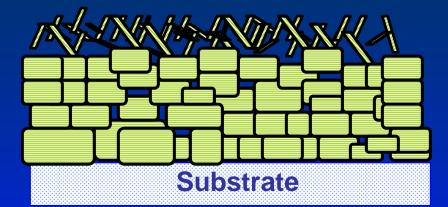


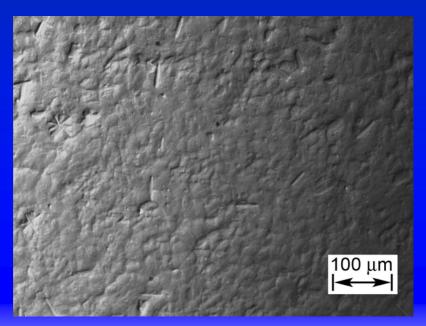


Can magnet designers use tapes?

Horizon: thick YBCO in long length 1000 A/cm tape @ 77K, 0.1T (= 2,500 A/cm @ >20T, 4.2K)

- Right now, basic research is making 100% gains in improving flux pinning
- Growth mechanisms are also better understood – new pathways toward thicker YBCO layers
 - Rapid nucleation provides grainboundary-like pinning for the first time!





V. Solovyov et al, BNL



Summary

- RRP strand is the engineering material that will allow LARP magnet construction!
- Lesson: Vertical nature of discussions within DOE-AARP greatly accelerated development to suit needs of LARP.
 - HEP Low Temperature Superconductor Workshop: End users, industry, and materials scientists together in one room for 2.5 days on an annual basis!
 - Efforts continue under Conductor Development Program
- Can we do better to facilitate R&D?
 - Example: DOE / NSF Center of Excellence



Summary, cont.

- LARP strand has room for further improvement. Incremental advances are likely given present effort.
 - Must sustain for long term
 - Must allow curiosity to produce breakthroughs
- Changing specs often has repercussions!
 - Example: Nb grain size affects roundness and shape stability, which affect efficiency of tin diffusion and thereby $J_{\rm c}$
- New materials offer new opportunities and present new challenges. We need to attack the scientific and technological questions with the same coherent vertical effort.



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... And many pleasant discussions with the AARP community!

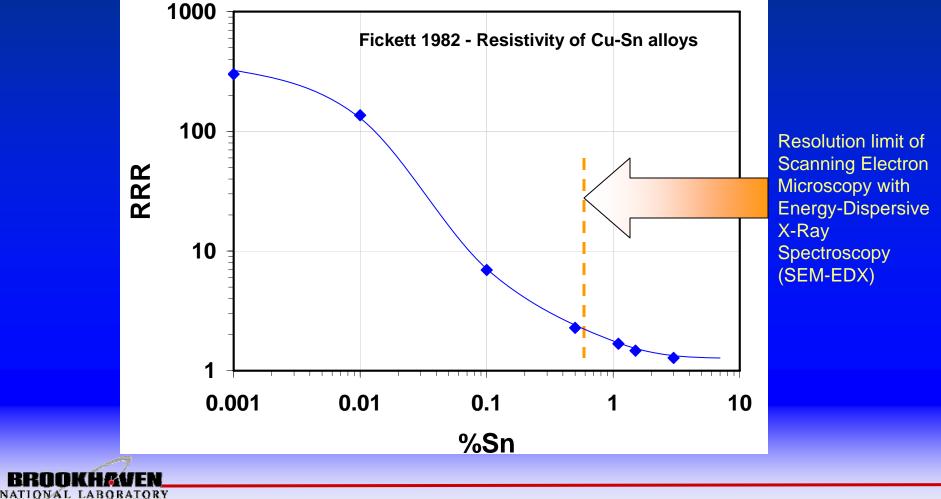


Reserve slides

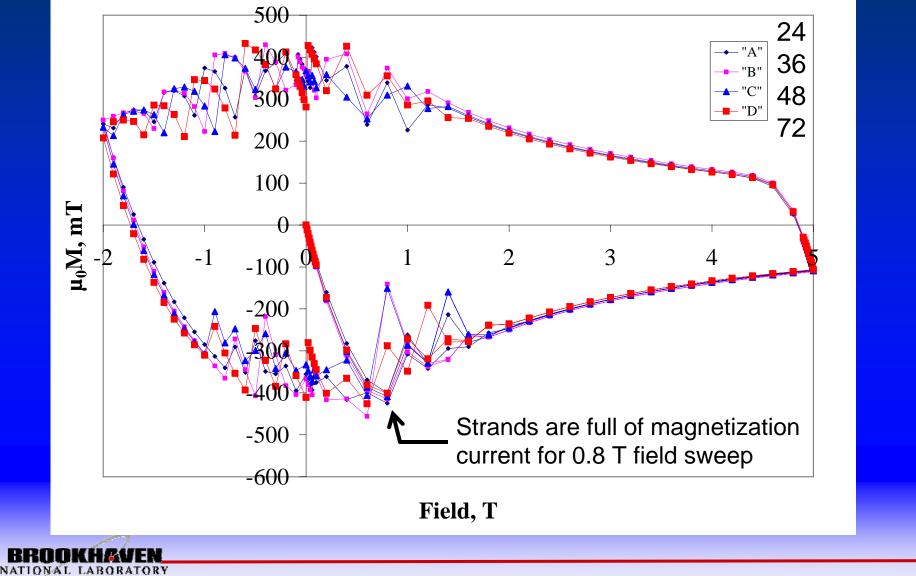


Tin destroys thermal conductivity of copper

 Tin is a potent electron scatterer: 0.1% Sn will reduce RRR from 300 to 7 — Fickett, Cryogenics vol. 22, p. 135 (1982)



All reactions produce unstable strands because barrier reacts to form ring of Nb₃Sn around subelement



How contaminated is the copper?

SEM-EDX microanalyses:

- 120-300 sec., 17 kV
- 2,500-10,000 counts
- no Nb detected

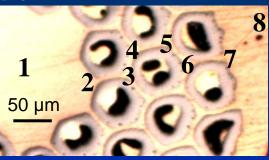


Table II. Atomic fraction of tin (%) determined by EDX as a function of location in Fig. 1 for various samples.

Location	-		Samples		
	Α	С	D	Е	F
1	< 0.5	< 0.5	< 0.5	< 0.5	0.66
2	< 0.5				1.0%
3	< 0.5	4.26	2.69	3.02 PAT4HW	
4	< 0.5	<u>*</u>	IERMAL	PA	/AY _{2.07}
5	< 0.5)EAD	1.88
6	< 0.5				
7	< 0.5		040	0.51	\cap
8	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5

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Traditional V-I curve

