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Title

A review of the properties of Nb3Sn and their variation with A15 composition, morphology and strain state

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A review of the properties of Nb_3Sn and their variation with A15 composition, morphology and strain state

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…Pushing the Limits of RF Superconductivity, Padua, Italy

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1954 → Discovery of Nb_3Sn

PHYSICAL REVIEW VOLUME 95, NUMBER 6 SEPTEMBER 15, 1954

Superconductivity of Nb_3Sn

B. T. Matthias, T. H. Geballe, S. Geller, and E. Corenzwit
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received June 10, 1954)

Intermetallic compounds of niobium and tantalum with tin have been found. The superconducting transition temperature of Nb_3Sn at 18°K is the highest one known.

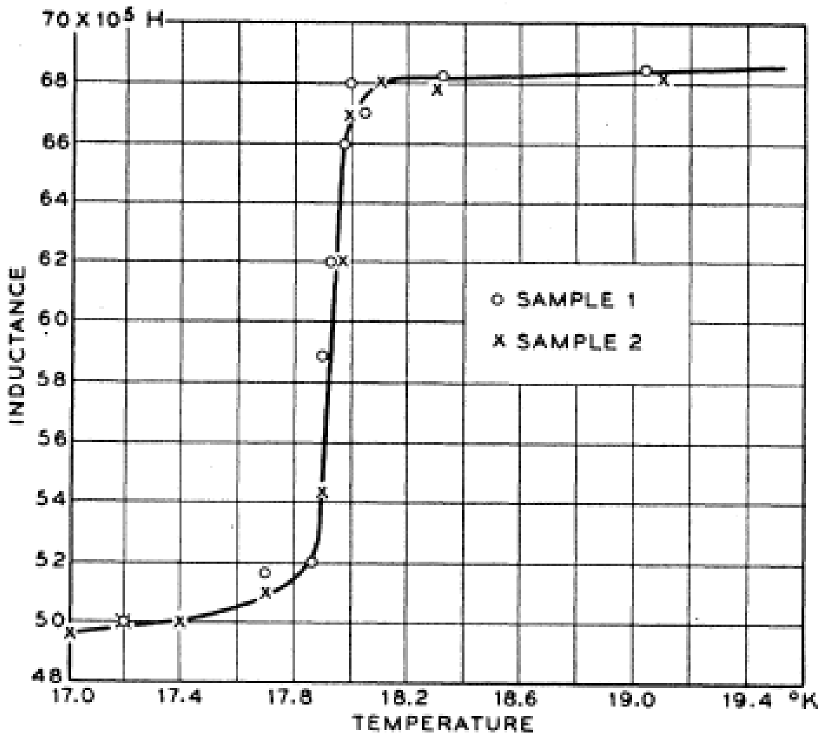


Fig. 1. Variation of susceptibility with temperature of Nb_3Sn.

compounds. No reference to Nb-Sn or Ta-Sn was found in the literature. The melting point of niobium is nearly 400° above the boiling point of tin, and an arc furnace is therefore out of place. A complete reaction can, however, easily be obtained by having molten tin run over Nb or Ta powder in a closed-off quartz tube at 1200°C. Nb_3Sn and Ta_3Sn seem to be formed by a peritectic reaction between 1200°C and 1550°C.

Pre-2005 literature values

Superconducting transition temperature	T_c	18	[K]
Lattice parameter at room temperature	a	0.5293	[nm]
Martensitic transformation temperature	T_m	43	[K]
Tetragonal distortion at 10 K	a / c	1.0026	
Mean atomic volume at 10 K	V_{Mol}	11.085	[cm^3/Mol]
Sommerfeld constant	γ	13.7	[mJ/K^{2}Mol]
Debye temperature*	Θ_D	234	[K]
Upper critical field*	$\mu_0 H_{c2}$	25	[T]
Thermodynamic critical field*	$\mu_0 H_c$	0.52	[T]
Lower critical field*	$\mu_0 H_{c1}$	0.038	[T]
Ginzburg-Landau coherence length*	ξ	3.6	[nm]
Ginzburg-Landau penetration depth*	λ	124	[nm]
Ginzburg-Landau parameter λ/ξ*	κ	34	
Superconducting energy gap	Δ	3.4	[meV]
Electron-phonon interaction constant	λ_{ep}	1.8	→ **Theory**

And obviously ρ_n

Moore, PRB 1979; Orlando, PRB 1979; Guritanu PRB 2004

Composition: $Nb_3Sn \rightarrow Nb_{1-\beta}Sn_{\beta}$

- Binary phase diagram → 18 to 25 at.% Sn → ‘A15’

Tetragonal distortion:

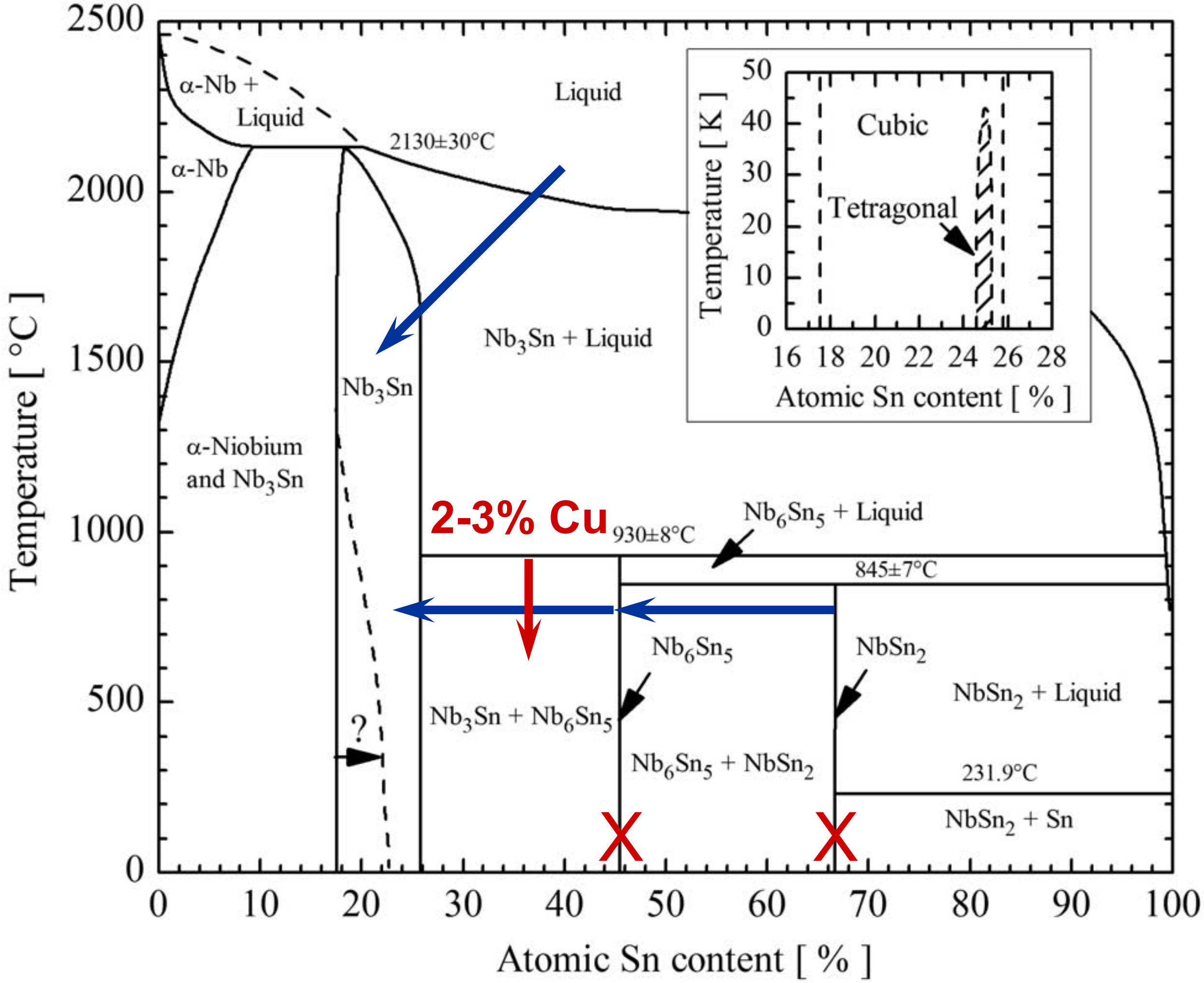
- C/a ~ 1.0035

Binary A15 formation:

→

Presence of 2 to 3% Cu:

X ↓ X



- A15 phase is insoluble with Cu
- Cu at Grain Boundaries
  - Charlesworth, JMS 1970, Flükiger, ACE 1982

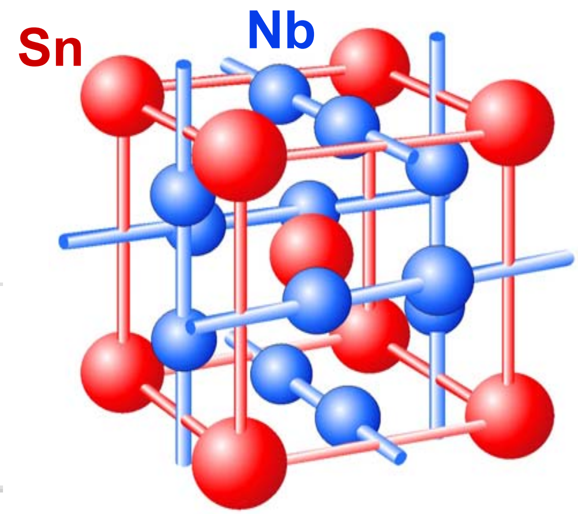
What happens with changing Sn content?

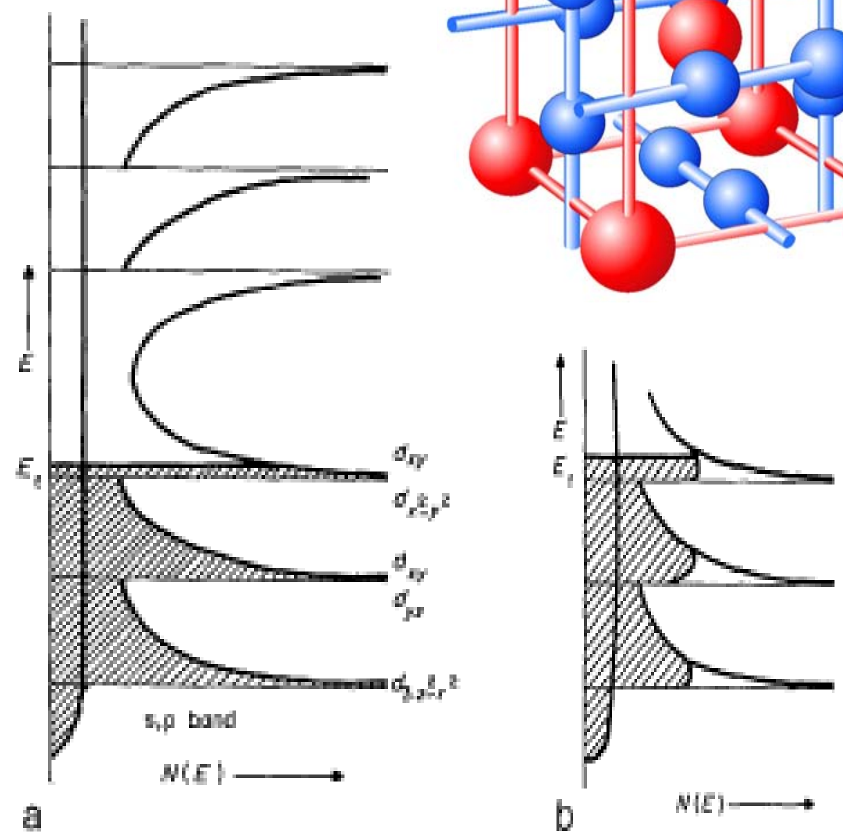
Pure Nb
- *bcc* Nb spacing 0.286 nm
- T_c = 9.2 K

Nb_3Sn → 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

A15 lattice and DOS

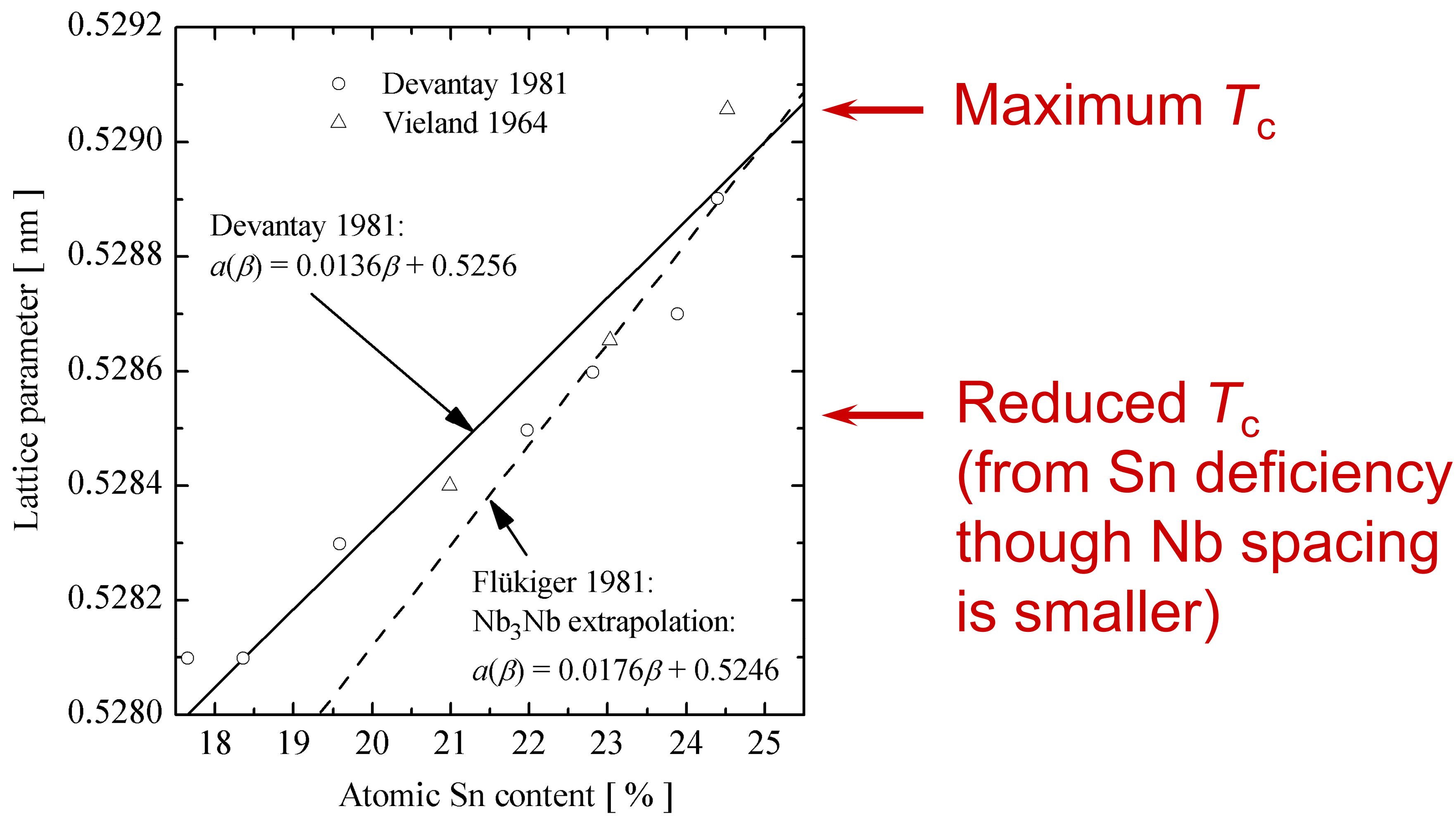




- Dew-Hughes, Cryogenics 1975

Sn content: Lattice parameter

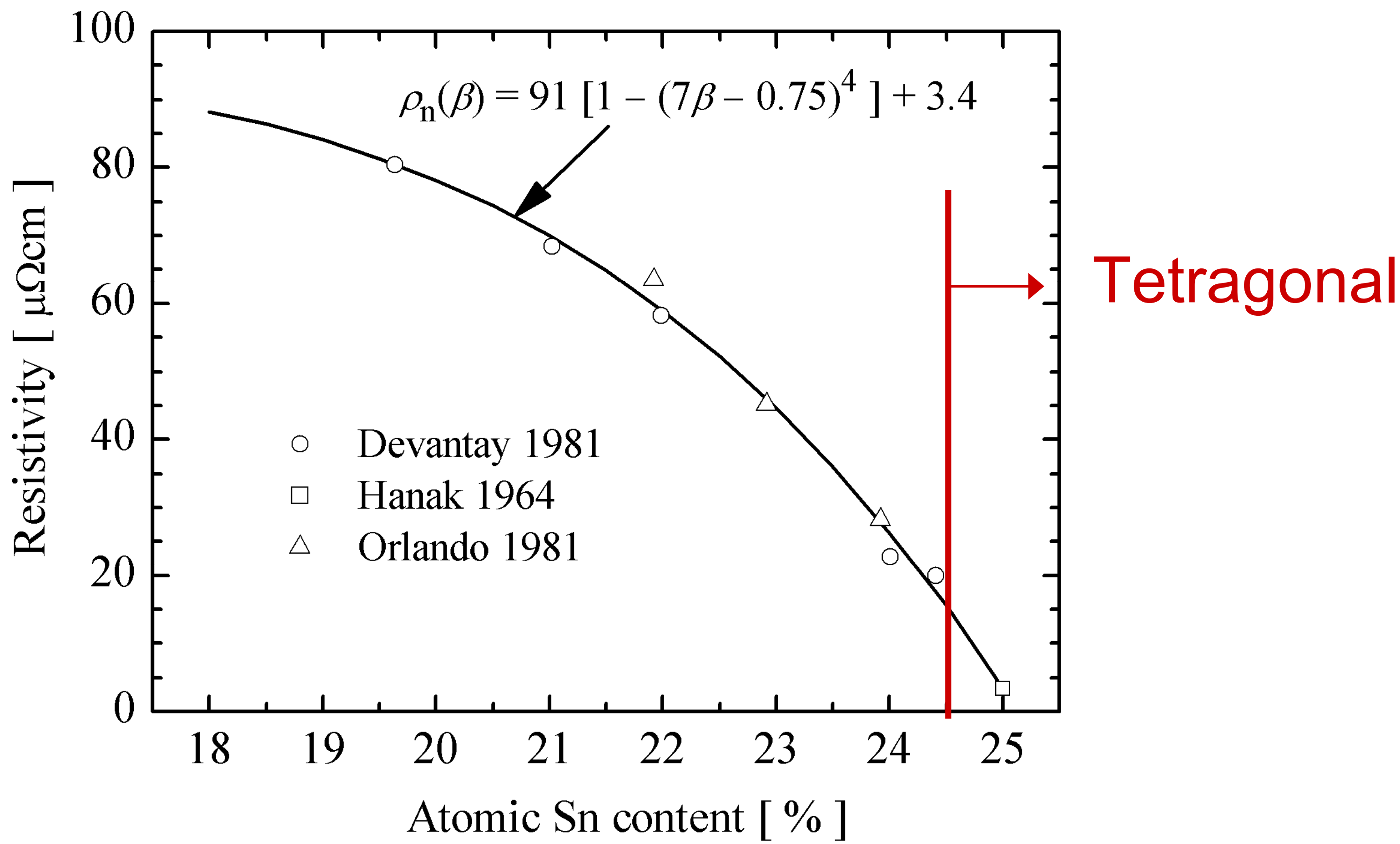
a increases with Sn content (as does T_c (below))



- Devantay, JMS 1981; Vieland, RCA Rev. 1964; Flükiger, 1981

Sn content: Resistivity

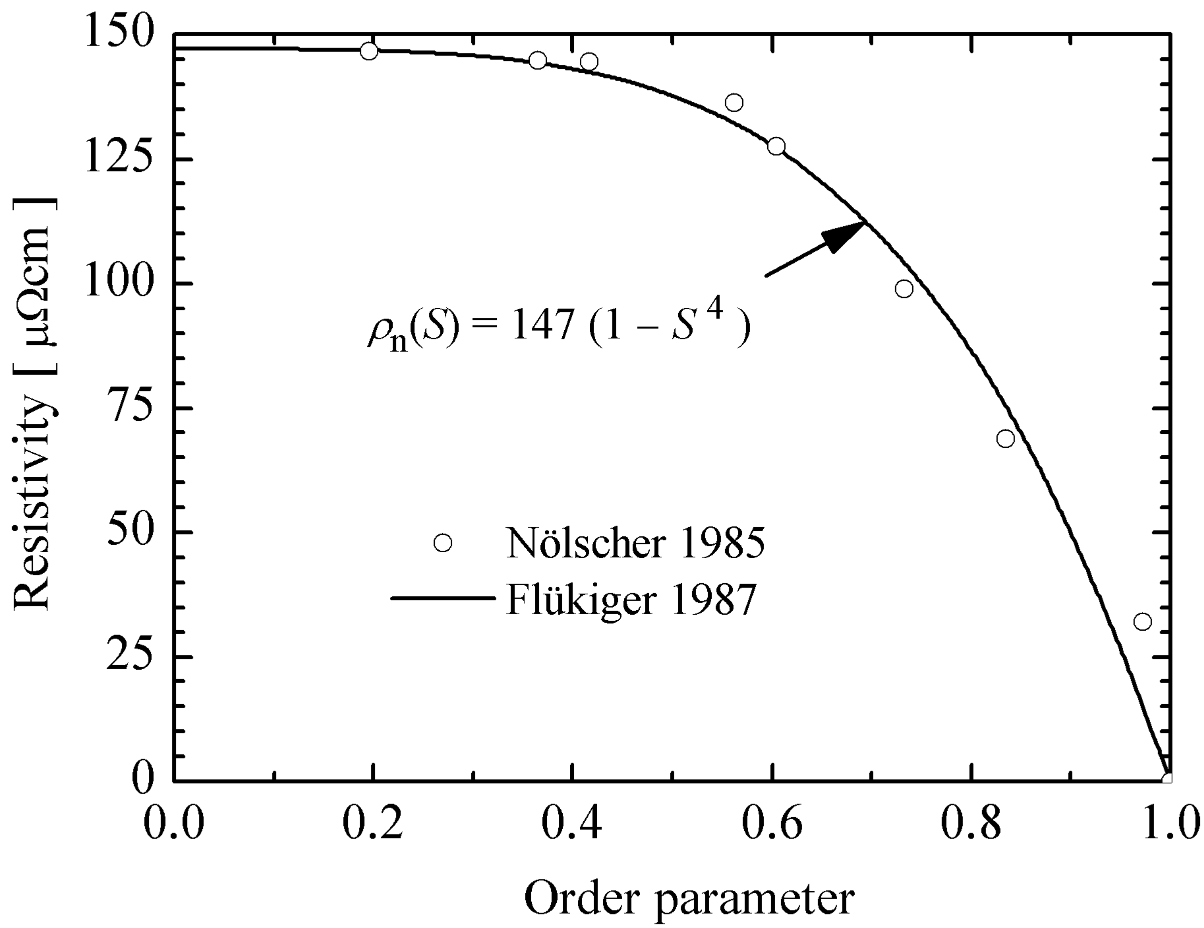
Nb_3Sn is cleanest at stoichiometry



- Devantay, JMS 1981; Hanak, RCA Rev. 1964; Orlando, TM 1981

Resistivity and Long Range Order

Bragg-Williams Order Parameter varied through irradiation



- Effect on ρ_n similar as changing Sn content
- *a*, *S* and ρ_n can all be related to atomic Sn content

Nb chain continuity, $N(E_F)$, λ_{ep}, T_c, H_{c2}

In general

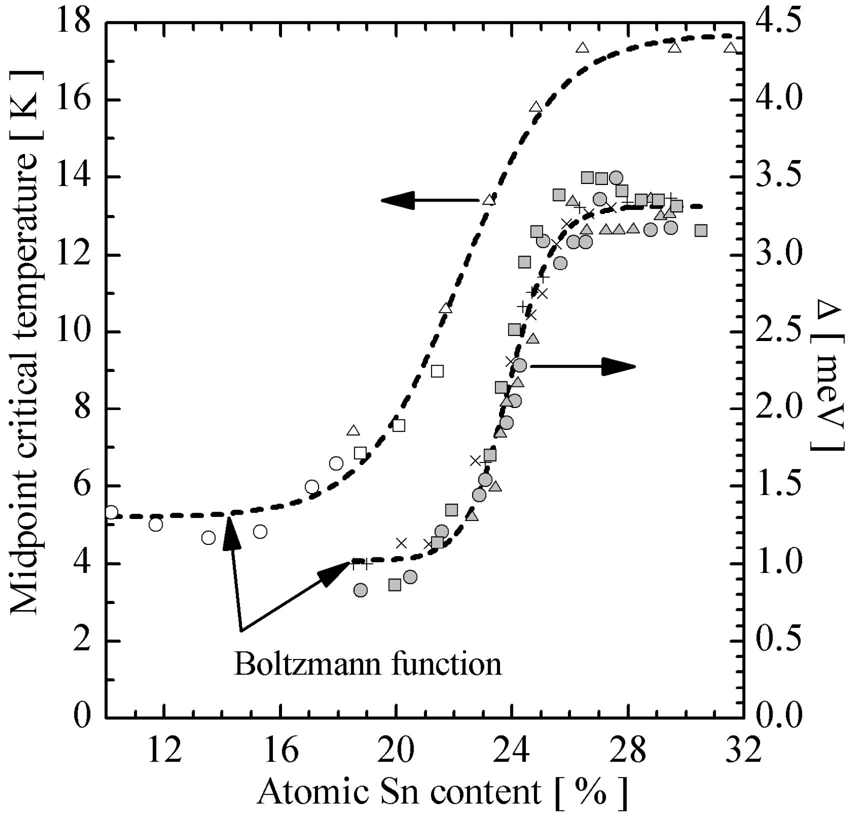
- Sn deficiency
- Tetragonal distortion
  - 24.5 – 25 at.% Sn
- Strain
- Alloying (e.g. Ti, Ta, …to increase H_{c2})
- Dislocations
- (Anti-site) disorder

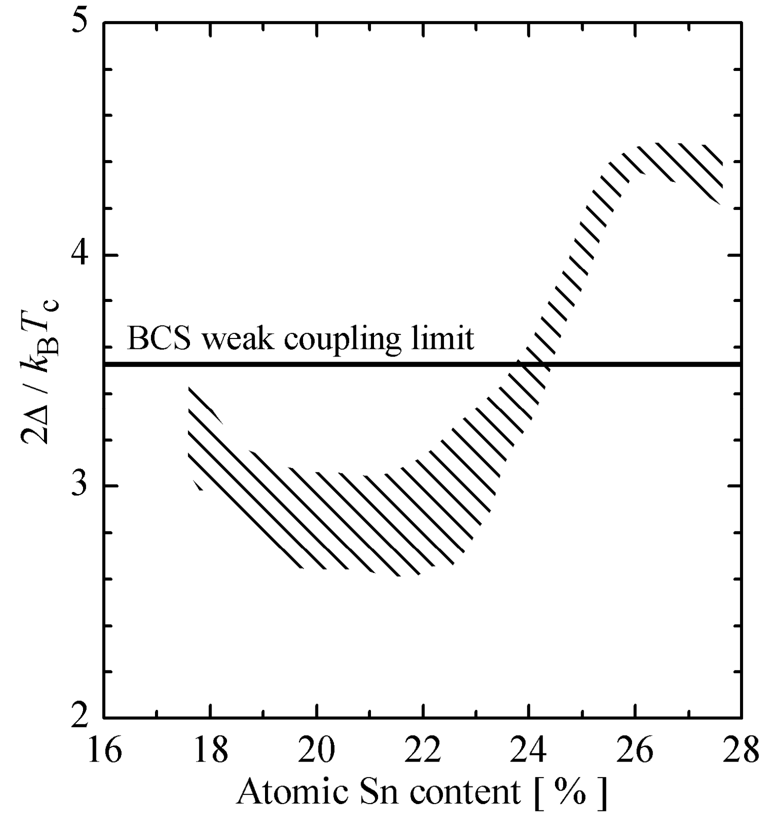
All affect Nb chain integrity ('Long Range Order')

- And thus $N(E_F)$ and λ_{ep}
- And thus T_c and H_{c2}

Sn content: Weak or strong coupling?

Moore, PRB 1979, thin film results



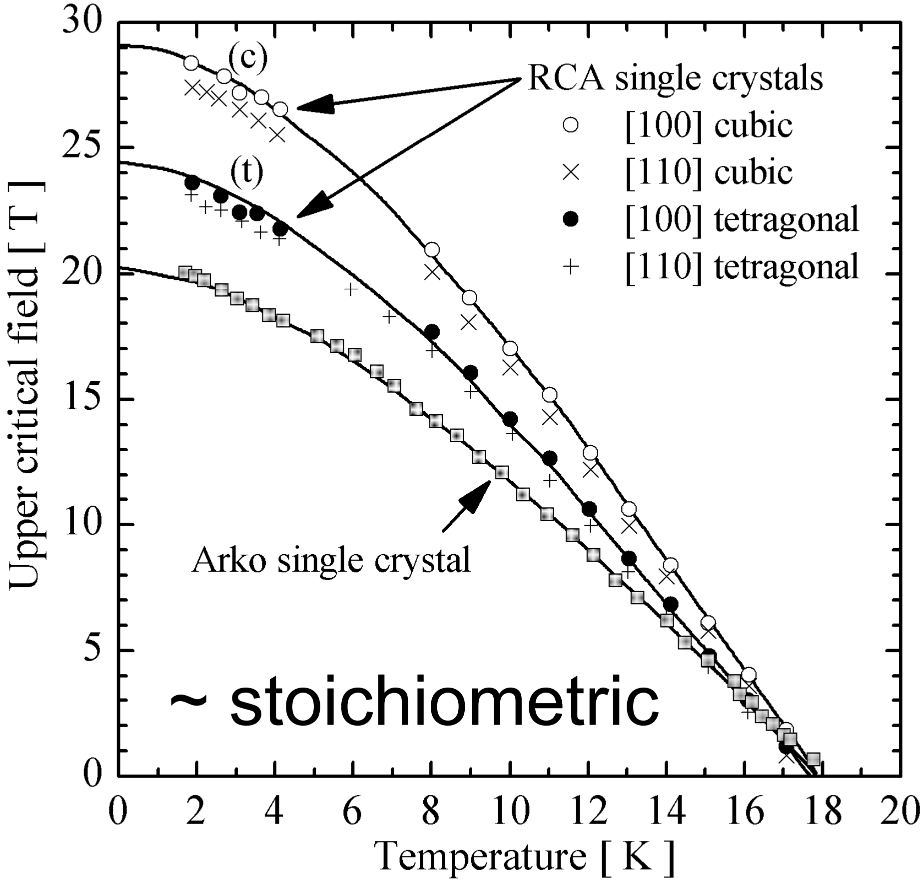


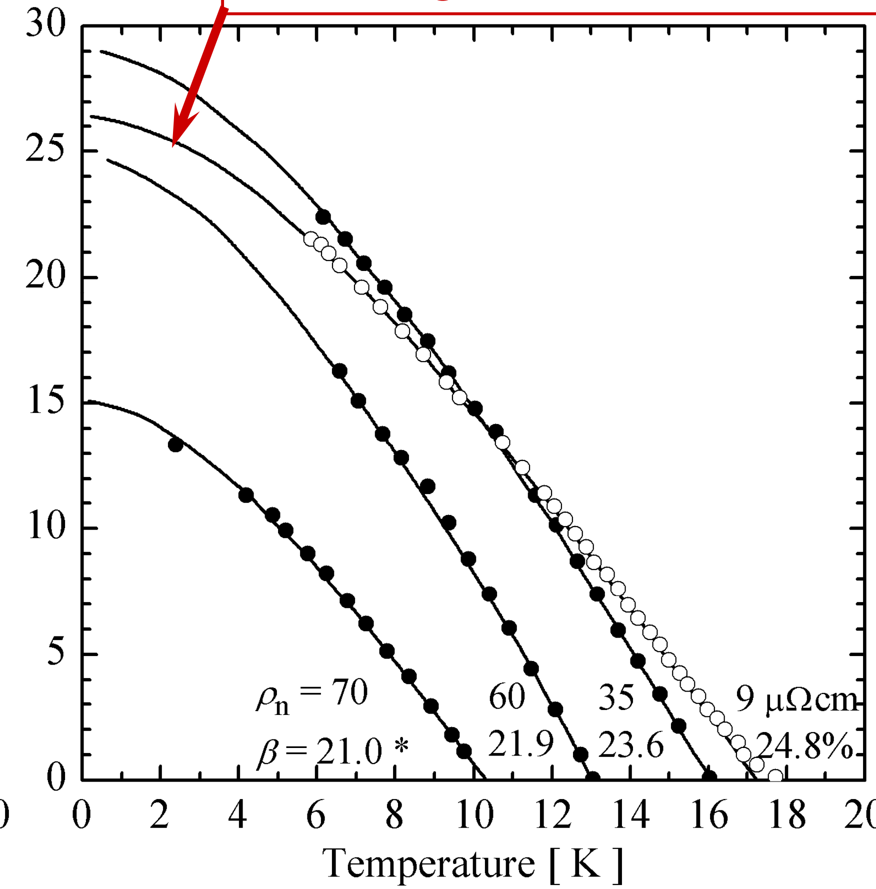
- Weak coupling below 23 – 24 at.% Sn
- Strong coupling approaching stoichiometry: λ_{ep} rising to ~ 1.8
- Strong coupling corrected BCS *insufficient* above ~ 23 at%Sn

Sn content: Tetragonal distortion, $H_{c2}(T)$

Single X-tal and thin films

Reduction at 24.8% due to tetragonal distortion





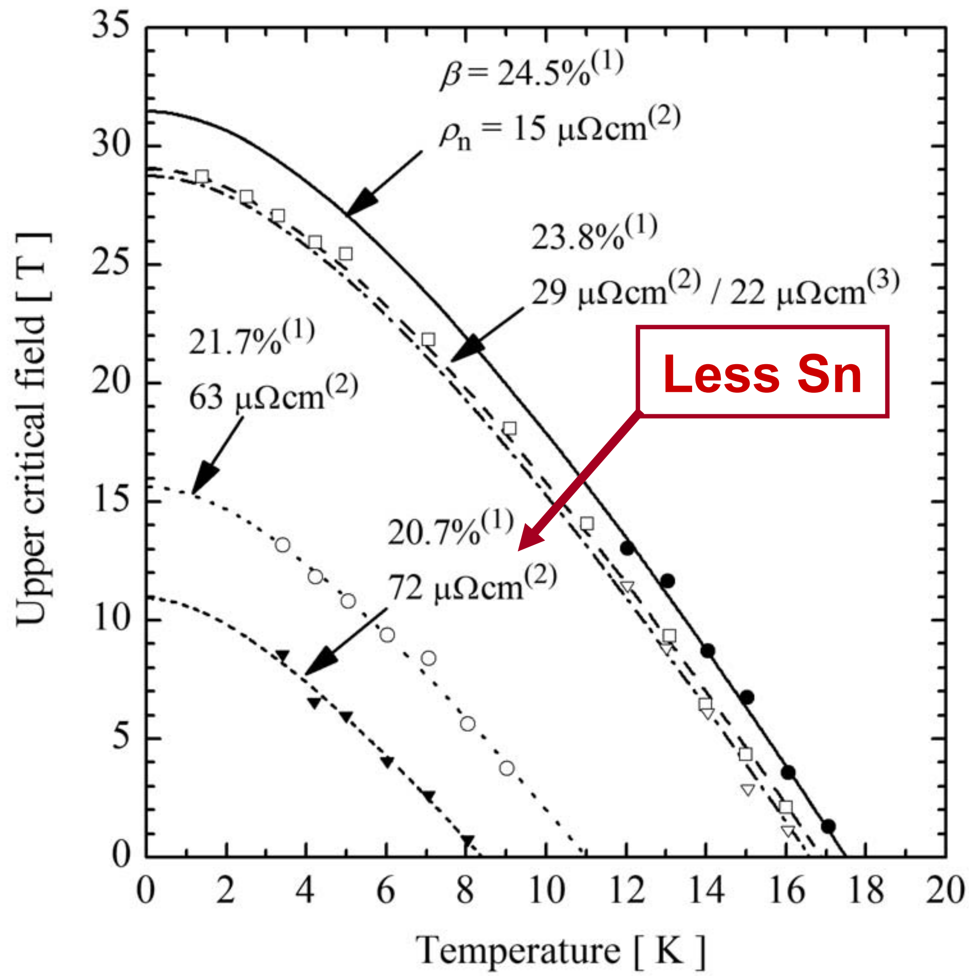
- Foner, Solid St. Comm. 1981
- Tetragonal distortion at stoichiometry

Orlando, TM 1981

Shift for < 24.5%

Sn content: $H_{c2}(T)$

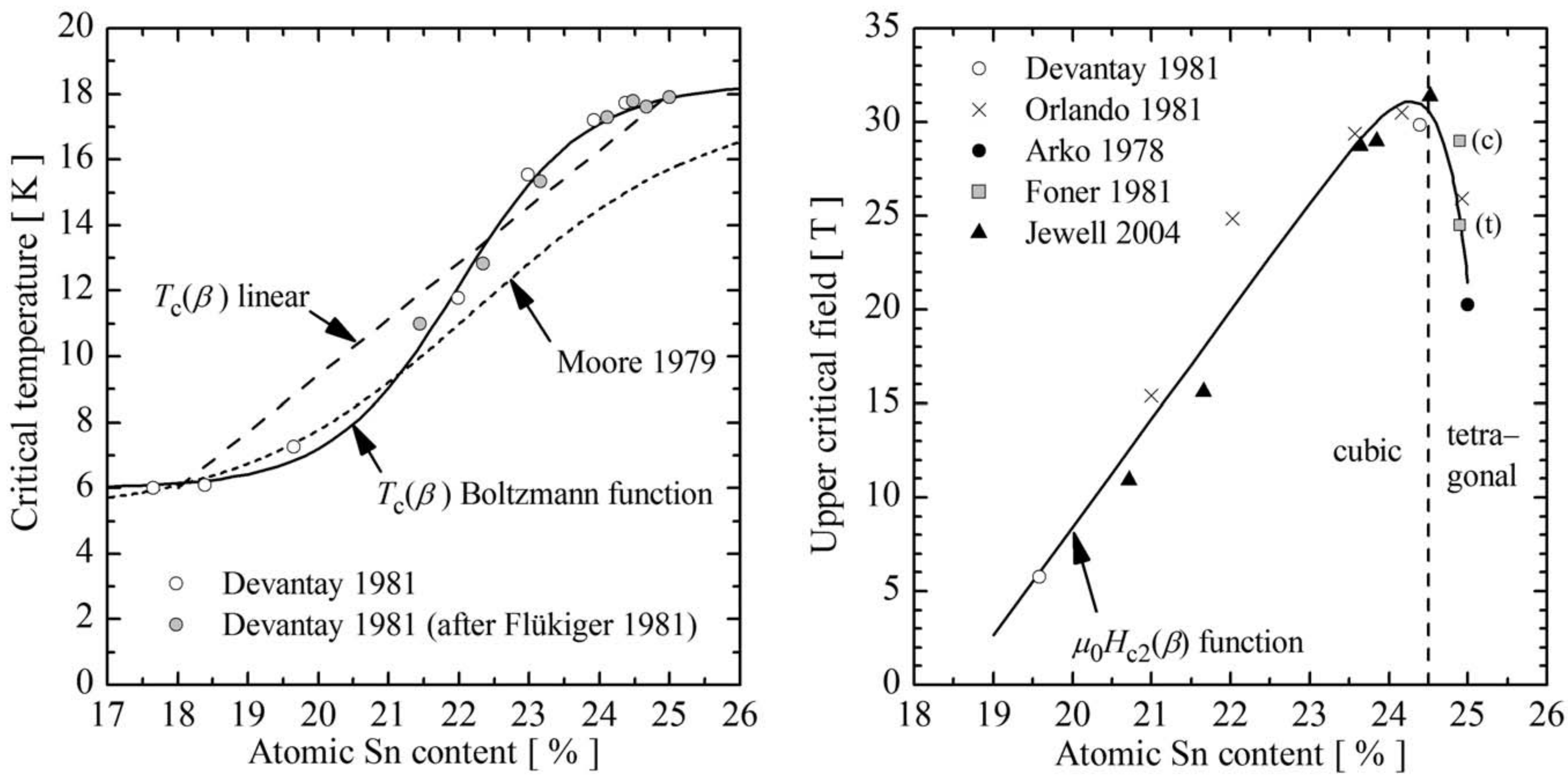
- Jewell, ACE 2004, bulk samples



- Sn richer A15 is cleaner
- Sn richer A15 has higher $H_{c2}(T)$ (until ~ 24.5 at.% Sn)

T_c and H_{c2} and Sn content summarized

Single crystal, bulk and thin film samples



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

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

How to make A15

Thin film deposition

- Hammond, J. Vac. Sci Tech. 1978
  - ➧ Multi-layers!

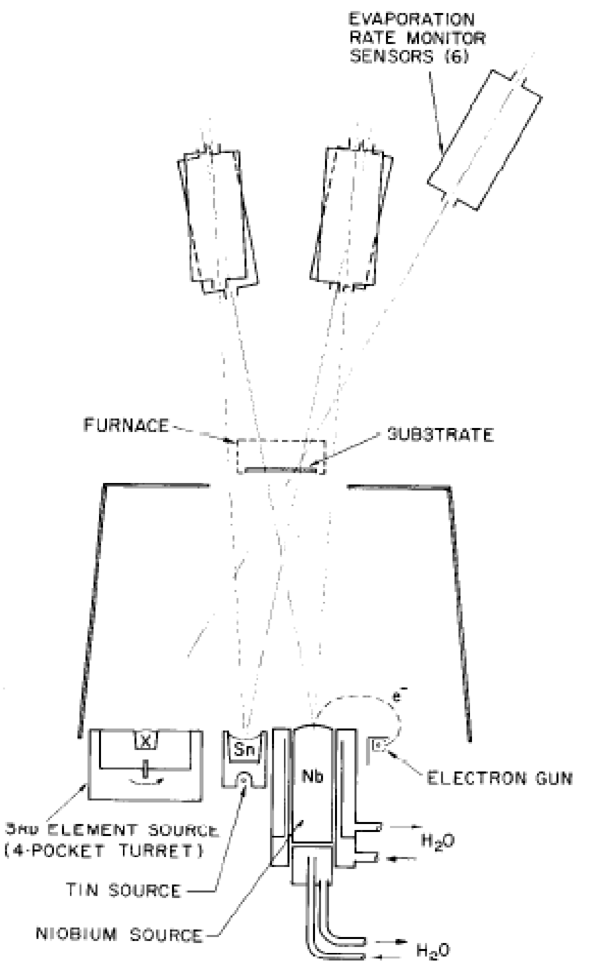
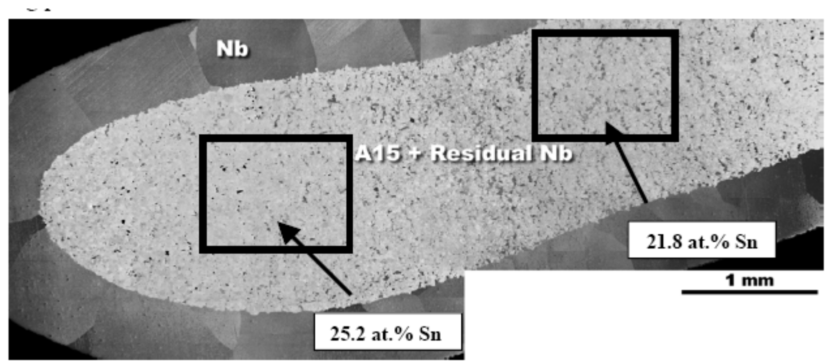


FIG. 1. Schematic of multisource deposition facility, showing three colinear

Bulk

Hot Isostatic Pressure

Goldacker, TAS 1993



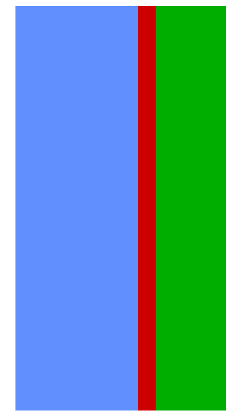
➧ Jewell, ACE 2004

Any Sn directly on Cu will poison Cu and lower RRR
➔ Use diffusion barrier e.g. Ta

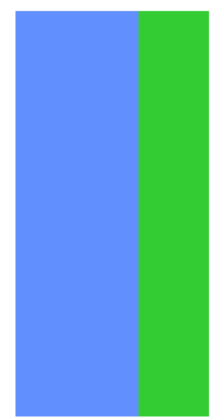
Diffusion

e.g. wires

Nb Cu Sn



Nb Cu+Sn



Diffusion based systems → Gradients

Example: Wires

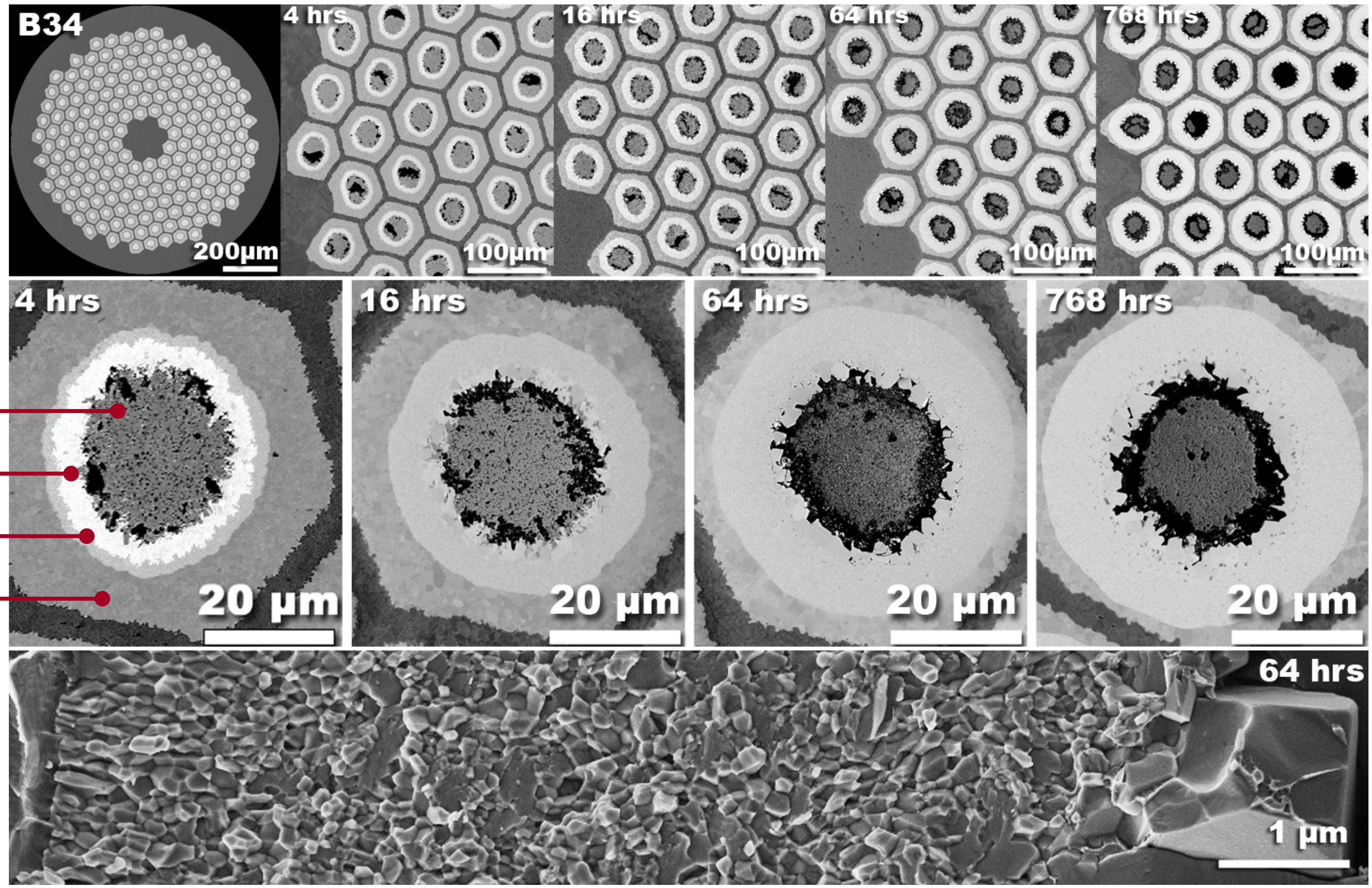
- Reaction at 675°C versus time in Powder-in-Tube wire (SMI)

$NbSn_2$, Cu, Sn

$Nb(Ta)_6Sn_5$

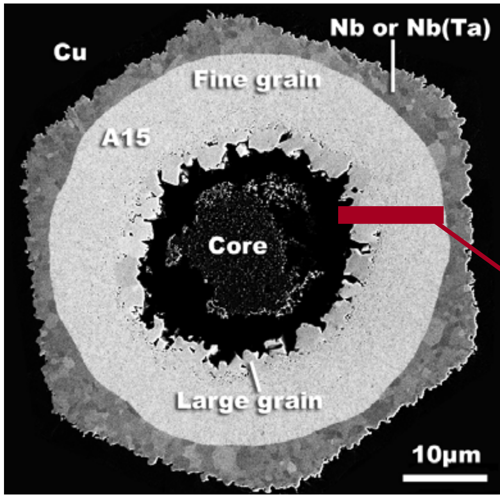
$Nb(Ta)_3Sn$

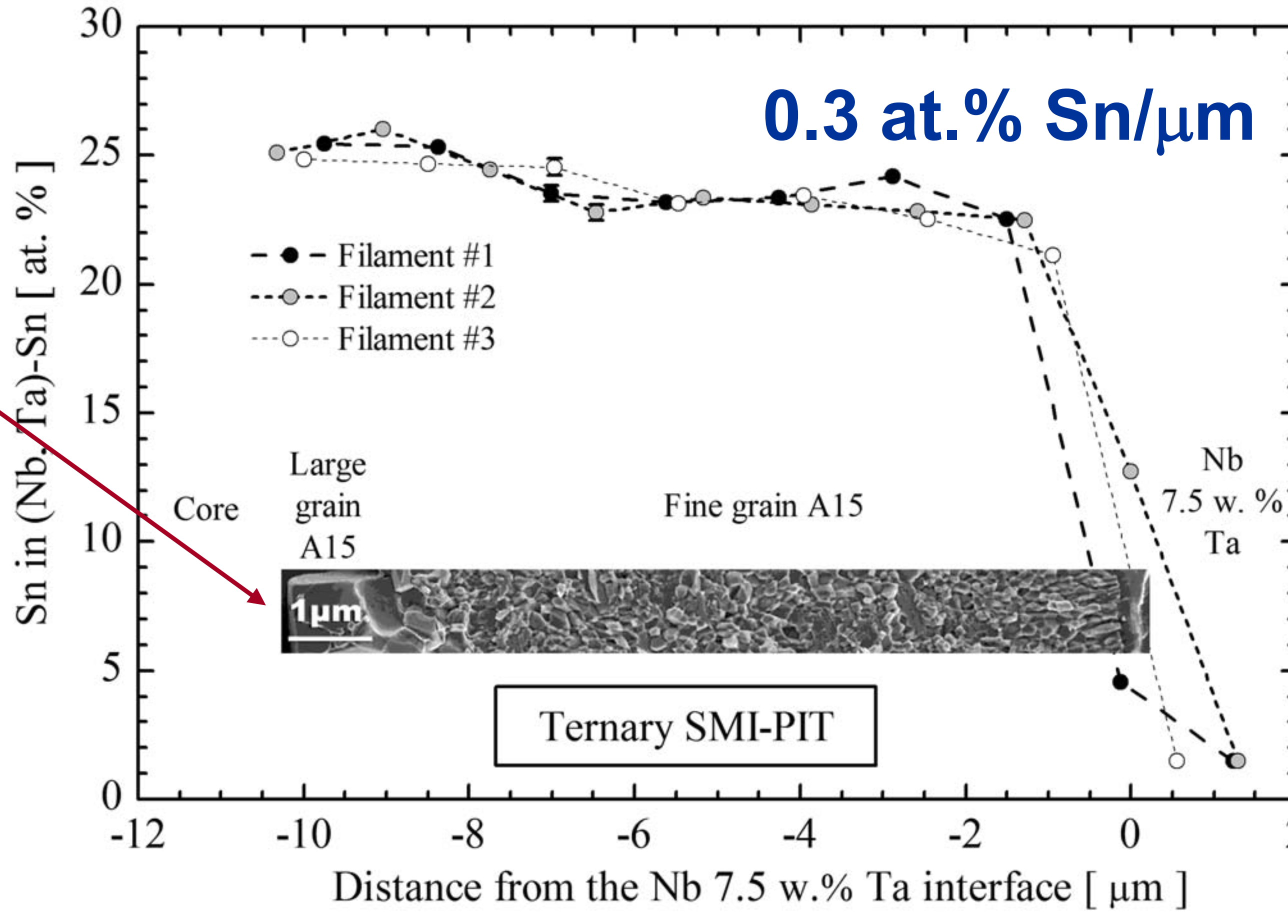
Nb(Ta)



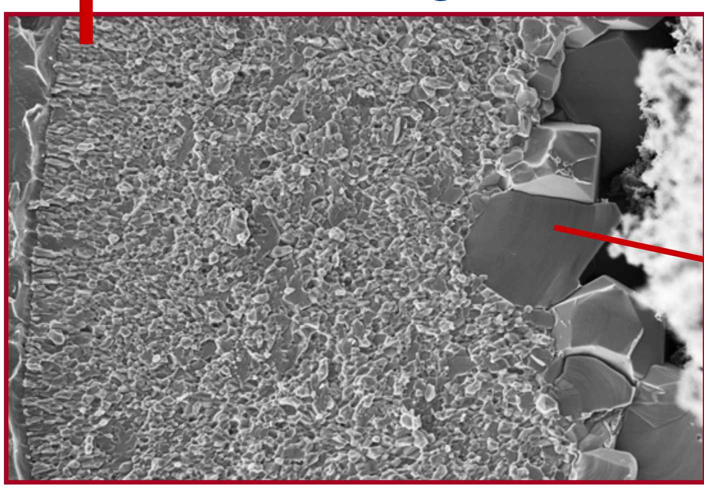
Resulting Sn gradients in wires…

Composition analysis on SMI Powder-in-Tube wire





Columnar grains



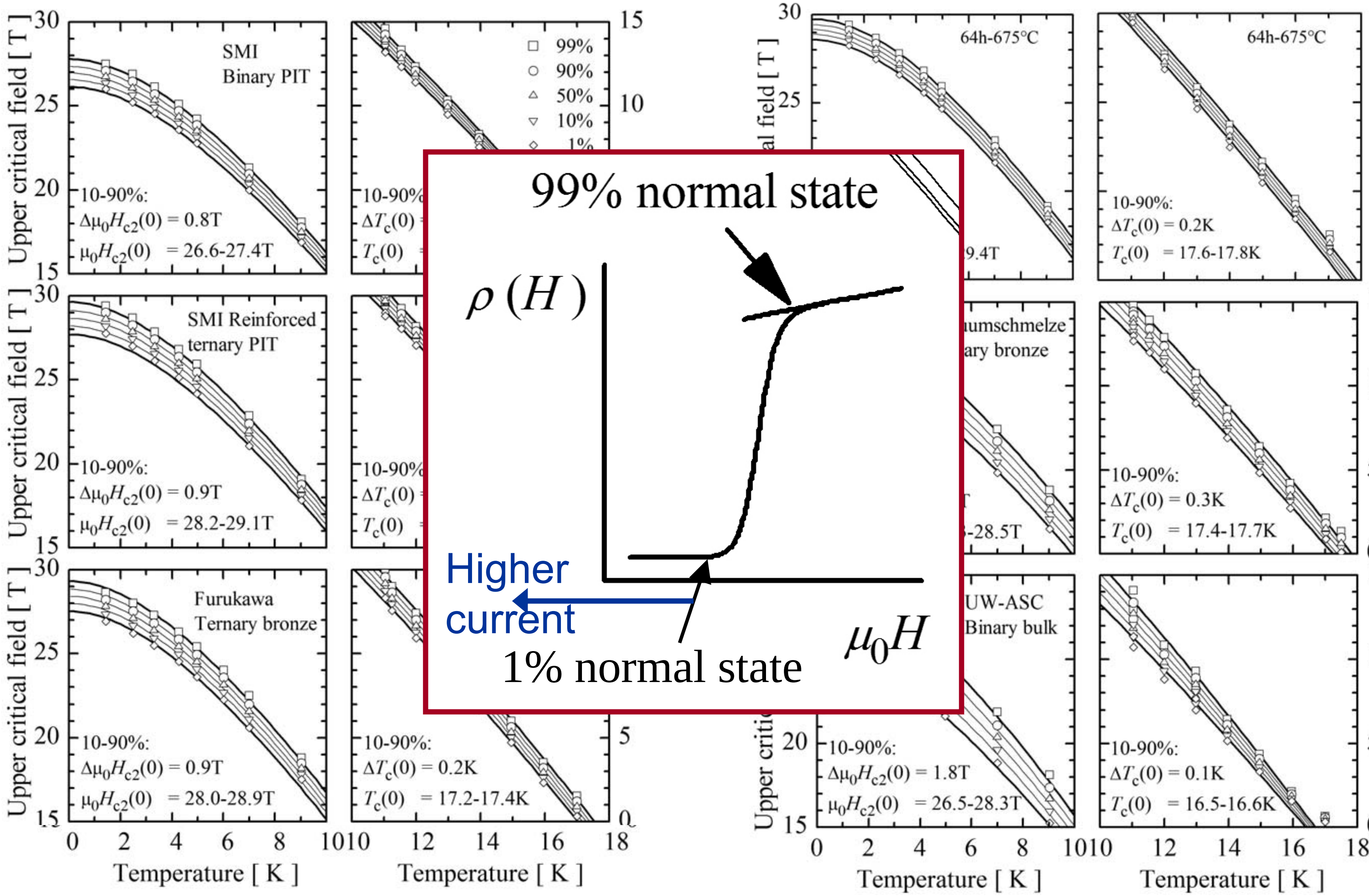
large grains (from initial Nb_6Sn_5)

Columnar grains when Sn deficient

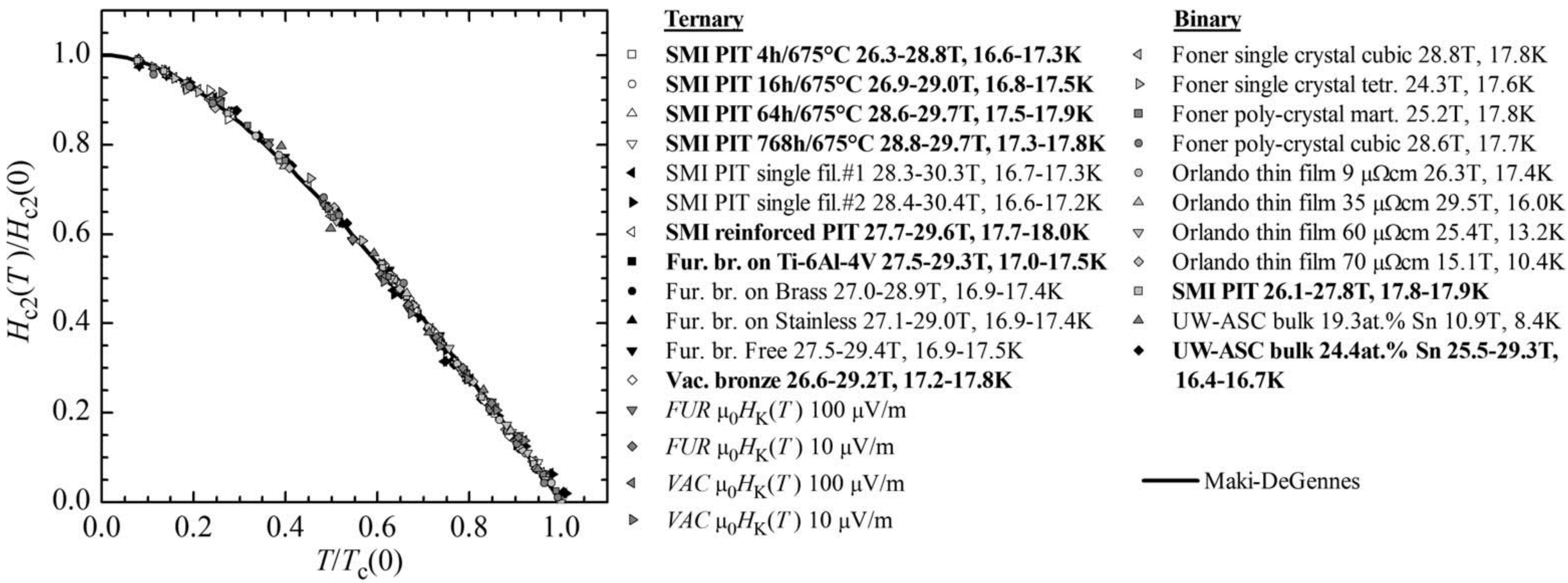
Otherwise typical 100 – 200 nm eqiuaxed

…and property gradients

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



Normalized $H_{c2}(T)$ all available results



Shape $H_{c2}(T)$ independent of

- Composition
- Morphology
- Strain state
- Applied critical state criterion

$$\ln\left(\frac{T}{T_c(0)}\right) = \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{\hbar D \mu_0 H_{c2}(T)}{2\phi_0 k_B T}\right)$$

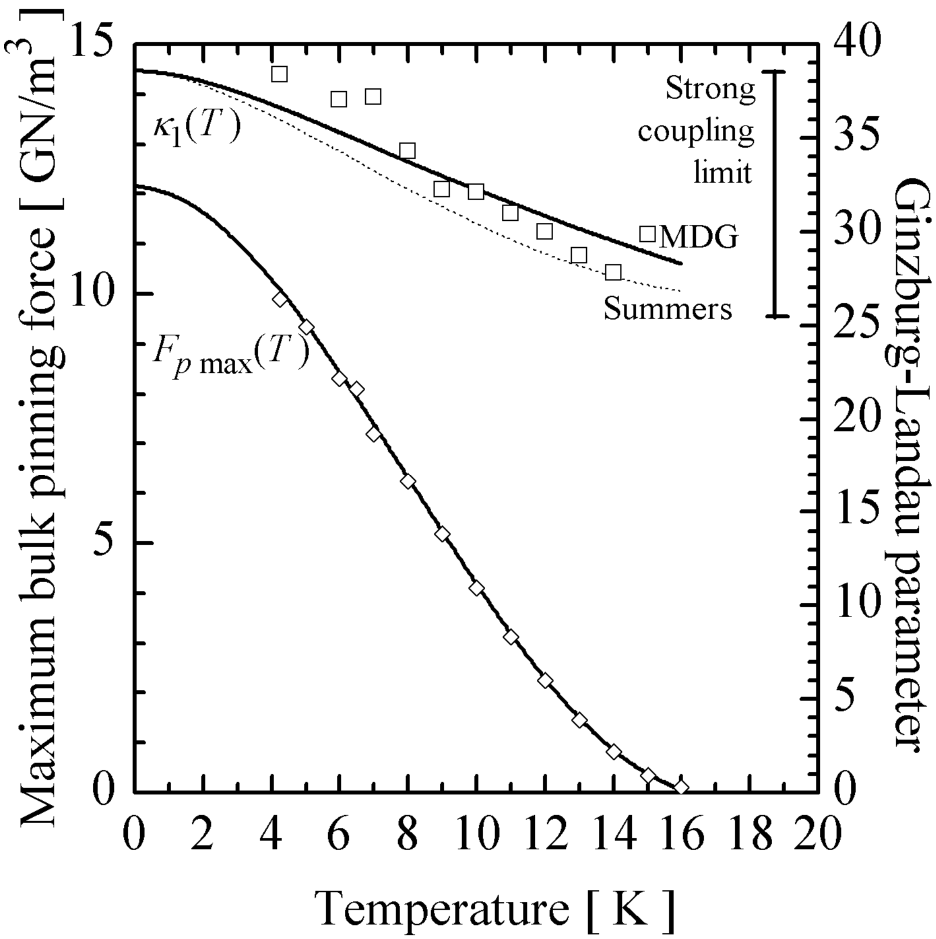
Approximation:

$$\frac{H_{c2}(t)}{H_{c2}(0)} \cong 1 - t^{1.52}, \quad t = \frac{T}{T_c(0)}$$

Ginzburg-Landau *T* dependence

Knowing $H_{c2}(T)$ and $H_c(T)$ (= $1 - t^{2.07}$ for Nb_3Sn) accurately

- means $\kappa_1(T) = \lambda(T) / \xi(T)$ can be calculated: $\kappa_1 = H_{c2} / (\sqrt{2}\, H_c)$



Weak limit:
$\kappa_1(0)/\kappa_1(T) = 1.2$

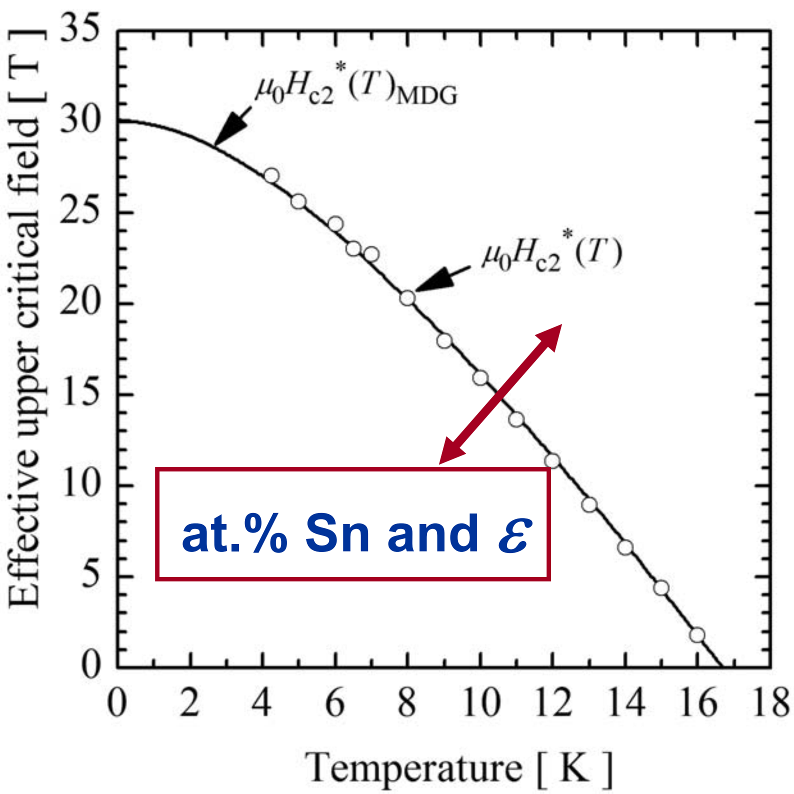
Strong limit:
$\kappa_1(0)/\kappa_1(T) = 1.5$

Rainer, J. Low T. Phys. 1974

Temperature dependence is accurately known

What determines J_c?

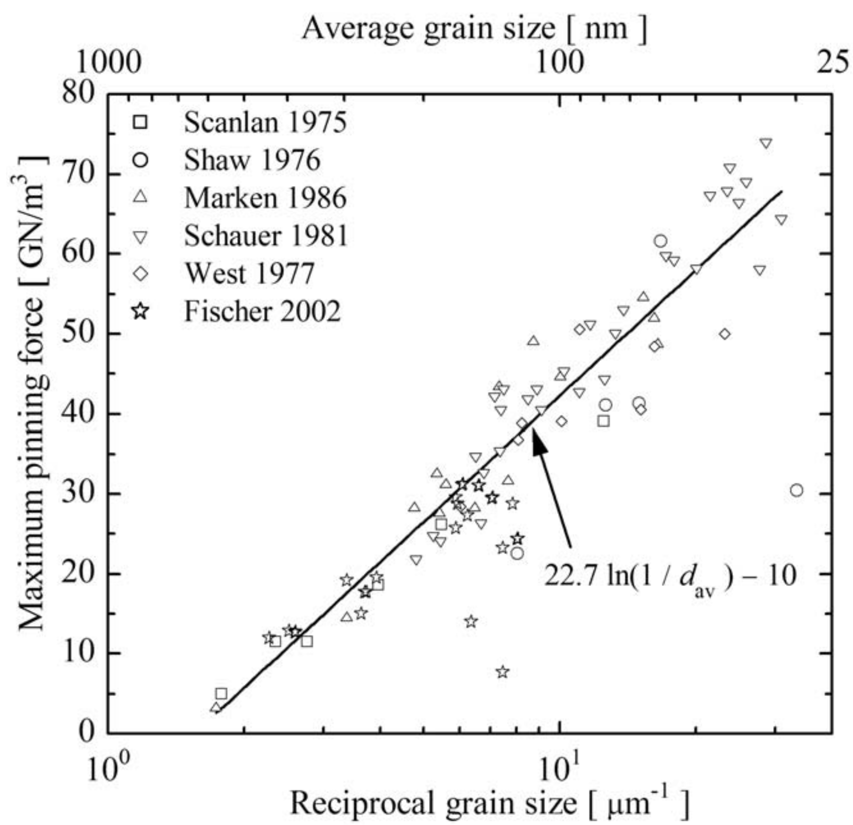
Effective $H - T$ phase boundary



- Composition
- Strain state (below)

Pinning capacity

+



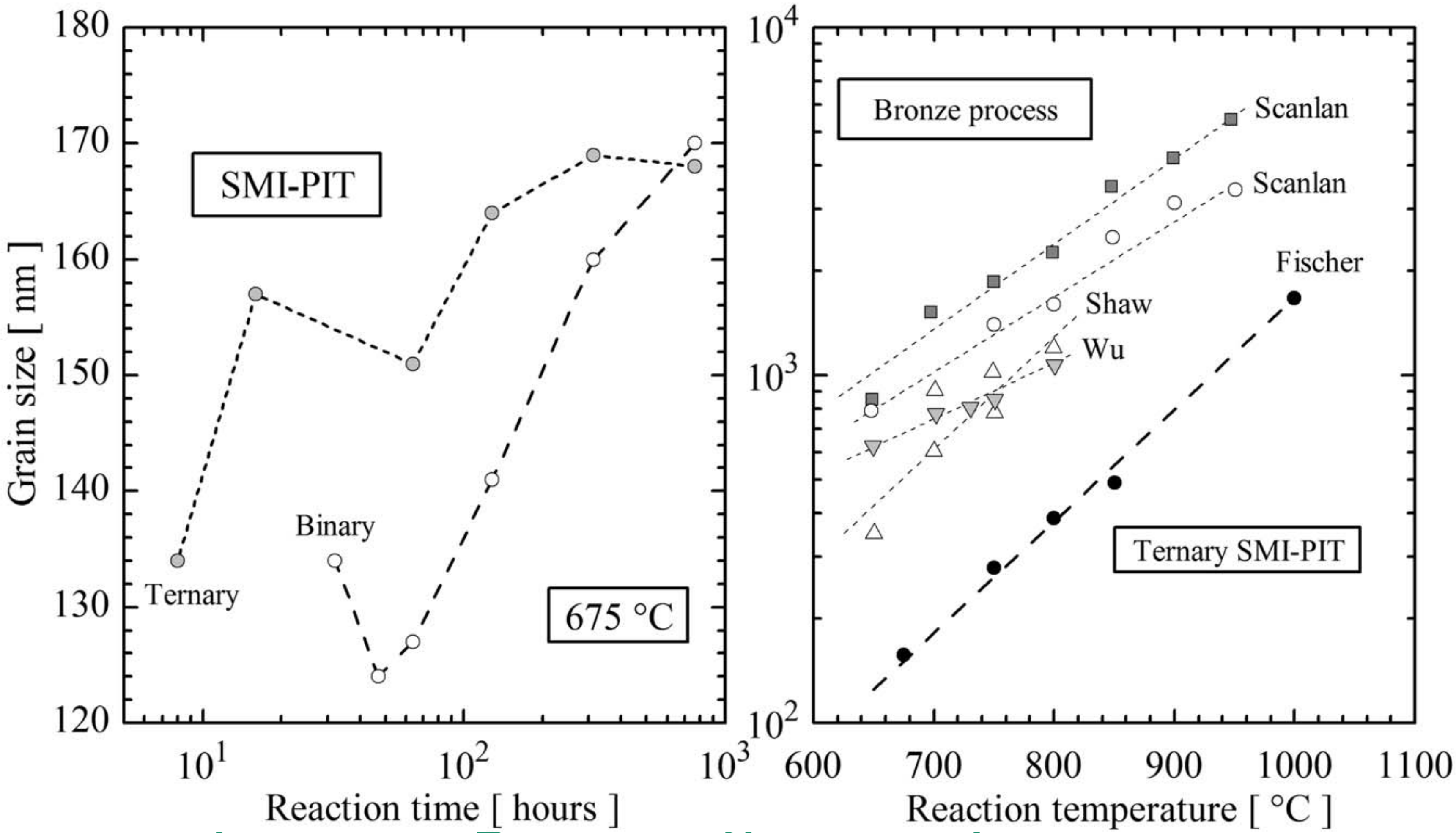
= J_c

- Average grain size

Nb$_3$Sn: Grain boundaries are main pinning centers
➔ Grain size determines F_{Pmax}

What determines grain size?

- Presence of grain nucleation points
- Reaction time and temperature
  - High *T* : Sn rich and large grains



Strain sensitivity

Strain ➔ Lattice deformations

- Modification of phonon modes and DOS
- All compositions requires interaction strength independent theory (Eliashberg based)

contains also $N(E_F)$

$$\lambda_{ep} = 2\int \frac{\alpha^2(\omega) F(\omega)}{\omega} d\omega$$

$$\lambda_{eff} = \frac{\left(\lambda_{ep} - \mu^*\right)}{\left(1 + 2\mu^* + 1.5\lambda_{ep}\mu^* e^{-0.28\lambda_{ep}}\right)}$$

$f(\varepsilon)$

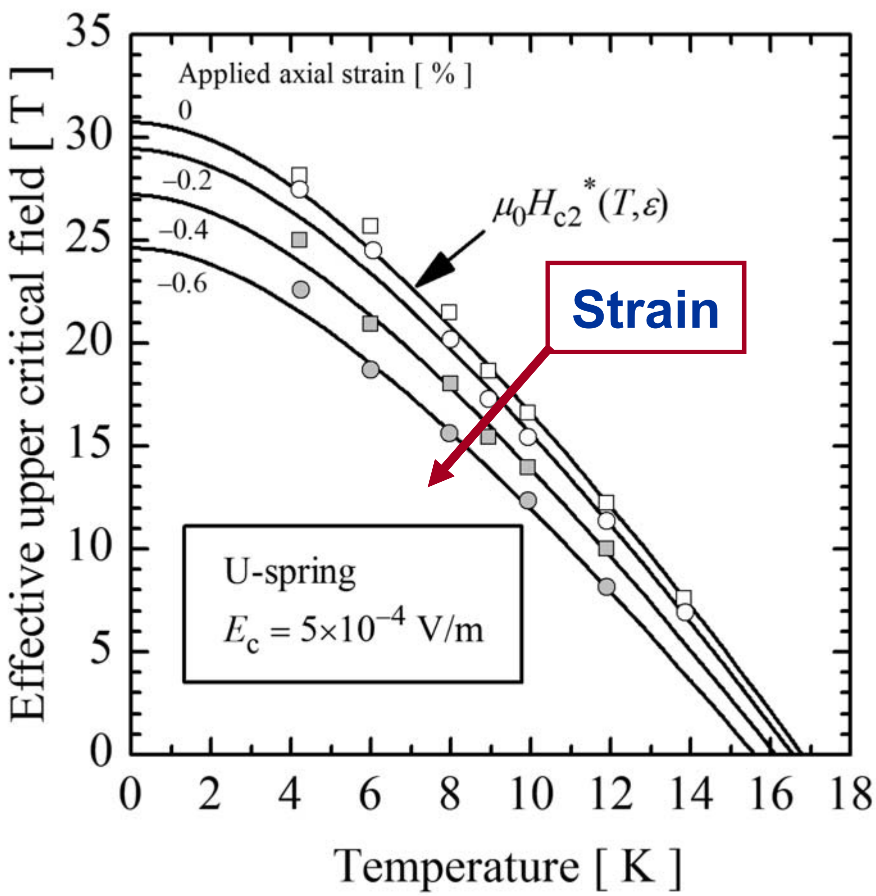
$$T_c = \frac{0.25\left\langle \omega^2 \right\rangle^{\frac{1}{2}}}{\left(e^{2/\lambda_{eff}} - 1\right)^{\frac{1}{2}}}$$

$$\mu_0 H_{c2} = \ldots$$

- Promising work: W.D. Markiewicz (NHMFL) and S. Oh (KBSI)

Strain sensitivity of $H_{c2}(T)$ (wires)

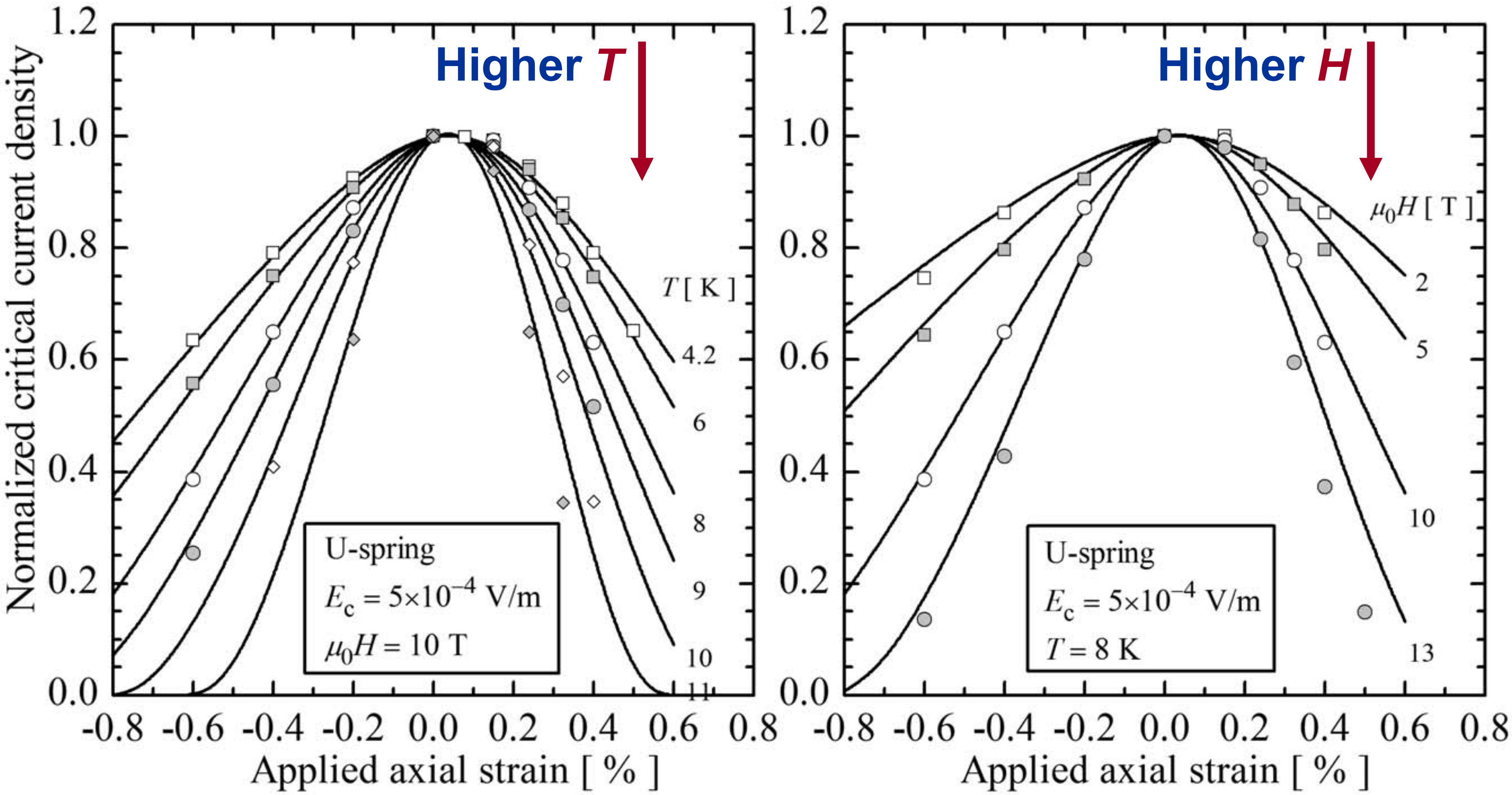
- Longitudinal strain effects on *effective* $H_{c2}(T)$*



- Strain and composition have similar effects
  - Need for a separation of parameters

Strain sensitivity of $J_c(H,T)$

J_c(10 T, T, ε_{axial}) J_c(H, 8 K, ε_{axial})

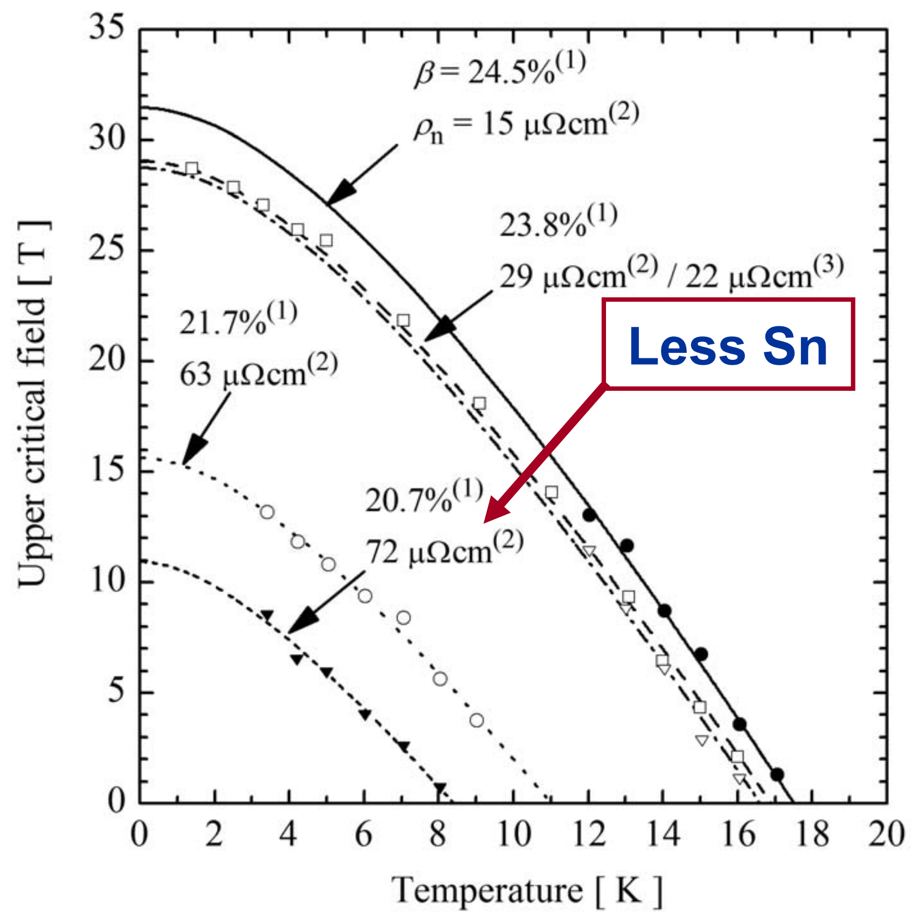


- Why is strain sensitivity increased at higher H and T ?
- Strain negligible at 4.2 K and < 1 T? (T_c: ~ -2 K / % strain)

Strain sensitivity versus composition

At higher *H* and *T:*

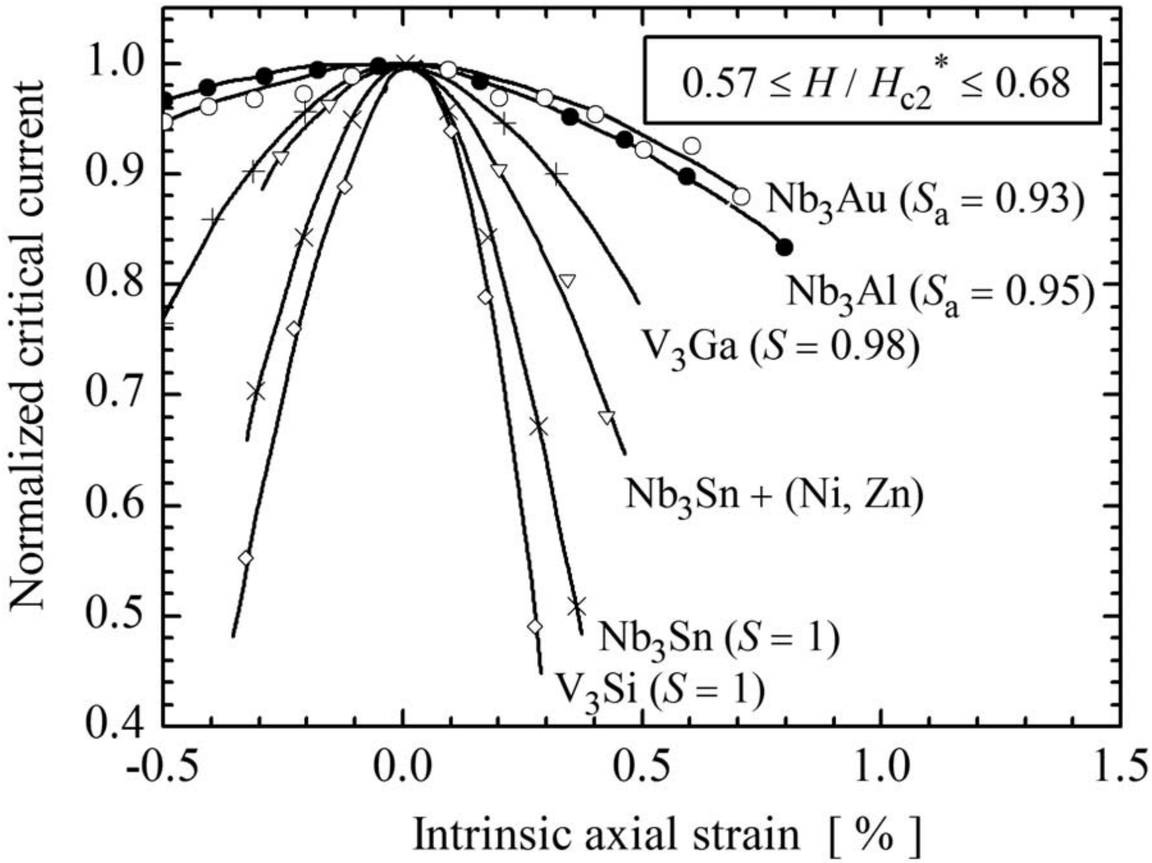
- Low Sn A15 sections "die out"
  - High Sn sections determine SC properties
- Increased strain sensitivity
  - Is Sn rich A15 more strain sensitive than Sn poor A15 ?



- Does optimization through Sn enrichment cause higher strain sensitivity?

Strain sensitivity versus LRO

- *S* → Bragg-Williams order parameter

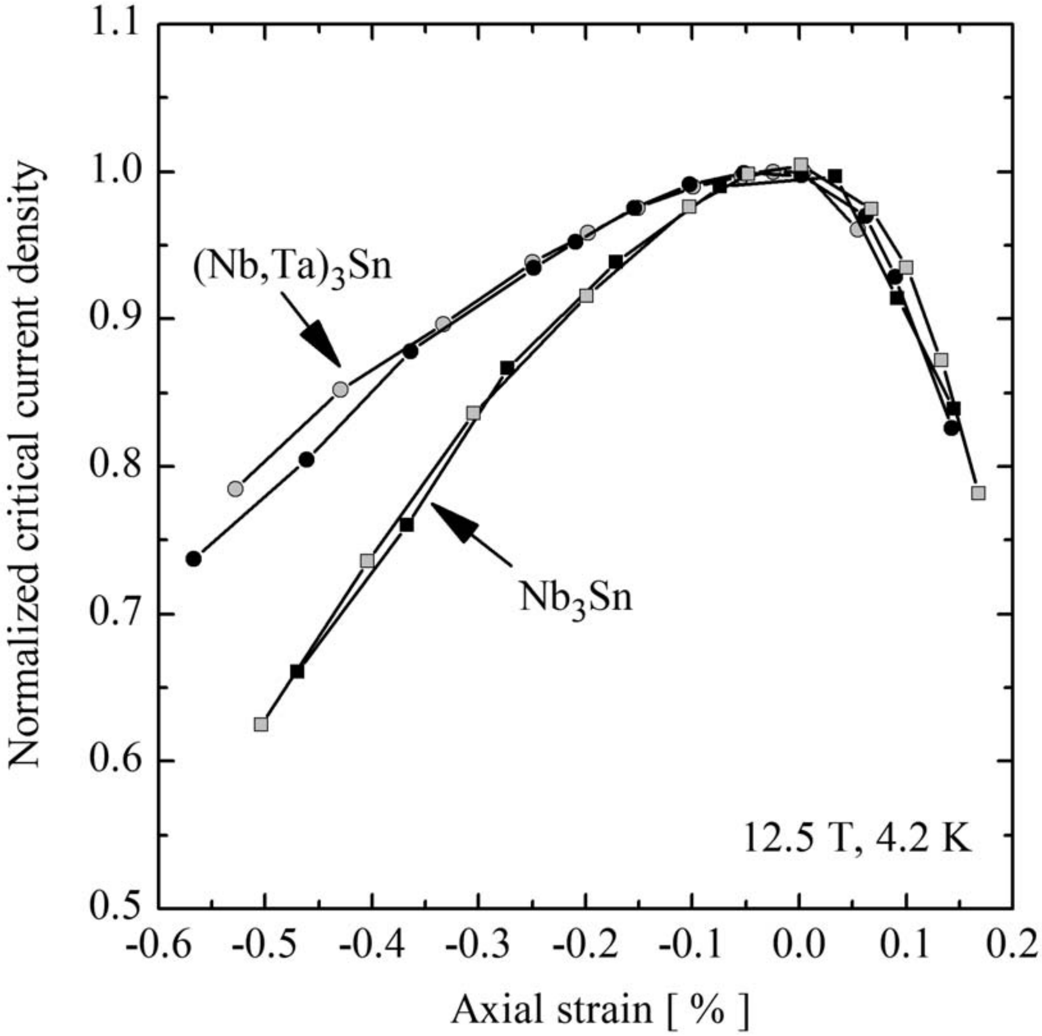


- Flükiger, ACE 1984

- Higher LRO (≙ more Sn in Nb_3Sn) → larger strain sensitivity

Strain in ternary and binary wires

- Alloyed → more disorder → reduced strain sensitivity?



Summary

- Nb_3Sn prefers stoichiometry
  - High T_c and ρ_n
- Watch out for:
  - Diffusion gradients
  - Tetragonal distortion above 24.5%
- Large grains easily obtainable (high *T* reaction + plenty Sn)
  - At the cost of pinning capacity
- Coupling constant independent theory is required (>23 %Sn)
- We're scratching the fundamental basis of strain dependence
  - If successful, is generalization possible?
  - Strain dependence appears more severe approaching stoichiometry

More info

- PhD Thesis (2005)
  - A. Godeke, "Performance Boundaries in Nb_3Sn Superconductors" Available on request: agodeke@lbl.gov
- Topical Reviews
  - A. Godeke, "A review of the properties of Nb_3Sn and their variation with A15 composition, morphology and strain state", *Supercond. Sci. Techn*. 19 R68 (2006) (invited)
  - A. Godeke *et al*., "A general scaling relation for the critical current density in Nb_3Sn", *Supercond. Sci. Techn*. 19 R100 (2006)
- Journal articles
  - A. Godeke *et al*., "The upper critical field of filamentary Nb_3Sn conductors", *J. Appl. Phys*. 97, 093909 (2005)
  - A. Godeke *et al*., "Inconsistencies between extrapolated and actual critical fields in Nb_3Sn wires as demonstrated by direct measurements of H_{c2}, H^* and T_c", *Supercond. Sci. Techn*. 16 1019 (2003)