

# How Superconductors became practical

A walk through history and science of flux pinning

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... a field of 100 000 Gauss could then be obtained in coils of say 30 centimeters in diameter.

... producing intense magnetic fields with the aid of coils without iron cores.

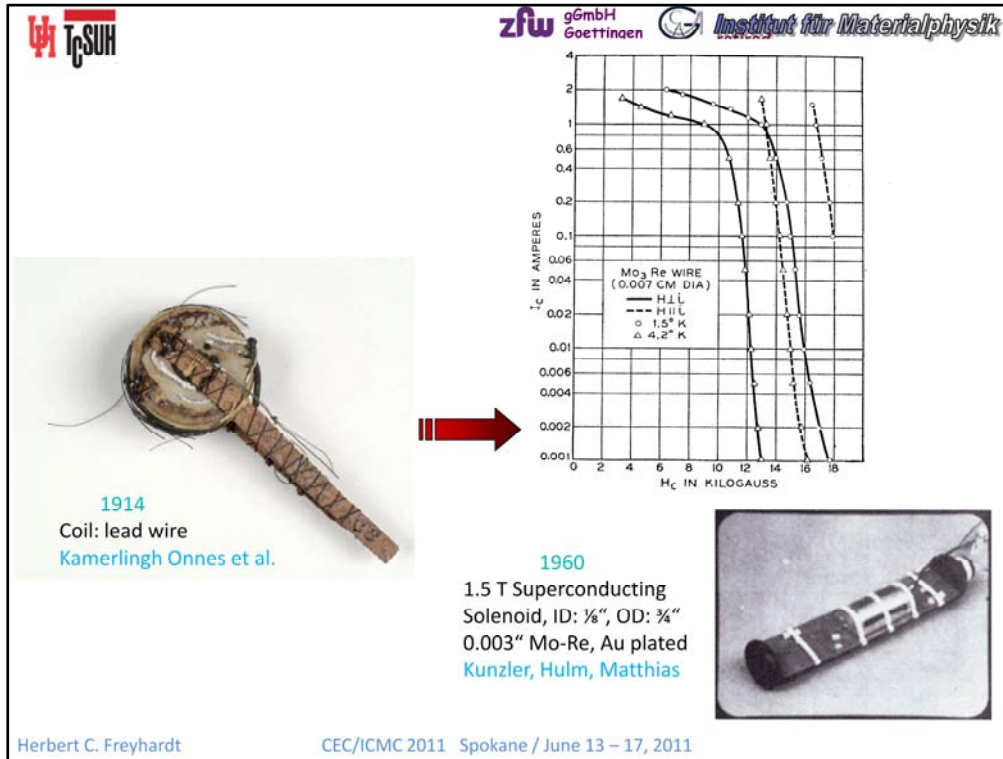
The appearance of resistance in superconductors which are brought into a magnetic field, at a threshold value of the field, ...

The centennial event:  
**1911 → 2011**  
 100 years since the discovery of superconductivity in mercury by **Heike Kamerlingh Onnes**

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In April 1911, a new age in physics began when Kamerlingh Onnes could demonstrate the disappearance of resistivity in mercury at 4.2 K, which marks the discovery of superconductivity. In December 1912, mercury was joined by tin and lead, with a transition at 3.8 K and 7.2 K, respectively. Already in the fall of 1913, at the 3<sup>rd</sup> International Congress of Refrigeration in Chicago, Kamerlingh Onnes pointed out the more than attractive possibility to generate magnetic fields of 100 000 Gauss in a coil, a solenoid, of only 30 cm in diameter, cooled down to liquid He temperatures. These dreams, however, were sustainably shattered when he and his coworkers found out that a magnetic field would destroy superconductivity. In a coil wound from Pb wires, superconductivity disappeared in a field of only 600 Gauss at 4.25 K. It took then decades to explain the phenomenon of superconductivity and to process and understand what we now call type-II superconducting materials, which can carry loss-less currents also in high magnetic fields of, e.g., 10 T, which is about the upper critical field of the alloy superconductor NbTi at 4.2 K.



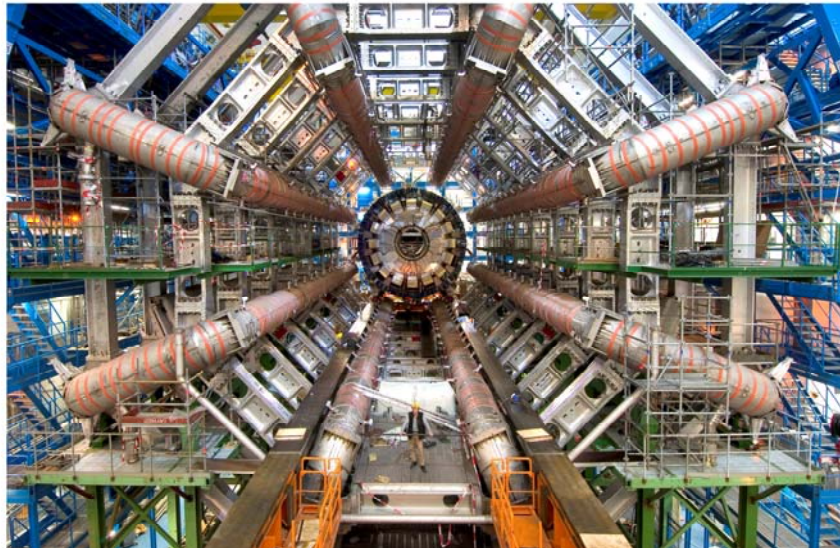


The progress can further be exemplified by considering the development of coils or coil systems to generate magnetic fields.

In 1914, the field of only 600 gauss destroyed superconductivity at a temperature of 4.25 K in the small coil wound by Kamerlingh Onnes from lead wires, already at a field of only 600 Gauss.

Although in 1955 Yntema reported on the first superconducting electromagnet wound from Nb, it was Hulm's discovery (1955) of superconductivity in ductile Mo-Re alloys, which allowed Kunzler et al. in 1960 to process superconducting wires with attractive current densities and to wind a superconducting solenoid, which generated a magnetic field of 1.5 T.

Nowadays possibilities and engineering skills are impressively demonstrated by the ATLAS detector magnet of CERN (next slide)



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The ATLAS particle detector magnet of the Large Hadron Collider at CERN, which is searching for new discoveries in the head-on collision of protons at ultrahigh energies. Depicted is the Barrel Toroid, the largest superconducting magnet ever built. It consists of 8 well aligned rectangular-shaped superconducting coils, 5 m wide and 25 m long. The Barrel Toroid can store an energy of about 1GJ and weights 100 tons.





... to become practical for, e.g.:

**Energy savings**

 SMES
  LIPA cable
  ISFCL

**Rotating Machinery**

 5 MW motor AMSC
  Windgenerator ENERCON

**Medical Imaging**

 MRI

**High-Energy Physics**

 LHC dipole CERN

Specific LTS/HTS needed

- stable conductors for high enough operational temperatures
- sufficient mechanical strength and good electromechanical properties
- protection against thermal quenching and flux jumping
- stable conductors, which can be manufactured with a robust processing, with sufficiently high yield and throughput, i.e., in an economically viable fashion with the specification needed for applications

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### ... to become practical. What does this mean?


Based on an understanding of the origin of superconductivity one has to economically process superconducting wires for devices which will exhibit a better performance than conventional, normal conducting equivalents or which allow to exploit new features of a superconductor.

Essential for the application of superconductors in electrical and power engineering is the control of critical currents as a function of temperature of operation, magnetic field, and field direction in a conductor which can be robustly produced with an optimum cost-performance relation. To a large extent this means the control of flux pinning. Therefore, one can use the progress achieved in flux pinning - which is determined by (i) the basic understanding of the physics of superconductors, and (ii) by an optimum layout of the pinning landscape which is given by the well-designed microstructure of optimally processed superconductors - as a valuable guide for bringing superconductors to the marketplace.

The impressive progress achieved over the years in producing (high) current carrying conductors can best be exemplified by starting from the early mercury filled quartz capillaries, via commercial NbTi alloy or Nb<sub>3</sub>Sn compound conductors, all the way to a modern power cable.

## Essential for the application of superconductors

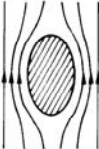
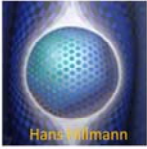
- Control of critical currents versus temperature, magnetic field and field angle, which is directly connected to a
  - Control of flux pinning



## The Early Days & Importance of Flux Pinning


- **1933: W. Meissner, R. Ochsenfeld**

  - complete flux expulsion; type-I superconductor
  - supercurrents flow only on the surface

- **1933/4 ff: Leo Shubnikov et al.**

  - complete flux expulsion confirmed
  - metallic alloy sc: mixed state with partial flux penetration  
→ Shubnikov phase





- **1935 Fritz & Heinz London**

London equations : electromagnetic properties of a superconductor

$$\mathbf{E} = \partial(\mathbf{A}_s)/\partial t \quad \nabla \times (\mathbf{A}_s) = -\mathbf{B}$$

$$\Lambda = m_s / (n_s e_s^2) = \mu_0 \lambda^2$$


$\lambda$ : London penetration depth


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One did not understand the phenomenon of superconductivity before 1957, when the microscopic theory of superconductivity was developed by Bardeen, Cooper and Schrieffer. However, before that time, the thermodynamic behavior of superconductors was studied and far reaching conclusions could be drawn, which brought quite some insight into the origin of superconductivity. Not only the resistivity disappears in the superconducting state but also the magnetic field is completely expelled from what's now called type-I superconductor, as Meissner and Ochsenfeld could demonstrate in 1933. As a consequence, the superconducting currents are flowing only in a thin layer on the surface of the superconductor. Complemented with the artist's (Hans Hillmann, who developed NbTi conductors at the Vacuumschmelze) view of flux expulsion. Leo Shubnikov (founder of low-temperature physics in the Ukraine, worked at Leiden, which he left 1930) and his coworkers confirmed these findings, and moreover discovered that in alloys (e.g., Pb alloys) magnetic flux penetrates the superconductor between the lower and the upper critical fields  $H_{c1}$  and  $H_{c2}$ , respectively (for which they used the original notation  $H_{k1}$  and  $H_{k2}$ ). These materials are now called type-II superconductors. The basic electrodynamic properties of superconductors were well described phenomenologically in 1935 by the two brothers Fritz and Heinz London. The London equations are derived by assuming a metal, which is an ideal conductor and exhibits expulsion of magnetic fields. This leads to the first important characteristic length scale, namely the London penetration depth  $\lambda$ .







## The Early Days & Importance of Flux Pinning ctd.



V. L. Ginzburg



Lev D. Landau



A. A. Abrikosov

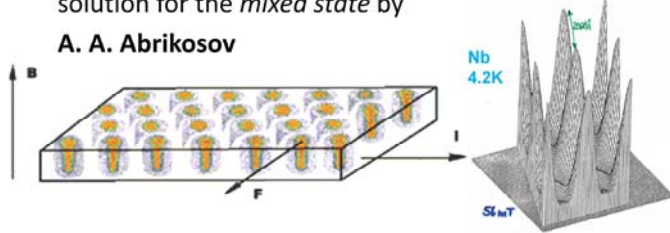
➤ **1950 ff: Ginzburg-Landau, Abrikosov (1957)**

Phenomenological theory of superconductivity based on a complex pseudowavefunction  $\psi$  as order parameter, where  $|\psi|^2$  is the density of sc electrons

$$F_s - F_n = \alpha|\psi|^2 + (\beta/2)|\psi|^4 + (1/2m^*)|(-i\hbar\nabla - 2e\mathbf{A})\psi|^2 + \mu_0 h^2/2$$

Ginzburg-Landau free energy per u.v. → **Ginzburg-Landau equations** ;  
 solution for the *mixed state* by

**A. A. Abrikosov**



**FL's & FLL**

$\phi_0 = 2 \times 10^{-15} \text{ Tm}^2$   
*flux quantum*

$\langle B \rangle = 2\phi_0 / (a_{FL}^2 \sqrt{3})$   
 $a_{FL}$  = FL spacing

$B_{c2} = \phi_0 / (2\pi\xi^2)$   
*upper critical field*

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The theoretical understanding and thermodynamic treatment brought the phenomenological theory of Ginzburg and Landau, which is based on a complex pseudo-wavefunction  $\psi$  as order parameter, whereby the Ginzburg-Landau free energy depends on the local density of superconducting electrons  $n_s = |\psi(x)|^2$ .

The solution for the flux distribution in the mixed state was finally derived by A. A. Abrikosov. Magnetic flux penetrates the type-II superconductor above  $B_{c1}$  in the form of flux lines (FLs), each carrying one flux quantum  $\phi_0 = 2 \cdot 10^{-15} \text{ Tm}^2$ , with a flux line spacing,  $a_{FL}$ , given by  $B a_{FL}^2 = 2\phi_0 / \sqrt{3}$ , until at  $B_{c2} = \sqrt{2} \kappa B_{cth} = \phi_0 / 2\pi\xi^2$  ( $B_{cth}$  is the thermodynamic critical field) the material is completely filled with flux. Three most important length scales are introduced here: the spacing of flux lines  $a_{FL}$  (which is  $150 \text{ nm} / \sqrt{B}$ , where  $B$  is given in kG or 0.1 T), the coherence length  $\xi$ , which is the Ginzburg-Landau coherence length, and  $\lambda$ , the penetration depth of the magnetic field, where  $\kappa = \xi/\lambda$  denotes the Ginzburg-Landau parameter.

# The Early Days & Importance of Flux Pinning ctd.



## ➤ 1950 ff: Ginzburg-Landau, Abrikosov (1957)

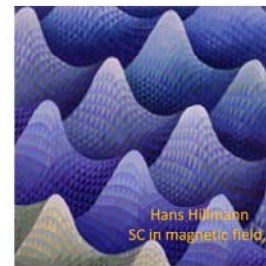
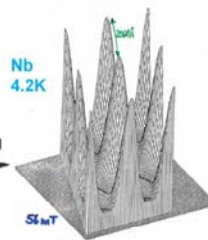
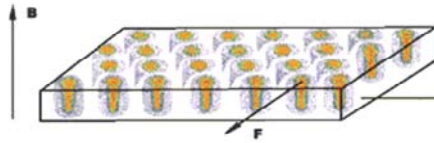
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Ginzburg-Landau free energy per u.v. → **Ginzburg-Landau equations** ;

solution for the *mixed state* by

**A. A. Abrikosov**



Now with Hans Hillmann's artistic imagination of a flux line distribution in a type-II superconductor.

## The Early Days & Importance of Flux Pinning ctd.

### Transport currents in the mixed state of a type-II superconductor?

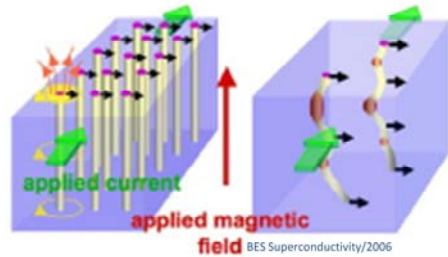
FLs move under the action of a Lorentz force

$$\mathbf{F}_L = \mathbf{J} \times \mathbf{B}$$

and dissipate energy (J. Bardeen & M. J. Stephen, 1965)

**FLs must be pinned** to create a flux gradient  $\nabla \times \mathbf{B} / \mu_0 = \mathbf{J}$  and a **volume pinning force**  $\mathbf{F}_p$  has to balance  $\mathbf{F}_L$

$$\mathbf{F}_p = \mathbf{F}_L = \mathbf{J}_c \times \mathbf{B} \quad \mathbf{J}_c : \text{critical current density}$$

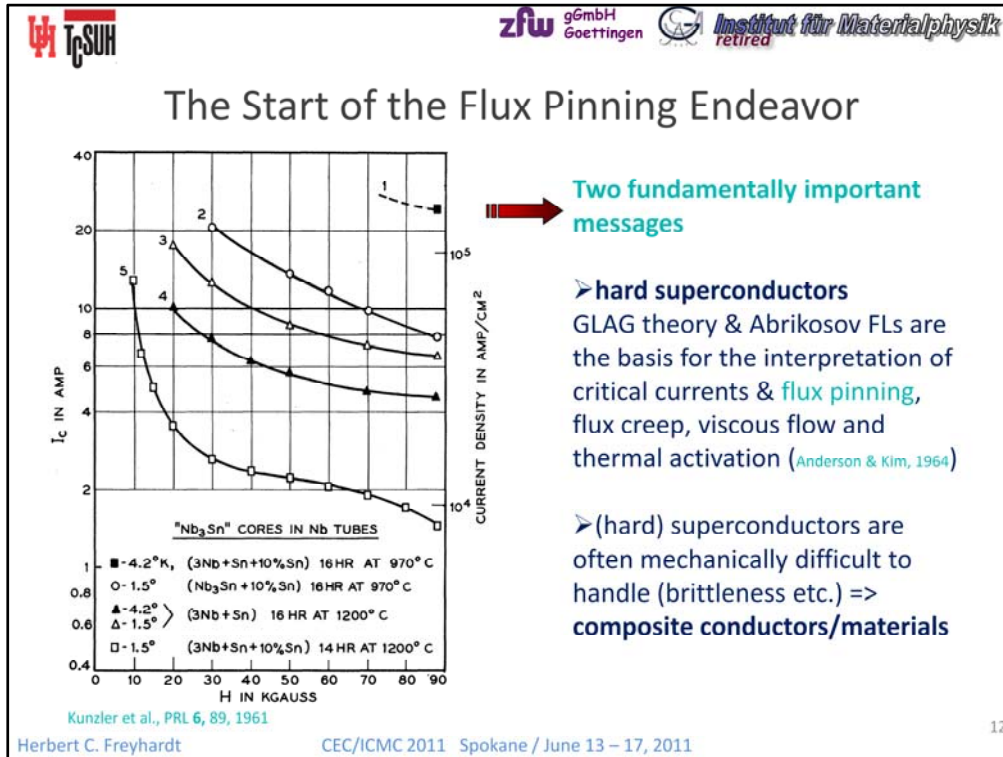


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If one passes a current of density  $\mathbf{J}$  through an ideal, reversible type-II superconductor (in the mixed state) in an external magnetic field, a Lorentz force  $\mathbf{F}_L = \mathbf{J} \times \mathbf{B}$  is exerted on the flux lines, which move and dissipate energy, and no lossless current is observed. A non-zero (transport) current density becomes only possible if a gradient in the flux density,  $\nabla \times \mathbf{B} / \mu_0 = \mathbf{J}$ , is established by pinning the flux lines by microstructural defects to prevent the FLs from moving. The critical current density  $\mathbf{J}_c$  is reached when the Lorentz force is balanced by the volume pinning force  $\mathbf{F}_p = \mathbf{F}_L = \mathbf{J}_c \times \mathbf{B}$ .

Again, with Hans Hillmann's artistic view of flux pinning



In 1961, Kunzler et al. from Bell Labs reported: “We have observed superconductivity in  $Nb_3Sn$  at average current densities exceeding  $100\,000\text{ A/cm}^2$  in magnetic fields as large as  $88\text{ kG}$ ”. In the years before, Pb, Pb alloys, Nb,  $Mo_3Re$  etc. showed only moderate critical currents ... nonetheless enabling the use of superconducting wires to manufacture, e.g., superconducting solenoid magnets.

These experiments, together with the introduction of the term ‘**hard superconductors**’ and Anderson & Kim’s discussion strengthening the implications of the GLAG theory and particularly the importance of the quantized flux lines (FLs) in the mixed state for flux pinning, critical states, flux creep, flux flow and thermal activation, paved the way for a deeper understanding of the irreversible properties of these materials.

Neutron diffraction investigations of Cribier et al. in 1967 clearly demonstrated the existence of a regular flux line lattice (FLL). FLs and defects in the FLL were then visualized by Träuble & Essmann (since 1967) and consequently a detailed investigation of the pinning behavior of superconductors & hard superconductors commenced.



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**Flux Vortices and Transport Currents in Type II Superconductors**

By A. M. CAMPBELL and J. E. EVETTS  
Department of Metallurgy and Materials Science,  
University of Cambridge, Cambridge, England




**Reinhausen, March 1982**

PROCEEDINGS  
OF THE  
INTERNATIONAL DISCUSSION MEETING  
ON  
FLUX PINNING IN SUPERCONDUCTORS

23.–27. September 1974  
INTERNATIONALES HAUS SONNENBERG  
St. Andreasberg/Harz  
Germany

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**Fukuoka, November 1985**

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Matsushita, Jan Evetts, Herb Freyhardt, Doug Finnemeore, Irie san, Archie Campbell, John Clem, Jack Ekin, David Larbalestier, Peter Kes, Si Foner; Chris Heiden. On top slide in addition: Brandt, Küpfer, Welsh


Allow me to mention some personal memories:

At LT11 (11<sup>th</sup> Conference on Low Temperature Physics), 1968 in St. Andrews (when superconductivity at 100 K was predicted by a novel pairing mechanism and our colleagues from the former USSR were forced to disappear after the invasion of Czechoslovakia (Alexander Dubček era)) a core group interested in pinning studies met for the first time (Campbell, Evetts, Dew-Hughes, Clem, Deutscher, Flükiger, Foner, Freyhardt, ... , not always investigating pinning directly at that time.)



Shortly afterwards, the standard publication “*Flux Vortices and Transport Currents in Type-II Superconductors*”, was published in 1972 by Archie Campbell & Jan Evetts, in *Advances in Physics* (**21**, 1972, 199 – 428).

Subsequently, in September 1974 the first *INTERNATIONAL DISCUSSION MEETING ON FLUX PINNING IN SUPERCONDUCTORS* was held in Sonnenberg /Germany by Peter Haasen and myself, where most of the essential players in the field gathered. This commenced a series of discussion meetings, which were regularly organized in Europe, USA and Japan (globalization of science!), e.g. at Reinhausen (Germany) in 1982 and at Fukuoka (Japan) in 1985.



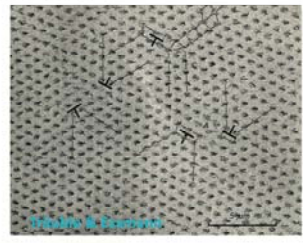


## Flux Pinning: Essential Issues

**Pinning obstacles**

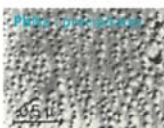
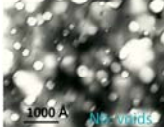
- 0D: point defects, vacancies, ...
- 1D: dislocations, nanorods, ...
- 2D: grain boundaries, stacking faults, surface



reversible type II superconductor

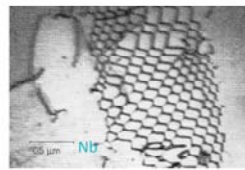
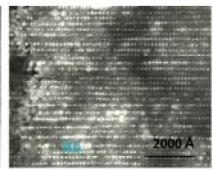
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graph LR
    A[reversible type II superconductor] --> B[statistics summation]
    B --> C[macroscopic behaviour]
    C --> D[obstacle distribution and geometry]
    D --> B
    E[elementary interaction force p_max] --> B
    B --> F[Pv / Fp]
    
```

barrier, ... , intrinsic layered structure, ...

- 3D: precipitates, voids, bubbles, ... , nanoparticle inclusions, ... , magnetic particles, ...

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To visualize the key flux pinning issues, let us use one of the central schemes (H. C. Freyhardt; Sonnenberg ,1974), to discuss the essential ingredients:

**Geometry and distribution of the pinning obstacles**, i.e., the pinning landscape have to be investigated, if necessary with high-resolution techniques. A few examples:

- 0 dimensional: vacancies, point defects, ...
- 1D: dislocations (in Nb, pinned by radiation induced defects), nanorods, ...
- 2D: grain boundaries (V<sub>3</sub>Ga: 700°C / 48h ;Tachikawa et al.), stacking faults, ...
- 3D: precipitates (in Pb-Na alloys), voids (produced by irradiation of Nb with 3.5 MeV <sup>58</sup>Ni<sup>+</sup> (3x10<sup>16</sup> ions/cm<sup>2</sup>)@ 900°C)

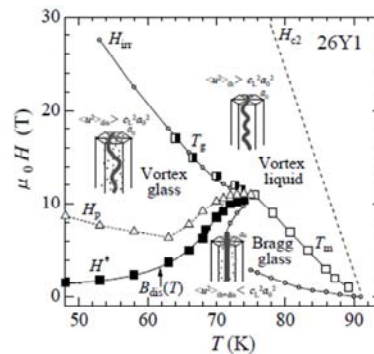
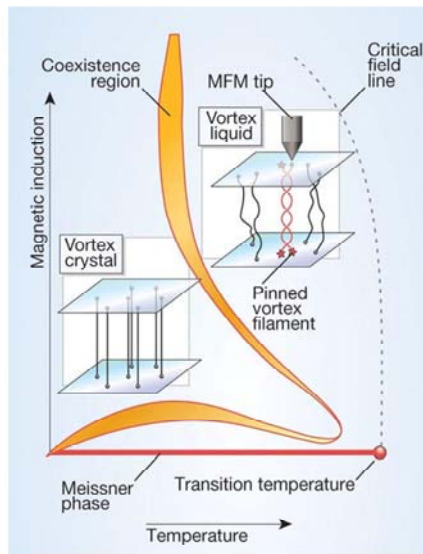
It is possible to generate regular obstacle arrays:

- Screw dislocation networks in Nb single crystals, twisted by 124°/cm + heat treated at 1150°C/30min
  - Regular void array: Nb with 900wt.%O<sub>2</sub> ,irradiated @ 785°C, 6x10<sup>16</sup> Ni<sup>+</sup>ions/cm<sup>2</sup>
- Träuble & Essmann visualized the flux line lattice/arrangement in the mixed state of a type-II superconductor by an elaborate decoration technique.

The use of model systems to investigate individual elementary interaction mechanisms as well as the summation problem paved the way for an optimization of practical conductors.



## Flux Pinning: Essential Issues ▶ Reversible superconductor, ctd.




Complex H-T Diagram for HTS/YBCO

Nishizaki & Kobayashi, SUST 13 (2000)1-11

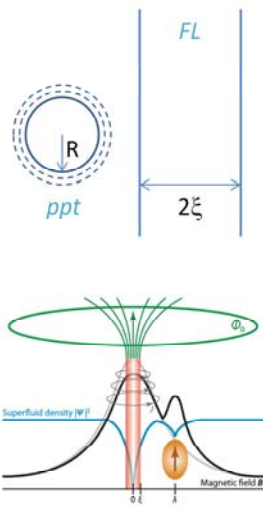
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The anisotropy and the layered structure of **high-temperature superconductors** (HTS) results in a more complicated flux structure. For the (highly) anisotropic HTS with their layered appearance, the flux lines can decompose into a stack of pancake vortices.



## Flux Pinning: Essential Issues ▶ Elementary Pinning Force



- Local variations of superconducting GL parameters  $\alpha$  and  $\beta$ , which means of  $H_{c2}$  and  $\kappa$ :

$$\delta F_{GL} = \int d^3r \mu_0 H_c^2 \{ -(\delta H_{c2}/H_{c2}) |\psi_r|^2 + (\delta \kappa/\kappa) |\psi_r|^4 \}$$

$$= \int d^3r \mu_0 H_c^2 \{ -(\delta H_c/H_c) |\psi_r|^2 - (\delta \kappa/\kappa) (|\psi_r|^2 - |\psi_r|^4) \}$$

( in reduced units )

⇒ core pinning interaction  
(normal ppt or void of volume  $V = 4\pi R^3/3$ ;  $R < \xi$ )  
 $\delta E = V \mu_0 H_c^2 / 2$  and  $f_p = \delta E / \xi$

- Dielastic and parelastic interaction
- Magnetic interaction of obstacles with permanent magnetic moments
- Interaction with phase boundaries
- ❖ but for HTS: more complicated vortex structure !!


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**Elementary pinning interaction:** Local variations of the superconducting Ginzburg-Landau free energy: →  $\delta l$  &  $\delta T_c$  pinning, or  $\delta \kappa$  &  $\delta H_c$  pinning (length scale:  $\xi$ ). Optimum interaction forces are introduced by pinning centers of the size of the coherence length  $\xi$ . The core interactions leads to a very effective pinning force.

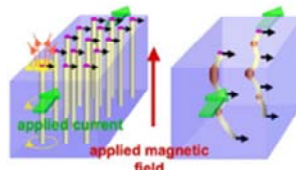
Elastic interactions: *parelastic* (normal conducting core of a FL with  $\Delta V/V \approx 10^{-7} \Rightarrow$  FL are surrounded by stress fields interacting with internal strain fields of defects), *dielastic* (stress field of defects is given by elastic constants  $C_{ik}$ , which are different in the nc & sc state ( $\Delta C_{ik}/C_{ik} \approx 10^{-4}$ ) causing an energy variation if a FL moves over the defect).

**Magnetic interaction:** (i) with surfaces (Bean-Livingston barrier), length scale  $\lambda$ , or (ii) of FLs with magnetic particles.

HTS with their more complex vortex structure experience more complicated flux pinning interactions.



## Flux Pinning: Essential Issues ► Summation Problem



$N_V$  = volume density of pins;  $a_o = \sqrt{(\phi_o/B)}$  = average FL spacing  
 $d$  = obstacle/FL interaction range

**DIRECT SUMMATION:**  $F_p = N_V f_p$

**STATISTICAL SUMMATION (R. Labusch):**  $F_p = 2dN_V f_p^2 / (a_o^3 8\pi\mu_{eff})$

**THRESHOLD AND THEORETICAL CRITICAL CURRENT OR  $F_p$**  (A. Campbell / Sonnenberg 1974)

→ **DYNAMICAL THEORY** (based on: Yamafuji & Irie 1967)

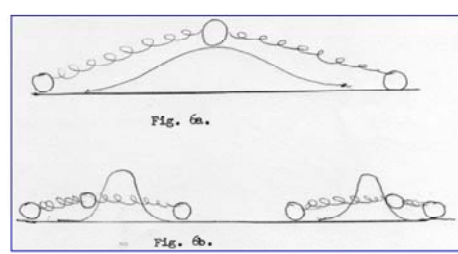


Fig. 6a.

Fig. 6b.

energy released in one event:  
 $f_p^2 / 8\pi\mu_{eff} a_o$

⇒ volume pinning force

$$F_p = \frac{2dN_V f_p^2}{8\pi\mu_{eff} a_o^3}$$

**COLLECTIVE FLUX PINNING** (Larkin & Ovchinnikov 1979)


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Central to the determination of  $J_c$  is the **summation problem**, which determines the way by which the individual interactions of the FLs and the pinning centers in a unit volume are summing up to a pinning force per unit  $\text{cm}^3$ , which very much is determined by the elasticity of the FLL.

*Summation* represents a central problem for different hardening phenomena in solids. P. Haasen, working together with R. Labusch (1969), developed a general view of the hardening of materials; mechanical, magnetic and superconducting hardening. They considered the interaction of dislocations with precipitates or solute atoms, Bloch walls with obstacles, and FLs with lattice defects. Labusch developed a statistical summation theory, whereby in type-II superconductors, the volume pinning force  $F_p$  depends on  $N_V f_p^2$  as well as on the FLL elasticity via an effective FLL-modulus  $\mu_{eff}$ .

I am not dwelling on the long disputes on the correct summation model. Only these remarks: The simplest model, applicable to dilute obstacle arrays, is the direct summation. The statistical theory of Labusch, on the other side, requires a threshold. Similar expressions to Labusch's can be found from a dynamical approach, originally suggested by Yamafuji & Irie in 1967. However, it turned out that although we can have sub-threshold pinning centers, a volume pinning force can be observed (particularly in systems with a near- $B_{c2}$  peak) as describes by the *collective pinning theory* of Larkin & Ovchinnikov (1979). For a large amount of weak pinning centers the long range order in the FLL lattice breaks down, whereby correlated regions of the FLL of volume  $V_c$  (which also could be determined by FLL defects) are created, in which a short range order is established. The net pinning force in a domain is proportional to  $\sqrt{N_V}$ .





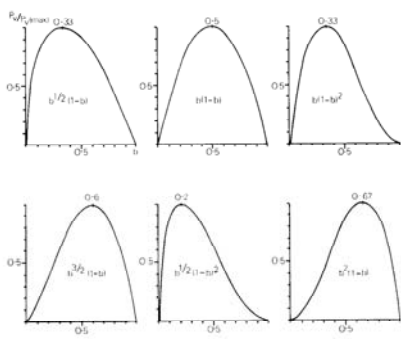
## Flux Pinning: Essential Issues ► Summation

Extension by E. J. Kramer (1972, 1978)

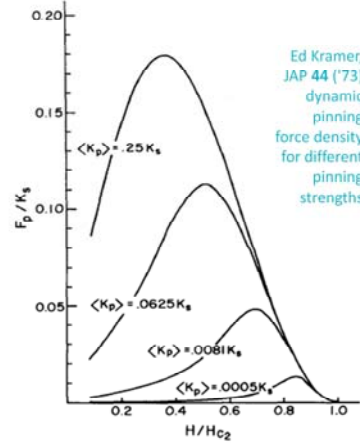
A master curve can be defined  
→ empirical relation

$$F_V = J_c \cdot B \propto B_{c2}^q b^n (1-b)^m$$

$b = B/B^*$ ;  $B^* = B_{c2}, B_{irr}$  ?



A.M.Campbell & J.E.Evetts, 1972, 'Flux vortices ...'



Ed Kramer, JAP 44 ('73)  
dynamic pinning force density for different pinning strengths

- weak pins;  $f_p$  small  
→ pin breaking,  $F_V \propto h^{1/2}(1-h)^2$   
small peak at high fields
- strong pins;  $f_p$  large  
→ FL shear,  $F_V \propto h^{1/2}(1-h)^2$   
high peak at low fields

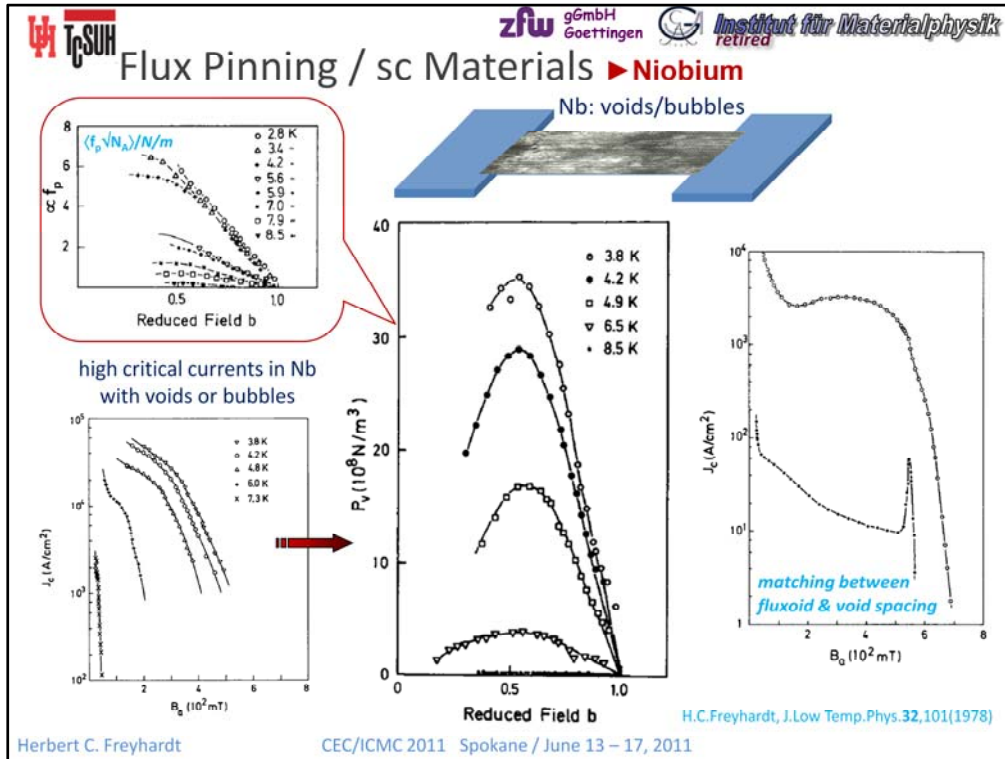
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To empirically describe the features/effects of the pinning obstacles and their arrangement, **master curves** have been defined. Particularly instructive has been Ed Kramer's discussion of the dynamic pinning force density (1973), which is characterized by different peak heights and peak fields, whereby at low FL densities *pin breaking* and at high FL densities *local shear of FLs* become the dominant mechanisms.

These master curves can be exploited to tailor  $J_c$ .

**Undoubtedly, basic pinning investigations helped to pave the way for applications of superconducting wires.**

- Basic flux pinning investigations helped to pave the way for applications of superconducting wires.




## Basic flux pinning investigations helped to pave the way for the application of superconducting wires.

Wherever we had a good cooperation, a close teaming between physicists, materials and solid state scientists, chemists and engineers, we not only learned to understand the importance of pinning for tailoring critical currents but, moreover, to fully optimize the behavior of superconducting wires. One also had to realize that by knowing the way to increase  $F_V$  or  $J_c$ , one could not go beyond certain physical limits. By selecting the following prominent examples, we will also demonstrate where we encounter these limits, e.g., through current blocking, the depairing limit, ...


Basic routes to increase  $F_V$ , and to make a conductor practical by designing the desired  $J_c$  will be illustrated below by considering several major classes of superconductors of interest for applications.

### Elemental superconductors: *Niobium*.


Nb single crystalline samples were used as model systems to study the basic flux pinning interactions (through stress fields, core pinning, ...) caused by dislocations, voids or bubbles (H. C. Freyhardt, 1978). Elementary interaction forces as well as the governing summation model could be studied. For regular void arrays the matching between fluxoid and void spacing generated a matching peak.



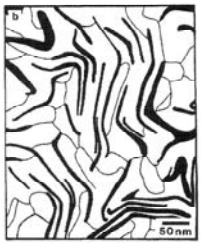
## Flux Pinning / sc Materials ► NbTi alloy superconductors



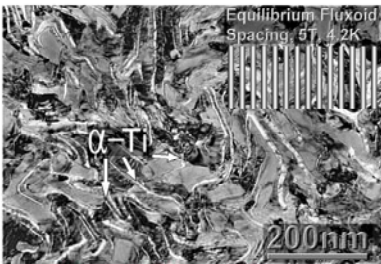
**a**



**c**



**b**



Equilibrium Fluxoid Spacing, 5T, 4.2K

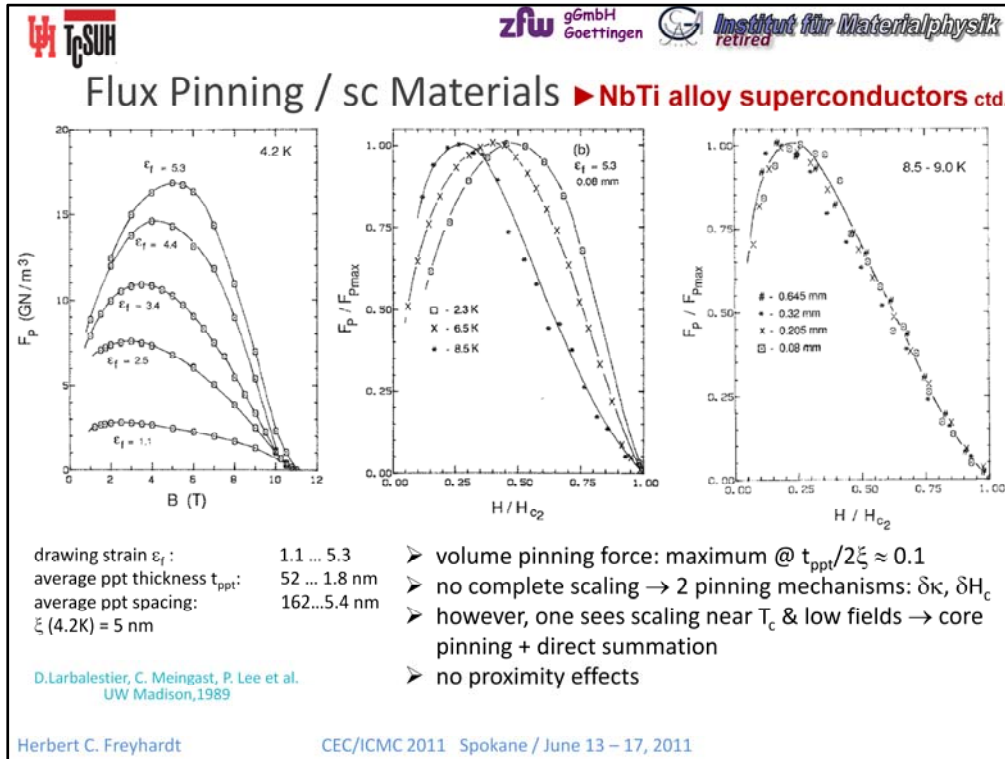
200nm

Nb 48 wt.% Ti  
 subsequent deformation and heat treatment steps:  
 → nc  $\alpha$ -Ti (15-20%), the pins in a  $\beta$ -Nb (42-39wt.%) Ti matrix  
 $T_c \approx 9.5$  K ( $\downarrow$  with ppt size/spacing)  
 $H_{c2} = 10.8$  T,  $H_{c2}^{res} = 11.9$  T  
 $H^* = 10.2$  T (@4.2 K)

D. Larbalestier, C. Meingast, P. Lee et al. UW Madison, 1989

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**Alloy superconductors:** In 1960, NbZr turned out to be one of the first candidates of alloy conductors for high current applications, and this was achieved by exploiting reactions in materials science to generate the desired pinning landscape. Typically alloys with 25 – 50 at.% Zr were used, where at 785°C one sees a eutectic decomposition into  $\beta$ Nb +  $\beta$ Zr, and at 560°C a eutectoid decomposition into  $\beta$ Nb +  $\alpha$ Zr. The  $T_c$  is around 10.8 K and  $B_{c2}(4.2K) = 10.5$  T. However, NbZr was quickly replaced by NbTi, starting at about 1965. NbTi, with an optimized Ti content of 48 wt.% is highly attractive because it's ductile, mechanically strong and straightforward to process. It can be readily used up to 9 T @ 4.2K and 11 T @ 1.7 K. Particularly two groups need to be mentioned here, David Larbalestier et al. in Madison (at that time) and Hans Hillmann at the Vacuumschmelze/Hanau. The NbTi  $T_c$  of about 9.5 K is slightly smaller than for NbZr, but NbTi exhibits an attractive  $B_{c2} = 10.8$  T. The size, shape and distribution of the  $\alpha$ -Ti precipitates can be tailored at wish by a combination of successive deformation and heat treatment steps to generate optimum pinning. By inspecting the actual precipitate distribution, one can immediately visualize the influence of current blocking for critical transport currents.



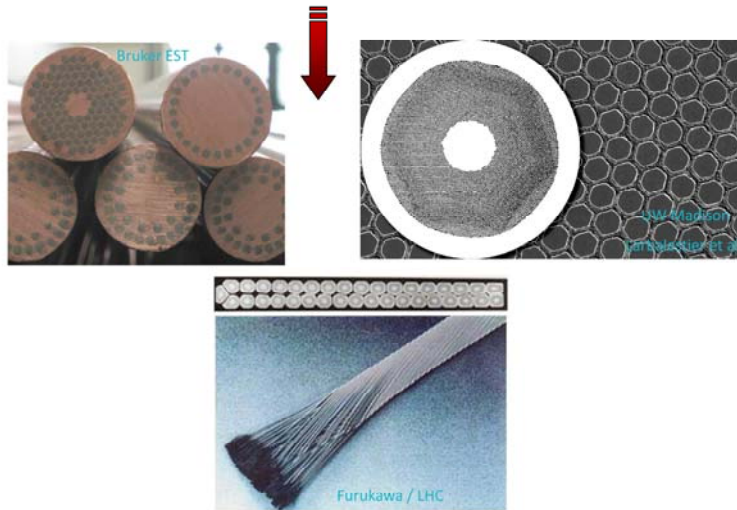
Details of the pinning in Nb 48 wt.% Ti through  $\alpha$ -Ti precipitates or clusters of precipitates were particularly studied by Larbalestier, Meingast and Lee, who investigated the characteristic features of the scaling of the volume pinning force  $F_p$ , whereby they succeeded in identifying the relevant obstacles or obstacle clusters as well as the origin of the elementary pinning mechanisms.



## Flux Pinning / sc Materials ▶ NbTi alloy superconductor ctd.

FURTHER OPTIMIZATION:

- ❖ Doping, e.g. by Fe, ↑ pinning efficiency
- ❖ APC: artificial pinning centers
- ❖ Ta alloying: increase  $B_{c2}$




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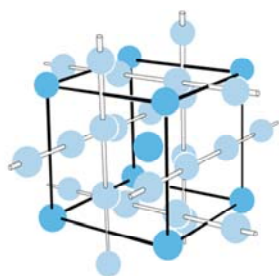
Based on this knowledge, technical conductors could be manufactured for, e.g., Magnetic Resonance Imaging or High Energy Physics, as depicted in the slide:

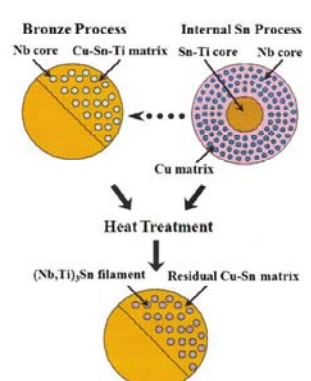
Examples:

- NbTi wires of *Bruker EST*,
- NbTi conductor (*UW Madison*),
- *Furukawa*: LHC conductor and Rutherford cable with 36 strands; wire: 8500 single cores, 6  $\mu\text{m}$  NbTi filaments. Non-Cu  $J_c$  of strand: 2300 A/mm<sup>2</sup> @ 4.2 K, 6 T.



## Flux Pinning / sc Materials ▶ A-15 sc: Nb<sub>3</sub>Sn, V<sub>3</sub>Ga, Nb<sub>3</sub>Al,...





Compound	T <sub>c</sub> /K	B <sub>c2</sub> (0)/T
Nb <sub>3</sub> Sn	18	23
V <sub>3</sub> Ga	15.9	21
Nb <sub>3</sub> Al	18.8	32

1961 J.E. Kunzler / Nb<sub>3</sub>Sn PIT: powder-in-tube  
 1970 ff. bronze process  
 1990 ff. internal Sn process

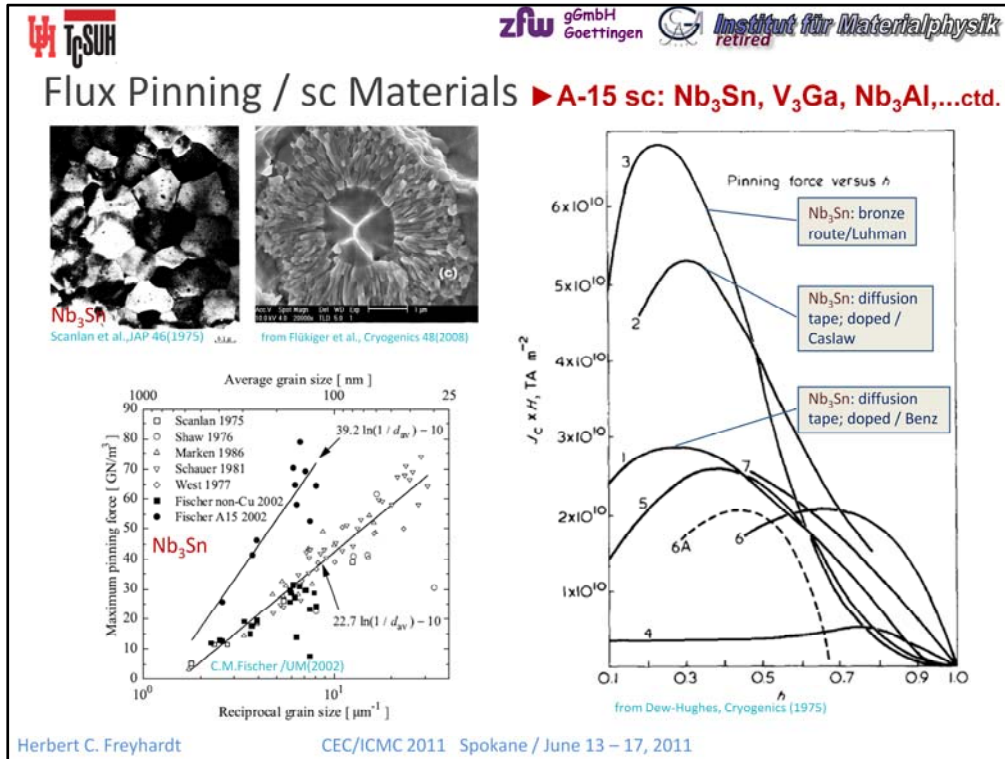
*alternative routes:*

- powder metallurgical process
- jelly roll process
- via metastable compounds

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**Compound superconductors. A-15 conductors, e.g., Nb<sub>3</sub>Sn, V<sub>3</sub>Ga, Nb<sub>3</sub>Al.**

A-15 structures are built from three orthogonal transition metal chains, with  $T_c$  values up to 23.2K (Nb<sub>3</sub>Ge). From the above mentioned A-15's, which are attractive for application, Nb<sub>3</sub>Sn was the first one investigated by J.E. Kunzler in 1961. Nb<sub>3</sub>Sn is required to produce fields above 10 T @ 4.2 K as used for NMR, fusion, etc. In the early 1970's, the bronze process was developed. Different methods were realized to embed the transition metal in a bronze matrix. After the deformation (wire drawing, ...) the A-15 phase is formed during a heat treatment. Upper critical fields and  $T_c$ 's depend on stoichiometry and additives like Ti, Ta. Because the solubility limit of Sn in the bronze is 15.8 wt.%, and because in general the Sn content of the matrix was varied between 13 - 16 wt.%, an increase in the Sn fraction, which subsequently would result in a larger A-15 fraction, was made possible by using the internal tin (IT) process. In such a way, the non-Cu  $J_c$  of the A-15 wires could be noticeably increased. Our goal is not to discuss the obstacles, which needed to be overcome to develop viable processing technologies, but rather to elucidate the important strategies to implement flux pinning.



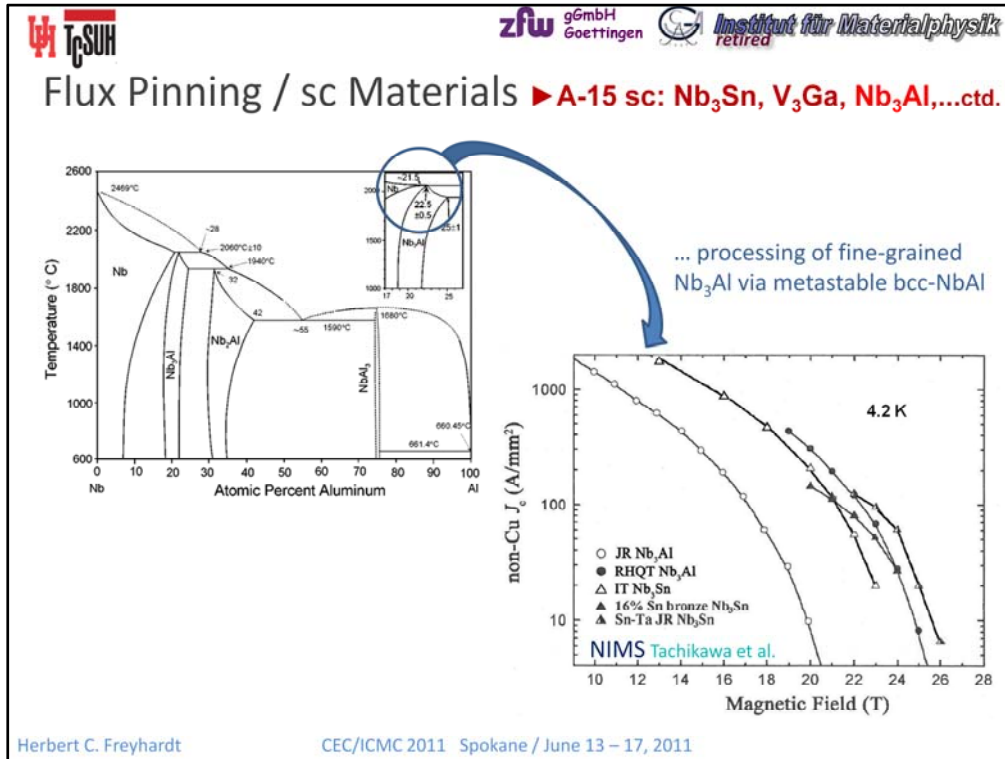
For the major A-15 conductors, grain boundaries clearly represent the major pinning centers, as pointed out and visualized by TEM imaging by Scanlan et al. (different grain sizes for Nb<sub>3</sub>Sn, with ZrO<sub>2</sub> particles). Size and shape (equiaxed, elongated) of the grains vary within the reaction layer (see, e.g., Flükiger et al.).

Pinning, particularly the volume pinning force is inversely proportional to the grain size. Similar dependencies can be found for V<sub>3</sub>Ga (Tachikawa et al.).


Inspecting, e.g., in Nb<sub>3</sub>Sn, the volume pinning force versus the reduce field, one finds that for large enough grains a scaling is observed/obeyed, which follows Kramer's empirical pinning behavior (see above): *pin breaking* at low fields or large FL spacings and *FL shearing* (FL flow or induced by FL dislocations) at larger fields. In general, maximum volume pinning forces are observed around 0.2B\*, with a high peak at low fields, resulting from strong pins, which induce flux shearing .

Furthermore, one finds well behaved Kramer plots ( $J_c^{1/2} B^{1/4}$  versus  $B$ ).

Similar progress has been achieved for V<sub>3</sub>Ga, to which particularly Tachikawa and his coworkers contributed.




More versatile methods have to be employed to process Nb<sub>3</sub>Al conductors with attractive properties. From the phase diagram we learn that an A-15 phase with a composition near to stoichiometry can only be obtained by solid state reactions at high temperatures, which would result in an unwanted coarse grain structure. Good stoichiometry and fine-grained Nb<sub>3</sub>Al, however, can be achieved through an alternative processing of A-15 Nb<sub>3</sub>Al via a metastable bcc-NbAl compound. The approach at the Uni. Göttingen (H. C. Freyhardt et al.) realized this in the early nineties via a rapid quenching of laser-molten Nb-Al powder mixtures (plus a subsequent appropriate heat treatment), and Tachikawa and coworkers by an elaborate technique of quenching the melt in liquid Ga (RHQT: Rapid Heating, Quenching and Transformation technology).




## Flux Pinning / sc Materials ▶ A-15 sc: Nb<sub>3</sub>Sn, V<sub>3</sub>Ga, Nb<sub>3</sub>Al, ...ctd.

**Nb<sub>3</sub>Sn**




Round Nb<sub>3</sub>Sn wires, up to Ø 1.4 mm & 18000 filaments

**Bruker EST**

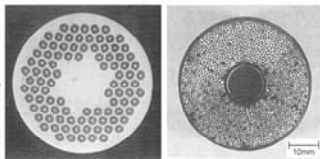


ITER CiC conductor  
1152 Nb<sub>3</sub>Sn strands

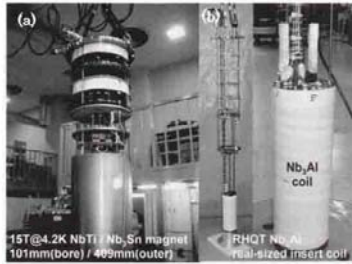


2009

**Nb<sub>3</sub>Al**



NIMS (NRIM) Tsukuba  
Tachikawa et al.



(a) 15T@4.2K NbTi / Nb<sub>3</sub>Sn magnet  
101mm(dore) / 499mm(outer)

(b) RHQT Nb<sub>3</sub>Al real-sized insert coil

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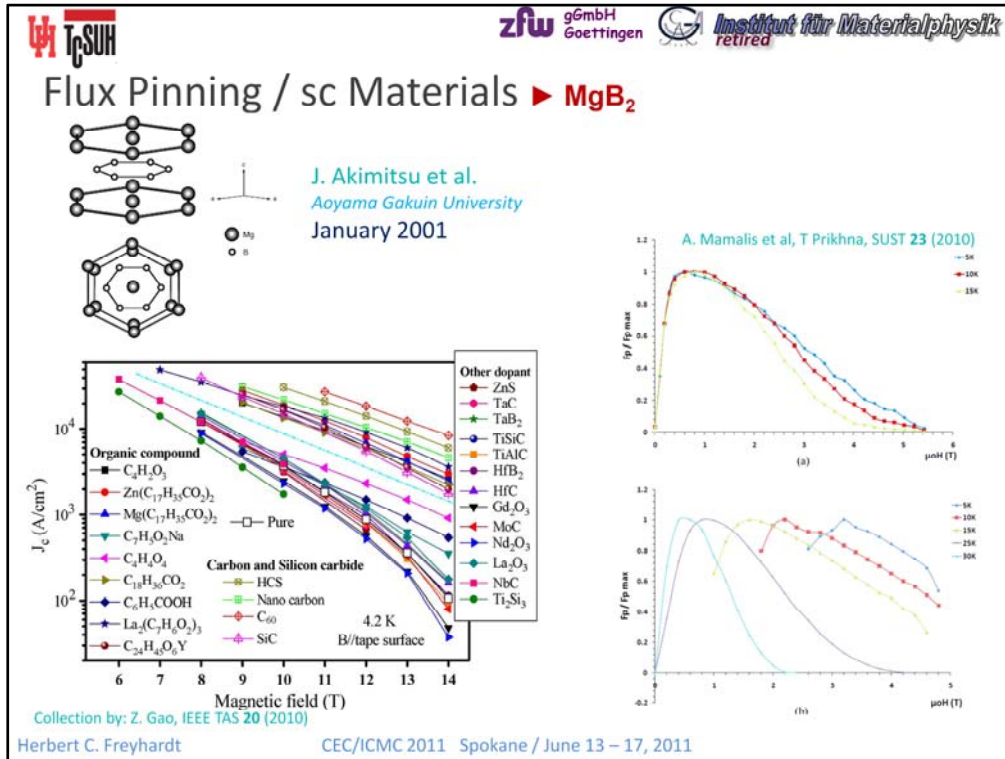
The remarkable progress in manufacturing technologies lead to the development of a number of reliable A-15 conductors, which can be produced in km lengths. A few examples are depicted:

- Internally stabilized round Nb<sub>3</sub>Sn with up to 18000 filaments (Bruker EST),
- ITER CEC (cable-in-conduit) cable with 1152 Nb<sub>3</sub>Sn strands and a non-Cu  $J_c$  of 600 A/mm<sup>2</sup> @ 12 T , 4.2 K (Bruker EST).

These high-quality A-15 conductors enabled Bruker BioSpin to built, with the AVANCE 1000, the world's first 1 GHz spectrometer with a 23.5 T persistent current superconducting magnet.


The Jelly-Roll-processed Nb<sub>3</sub>Al CIC conductor (employing RHQT) allowed the construction of a magnet, where the Nb<sub>3</sub>Al insert generated a field of 19.7 T @ 4.2 K in a 15 T back-up field of NbTi/Nb<sub>3</sub>Sn magnets (see K. Tachikawa, ESNF, 2011, issue 16, paper RN20).





**Pnictides** will not be discussed here, where one just starts to investigate and tries to tailor the irreversible properties, including  $J_c$ .

Only a few remarks about **MgB<sub>2</sub>**, discovered - as an unexpected 'surprise discovery' - in 2001 by J. Akimitsu in Japan. The  $T_c$  is 39 K, and one tried to increase it with all different kinds of additives ... without success. At that time, we were in the era of HTS ... and one was surprised to find that grain boundaries in MgB<sub>2</sub> did not act as weak links, they are rather pinning the FLs. However, because of the complex microstructure obtained by the different processing routes, a clear statement as to the origin of pinning is still not possible. Carbon doping, by various methods (organic compounds, like C<sub>4</sub>H<sub>6</sub>O<sub>5</sub> (malic acid; e.g., Flükiger et al.) SiC, C<sub>60</sub>, CNTs, ...), very often leads to a replacement of B by C and increases  $B_{irr}$ . Grain boundary pinning can play a role as well as  $\delta l$  and  $\delta T_c$  pinning (collective pinning!?)




Flux Pinning / sc Materials ▶ **Coated Conductors REBCO**

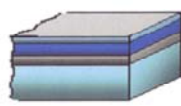
... a race

HTS must be available as wires, tapes, assembled conductors

HTS conductors  
 1st generation  
 Bi-2223, Bi-2212



HTS conductors  
 2nd generation  
 YBaCuO coated conductors



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The discovery of **high-temperature superconductors (HTS)** in 1986 by Bednorz and Müller opened a new chapter in R&D of superconducting materials. One now could envision to apply superconducting wires and cables at temperatures of the much more readily available liquid nitrogen. Because of the more favorable spectrum of properties, one sees nowadays a preference for the use of REBaCuO coated conductors, rather than of the HTS conductors of the first generation, i.e., Bi-2223 or Bi-2212 HTS wires.

**A possible solution**

zfw g6mbH Goettingen Institut für Materialphysik retired

**COATED CONDUCTORS: TWO ALTERNATIVES**

**IBAD-YBCO CONDUCTORS**

Protective Coating (Ag, Au, ...)  
 optional "cap layer":  $\text{CeO}_2, \text{Y}_2\text{O}_3$   
 Biaxially aligned YBCO film (PLD, Thermal Coevaporation, ...)  
 Polycrystalline Substrate Tape or Sheet  
 Ion-Beam-Assisted Deposition: IBAD  
 Inclined-Substrate Deposition: ISD  
 Biaxially aligned buffer layer:  $\text{YSZ}, \text{CeO}_2, \text{CGO}, \text{MgO}, \dots$

**RABITS-YBCO CONDUCTORS**

YBCO  
 Buffer-layer Architecture ...  $\text{CeO}_2, \text{RE}_2\text{O}_3, \dots$   
 $\text{YSZ}, \text{RE}_2\text{O}_3, \text{CeO}_2$   
 Rolling-Assisted-Biaxially-Textured Substrates  
 Ni, NiCr, NiV, ...

IS ▶ **Coated Conductors REBCO**

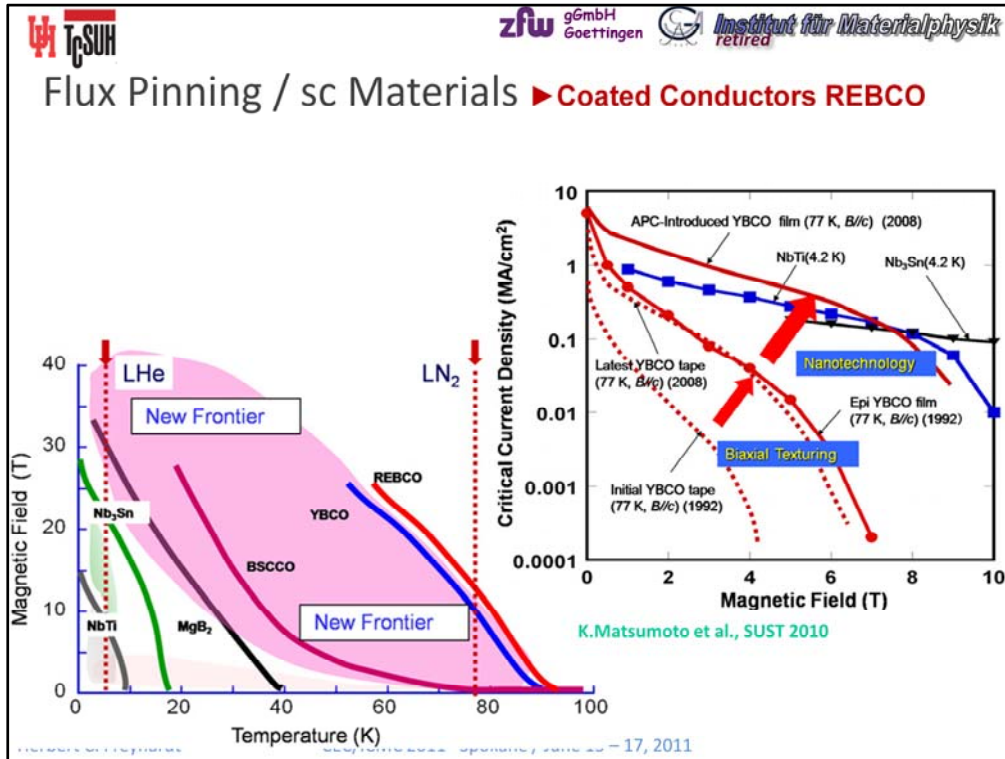
... a race

es, tapes, assembled conductors

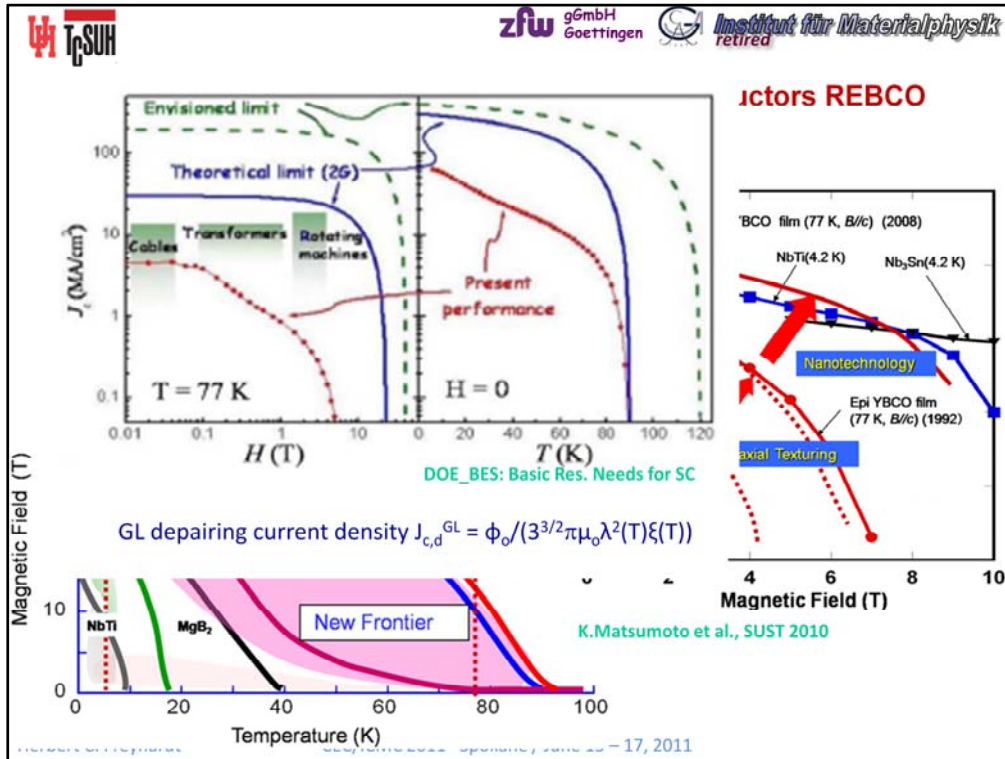
HTS conductors  
 2nd generation  
 YBaCuO coated conductors

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For the 2<sup>nd</sup> generation of HTS conductors, the **Coated Conductors (CC)** from **REBaCuO**, **mainly YBCO**, two basically different processing routes, IBAD (Ion-Beam Assisted Deposition) and RABITS (Rolling Assisted Bi-axially Textured Substrates) were developed, by utilizing both physical and chemical deposition techniques. Coated conductors possess a complex structure, consisting of a (mechanically strong) substrate tape covered with buffer, HTS and cap layers, and a protective coating. To bring it to application, a multitude of mechanical, electromechanical and electromagnetic specifications will have to be met. Nonetheless, here too, the current carrying capability in the superconducting state remains an essential hallmark.



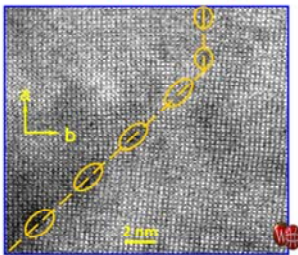
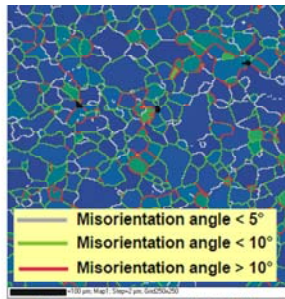
In the diagram of accessible temperatures and magnetic fields, with the introduction of REBaCuO CC, new frontiers opened up, enabling the generation of fields beyond 40 T and 10 T at 4.2 K and 77 K, respectively. However, one had to overcome two challenges. These REBCO-HTS are strongly anisotropic and their grain boundaries act as weak links. To increase the critical current densities to the expected levels, at first the biaxial texturing of the HTS layer had to be implemented before one could improve  $J_c$  by creating the desired pinning landscape.



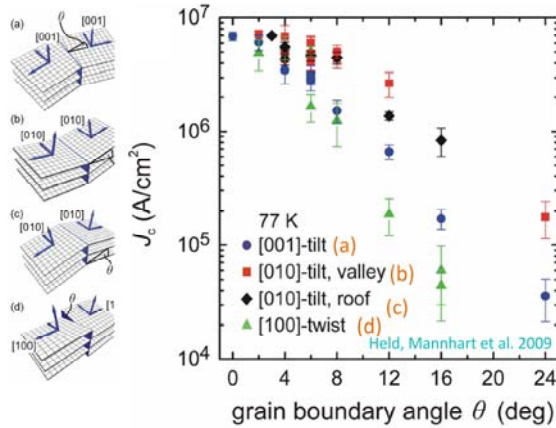
The imaginative and innovative implementation of these prerequisites brought us much nearer to the theoretical limit, the pair breaking limit in a superconductor.



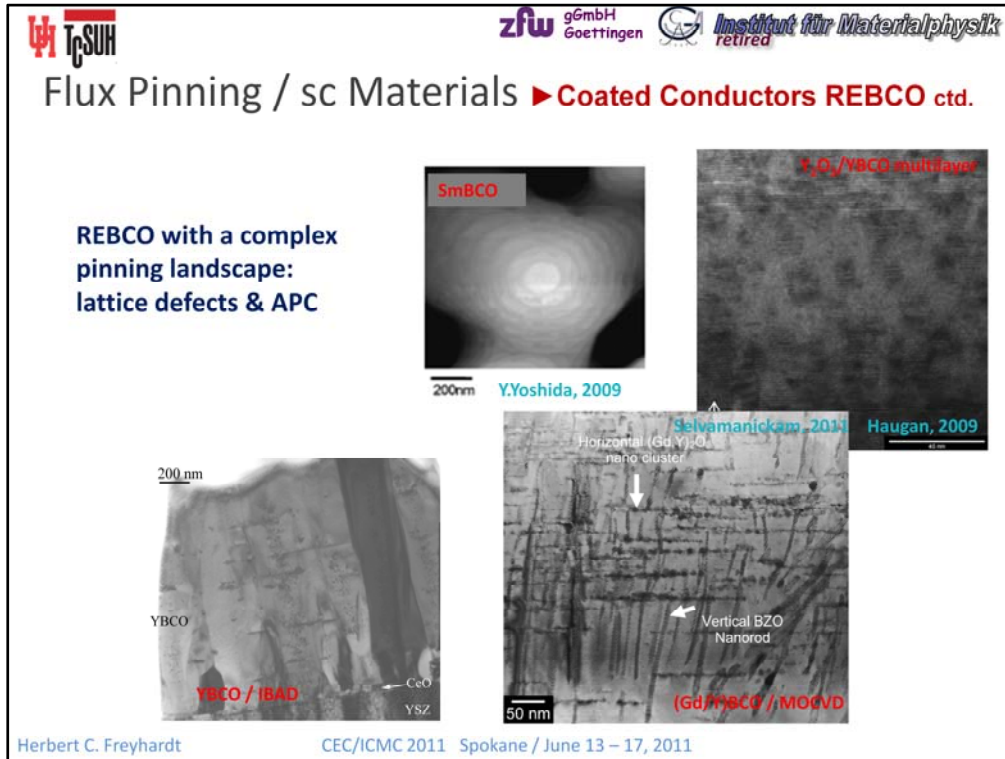
## Flux Pinning / sc Materials ▶ Coated Conductors REBCO ctd.



Grain boundaries act as weak links & determine current transfer

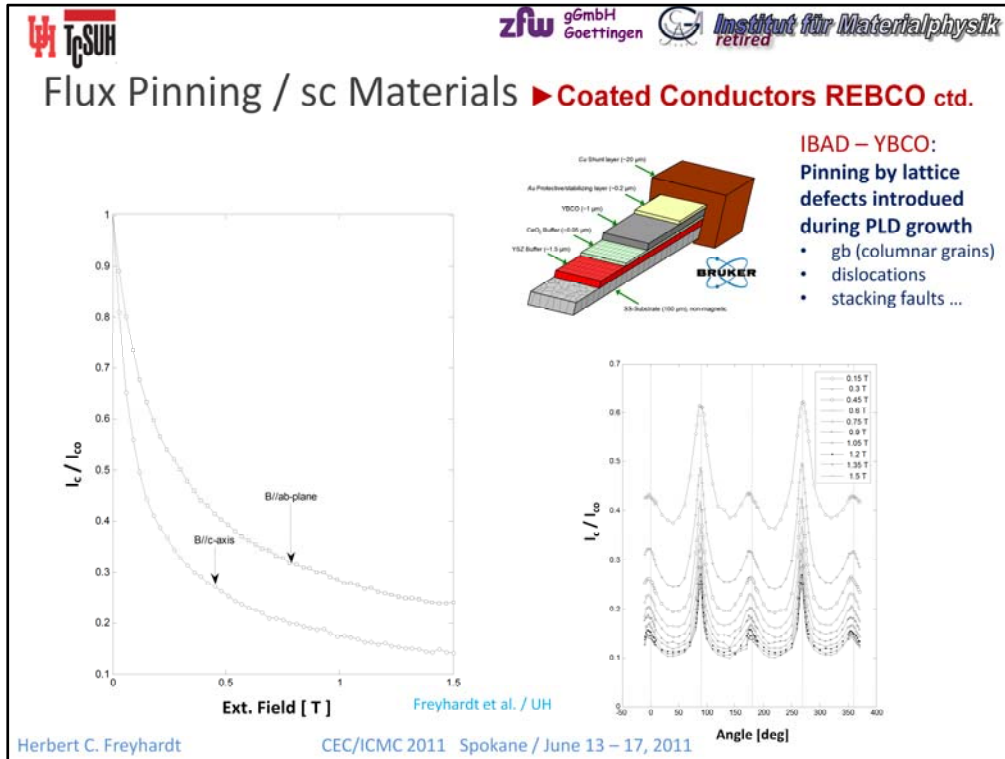


Dimos, Chaudari and Mannhart in 1990 were the first to point out that **grain boundaries in REBCO act as weak links**. The critical transport currents crossing the boundary decrease in a first approximation exponentially with increasing misorientation angles between adjacent grains. Further inspection of the microstructure of different configurations of grain boundaries (which can even mäander) as weak link junctions revealed though a more detailed insight, and also allowed to develop a sophisticated view of the interaction of FLs, pancake or Josephson vortices with the grain boundary. It was already stated, that it's a must to biaxially texture the HTS layer in CCs. Here EBSD, exemplified by a measurement of the IFW/Dresden, turned out to become one of the most valuable experimental tools.



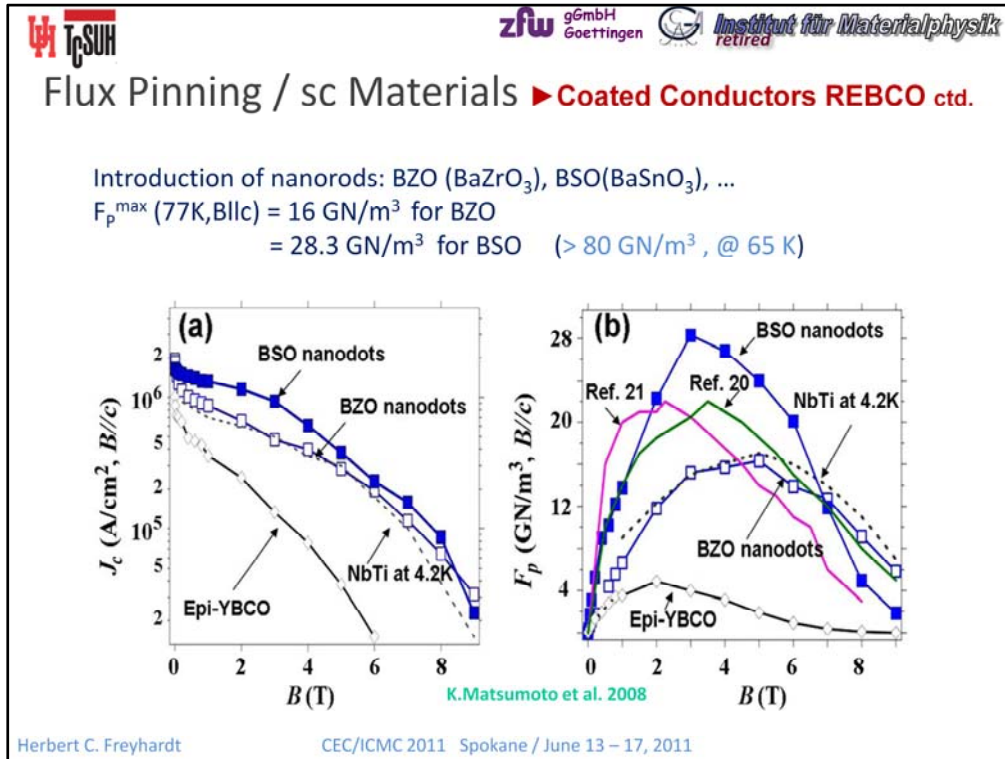
REBCO layers grown by physical or chemical deposition method reveal a complex pinning landscape, which can be characterized either as ‘natural’ (growth steps, lattice defects, 122 inclusions, intergrowth, phase boundaries ...) or ‘artificial’ (e.g., Artificial Pinning Centers, nanoparticles).

- Growth spiral induced by a screw dislocation in SmBCO,
- $Y_2O_3$  nanoparticles in  $Y_2O_3$ /YBCO multilayers,
- Columnar grain structure in laser-deposited YBCO (H. C. Freyhardt et al.) with defects (dislocations, stacking faults, ...),
- Gd/YBCO MOCVD films with vertical  $BaZrO_3$  nanorods and  $(Gd,Y)_2O_3$  nanoclusters



An example of how the columnar grain structure in laser-deposited YBCO (of the previous slide) with its characteristic defects (dislocations, stacking faults, ...) determines the field and angular dependent critical currents is depicted for technical IBAD-YBCO coated conductors.

It's particularly noteworthy that the planar defects introduced in the ab crystal planes of the YBCO and the intrinsic pinning, caused by the layered structure of the YBCO, lead to a pronounced anisotropy of the critical currents vs. angle, with a marked peak for external fields parallel to the ab planes.



The introduction of artificial pinning centers developed into one of the most innovative methods to not only increase the critical current density but, moreover, to tailor its angular dependence.

Critical currents and volume pinning forces of CC can be considerably raised through introducing BZO ( $\text{BaZrO}_3$ ) and BSO ( $\text{BaSnO}_3$ ) nanorods. Although the details of the impact of specific obstacles is not fully clarified, it is important to note that one finds  $J_c \propto B^{-\alpha}$ , with  $\alpha = 0.5$  in CC with 5-10nm BZO, which points to a FL shearing mechanism (s. above, Kramer), while for low fields  $\alpha: 0.5 \rightarrow 0.2$ , where pin breaking might be dominant.

The observed temperature dependence of the volume pinning force  $F_p(B)$  still needs to be clarified.

However, we have a positive accompanying effect in that with a stronger pinning, the irreversibility field  $B_{\text{irr}}$  is also increased.

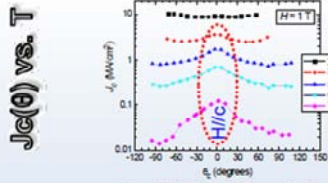
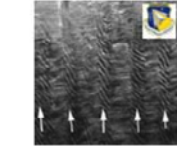
Furthermore, one learns that for a large volume fraction of nanoparticles, current blocking will play a role.

# Flux Pinning / sc Materials ▶ Coated Conductors REBCO ctd.

Combination of point & line pinning centers → tailoring of angular dependence of  $J_c$

**Case I:**  
Columnar defects

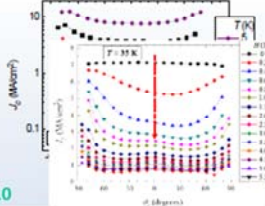
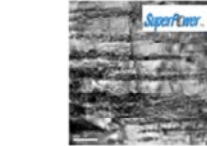
YBCO+BaSnO  
on LAO  
2.5  $\mu\text{m}$   
 $T_c = 85.5\text{K}$



Dominic Lee / Peer Review 2010

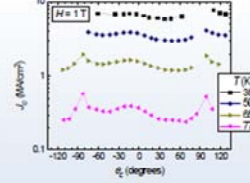
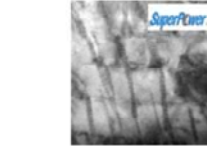
**Case II:**  
Precipitates

(Y,Gd)BCO on IBAD  
2.8  $\mu\text{m}$   
 $T_c = 90.7\text{K}$   
 $I_c = 760\text{ A/cm } 77\text{K sf}$




**Case III:**  
CD + ppt

(Y,Gd)BCO+BZO  
3.3  $\mu\text{m}$   
 $T_c = 90.5\text{K}$   
 $I_c = 944\text{ A/cm } 77\text{K sf}$




By artificially creating a combination of line and point pinning centers, the angular dependence of the critical current of coated conductor tapes can be tailored.






## ... impact on today's grid

**Efficiency  
Lost Energy**





62% energy lost in production / delivery  
8-10% lost in grid  
40 GW lost (US)  
~ 40 power plants  
2030: 60 GW lost  
340 Mtons CO<sub>2</sub>



Long Island / LIPA + AMSC


**Trafo (2 MVA)**  
Kyushu Electric,  
Fujikura, Showa






ISFCL / Bruker

sc wind generator




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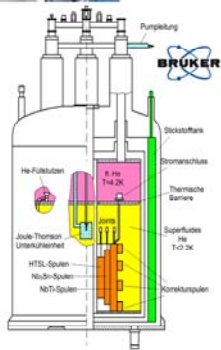
In the future one can foresee a sustainable application of coated conductors for energy saving devices and devices with new functionalities in the power grid. Novel ways of generating (high) magnetic fields will become feasible.



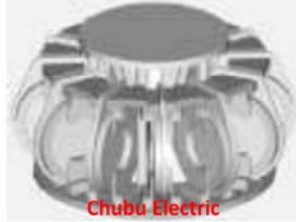
## Magnet Applications



**Avance 1000**




Pumpleitung  
 BRUKER  
 Hei-Füllboden  
 Jule-Thomson-Umkehrmagnet  
 HTSL-Spulen  
 Nb<sub>3</sub>Sn-Spulen  
 NbTi-Spulen  
 4 He 1.9 K  
 Chromschicht  
 Thermische Barriere  
 Superflüssiges He  
 T=2.2K  
 Stickstoffflüssig  
 Kommuterspulen

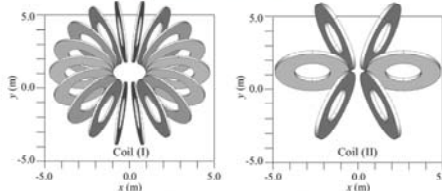


**Chubu Electric**

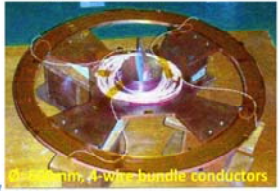
2 GJ class SMES



**SuperPower/NHFL**  
 Insert Coil / YBCO CC  
 ID/OD 25/36 mm  
 2.8 T in 31 T background



Coil (I)      Coil (II)



Superconducting coil with W bundle conductors

Herbert      ne / June 13 - 17, 2011

... and:  
 Novel ways of generating (high) magnetic fields will become feasible.



There is hope, that we find a way out of the R&D-of-HTS *Labyrinth*, and see a *sunrise* for the application of HTS.