# What to look for in new superconductors if we want to make them useful?

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Applied Superconductivity Conference, Salt Lake City, Sept. 6, 2024

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### **Useful for: power/magnet applications?**

**Research magnets** 



Medical MRI



HTS motors & generators



Power transmission lines



Figure 1 Schematic comparison of the  $3\times3$  duct bank of an underground copper distribution system vs. a single triaxial HTS cable operating at 13 kV and transferring 69 MVA of power.

MagLev





### **Useful for: high-Q SRF resonators?**



- Football-size Nb or Cu cavities 2-3 mm thick
- $TM_{010}$  resonance mode with f = 1-2 GHz
- Cooled by superfluid He at 2 K



### **Define useful superconductors**

#### What application?

- Materials parameters of merit depend on operating temperature, magnetic field and frequency
  - high field dc magnets, SMES: high J<sub>c</sub> and irreversibility field
  - motors and generators: high J<sub>c</sub>, low ac losses, twisted multifilamentary wires
  - transmission lines, FCL: high  $J_c$  at low fields
- Requirements for electronic, QIT and high-Q resonators are best satisfied by conventional superconductors
  - High Q resonators: no vortices, s-pairing, large SC gap, lowest density of subgap states
  - Engineering of optimum density of states to minimize very weak RF dissipation in the vortex-free state As a review, see Gurevich, SUST, 36, 063002 (2023)
- Lots of SC materials (conventional, cuprates, pnictides) are available for applications at 4.2K
- Very few SC can perform at 77 K, and  $T_c$  is not the only reason
- The best performance does not always require the highest  $T_{\!c}\,\text{and}\,\,H_{c2}$
- To operate at T>77K, tough materials requirements are to be satisfied
- Can they help in searching for new superconductors?

### **Superconducting materials**

Is T<sub>c</sub> the most important parameter of merits for applications?

Should the search for new superconductors be primarily focused on T<sub>c</sub>?

Does high T<sub>c</sub> ensure applications at high temperatures and fields?



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### Search for new superconductors

Matthias rules of maximizing T<sub>c</sub>: BCS physics + materials experience + legendary insights:

- 1. High symmetry is good; cubic symmetry is the best
- 2. High density of electronic states is good
- 3. Stay away from oxygen
- 4. Stay away from magnetism
- 5. Stay away from insulators

#### Works for LTS but fails for unconventional SC (HTS cuprates and FBS)

- The higher the operating temperature, the less relevant T<sub>c</sub> and H<sub>c2</sub> become.
- Performance at high fields and temperature is mostly limited by thermal fluctuations of vortices and current-blocking grain boundaries
- The higher the electron density and more isotropic the better (1,2)
- Avoid competing orders: superconductivity with antiferromagnetic or structural transitions (4,5)

**Dream and challenges of RTS**: If a superconductor with  $T_c > 300K$  and  $B_{c2}=1000$  T has been discovered, can it be used at 300K? What would be the materials requirements to enable RT magnet applications? M.R. Beasley, MRS Bull, 36, 597 (2011); Gurevich, Nature Mater. 10, 255 (2011); Annu. Rev. Cond. Mat. Phys. 5, 35 (2014); A.P. Malozemoff, MRS Bull. 36, 601 (2011), Physica C (2013)

![](_page_5_Picture_15.jpeg)

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### Is there a T<sub>c</sub> limit for conventional superconductors?

McMillan formula for intermediate coupling

$$T_c = \frac{T_D}{1.45} \exp\left[-\frac{1.04(1+\lambda_p)}{\lambda_p - (1+0.62\lambda_p)\mu^*}\right] \lesssim 0.2T_D \qquad \lambda_p \lesssim 1.5$$

 $T_c\simeq 0.2T_D\sqrt{\lambda_p}, \qquad \lambda_p\gtrsim 2$  Allen and Dynes, Phys. Rev. B 12, 905 (1975)

High Debye temperature  $T_D \propto M^{-1/2}$  for light atoms line H, strong electronphonon coupling  $\lambda_p = \langle VN(E_F) \rangle$  and peaks in DOS at the Fermi surface

T<sub>c</sub> enhancement by pressure:

**Ti:**  $T_c$  from 0.5 K to 25 K at 248 GPa (2.5M atm) Zhang et al, Nature Commun. 13, 5411 (2022)

**NbTi**:  $T_c$  from 9.6 K to 19.1 K,  $B_{c2}$  from 15.4T to 19.1T at 262 GPa , Guo et al, Adv. Mater, 1807240 (2019)

#### Superhydrides: $LaH_{10} T_c = 250 \text{ K} (-23C) @ 170 \text{ GPa} (1.7\text{ M atm})$ Drozdov et al Nature 569, 528 (2019)

![](_page_6_Figure_9.jpeg)

Tune the Fermi energy to match the DOS peaks by chemical or applied pressure

### In-field performance is controlled by vortices

![](_page_7_Figure_1.jpeg)

#### High-field superconductors:

- HTS cuprates, B<sub>c2</sub>(0) > 100-200 T
- FBS.  $B_{c2}(0) > 50-100 \text{ T}$
- PbMo<sub>6</sub>S<sub>8</sub>, B<sub>c2</sub>(0) = 50 T
   Seeber, Phys. Plasmas. 30, 120604 (2023)

- Quantized vortices in which supercurrents circulate over  $\lambda$  = 40-400 nm around normal cores of radius  $\xi$  = 2-40 nm.
- Short coherence length ξ ~ ħv<sub>F</sub>/2πk<sub>B</sub>T<sub>c</sub> = 2-4 nm yields high B<sub>c2</sub>, which can be further increased by alloying with nonmagnetic impurities

clean limit:  $B_{c2} = \phi_0/2\pi\xi^2 \propto mT_c^2/E_F$ 

dirty limit:  $B_{c2}=\phi_0/2\pi\xi l\propto T_c
ho_n$ 

• Higher  $T_c$  does result in high  $B_{c2} = 20 - 100$  T to be used in superconducting magnets

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### Ten-fold increase of B<sub>c2</sub> by materials disorder in MgB<sub>2</sub>

![](_page_8_Figure_1.jpeg)

From 3 to 10 - fold increase of  $B_{\rm c2}$  as compared to  $MgB_2$  single crystals. Anomalous increase of  $B_{c2}$  by two-band SC. Selective tuning of impurity scattering in  $\pi$  and  $\sigma$  bands in MgB<sub>2</sub>

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#### Gurevich, PRB 67, 184515 (2003)

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### Critical current density J<sub>c</sub>(B) – one of main parameter for applications

Type –II superconductors can carry nondissipative currents if vortices are pinned by materials defects

![](_page_9_Figure_2.jpeg)

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IEEE-CSC, ESAS and CSSJ SUPERCONDUCTIVITY NEWS FORUM (global edition), Issue No. 57, Oct 2024. Plenary presentation given at ASC 2024, Sept 2024, Salt Lake City, Utah, USA.

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### High-J<sub>c</sub> at low T: the more pinning centers the better

![](_page_10_Picture_1.jpeg)

 $\alpha$ -Ti ribbons in a Nb-Ti alloy (Larbalestier & Lee)

Can produce self-field  $J_{\rm c}$  up to 10-30% of the depairing current density:

$$J_d \approx \frac{H_c}{\lambda} \approx \frac{\phi_0}{3\sqrt{3}\pi\mu_0\lambda^2\xi}$$

![](_page_10_Picture_5.jpeg)

- Vortices are chopped into short, strongly pinned segments
- From weakly pinned vortex spaghetti to strongly pinned vortex pasta.
- Can produce very high  $J_c \sim (0.1-0.3)J_d$ .

Haugan, et al. Nature 430, 867 (2004)

8 nm YBa<sub>2</sub>CuO<sub>5</sub> nanoparticles

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### **Conventional materials optimization for low-T applications**

- Materials with higher  $T_c$  tend to have higher  $B_{c2}$  and  $J_c$  and generate stronger magnetic fields
- $B_{c2}$  can be further increased by alloying with nonmagnetic impurities
- Incorporate defect structures or APC to pin vortices. The more pinning defects the better
- Produce twisted multifilamentary round wires to suppress thermo-magnetic instabilities and reduce ac losses

![](_page_11_Picture_5.jpeg)

**Bi-2212 round wire** Larbalestier et al, Nature Mater Jiang et al IEEE TAS 29, 6400405 (2019)

NbTi

All work fine at 4.2 K

![](_page_11_Picture_8.jpeg)

Sr<sub>06</sub>K<sub>04</sub>Fe<sub>2</sub>As<sub>2</sub> Yao et al, APL 102, 082602 (2013)

 $MgB_2$  in Cu

![](_page_11_Picture_11.jpeg)

![](_page_11_Picture_12.jpeg)

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![](_page_11_Picture_14.jpeg)

### Why cannot it work at 77 K?

![](_page_12_Figure_1.jpeg)

- Irreversibility field H\*(T) above which J<sub>c</sub>(T,H) = 0
- H\* is limited by thermal fluctuations of vortices and strong magnetic flux creep

![](_page_12_Figure_4.jpeg)

- It is neither T<sub>c</sub> nor H<sub>c2</sub>, but high J<sub>c</sub> and H\*(T), which make superconductors useful
- Above 77 K, very high H<sub>c2</sub> of cuprates becomes irrelevant and H\* << H<sub>c2</sub>
- Mechanisms responsible for high T<sub>c</sub> can cause problems with magnet applications at 77K

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### **Conventional vs unconventional pairing**

Conventional: the wave function of the Cooper pair  $\Psi$  has the same symmetry as the Fermi surface: s-wave BCS superconductivity mediated by phonons. Weak sensitivity to nonmagnetic impurities. Majority of high-field LTS

Unconventional: d-wave or multiband superconductivity mediated by magnetic excitations. Stronger sensitivity of T<sub>c</sub> to impurities, the grain boundary problem. HTS cuprates, heavy fermions, iron pnictides.

![](_page_13_Figure_3.jpeg)

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### **Competing orders. What helps T<sub>c</sub> can harm J<sub>c</sub>**

![](_page_14_Figure_1.jpeg)

![](_page_14_Picture_2.jpeg)

Left: The nanoscale superconducting energy gap disorder in  $Bi_2Sr_2CaCu_2O_x$ . Right: A simultaneous image of the dopant atom locations from SI-STM.

![](_page_14_Figure_4.jpeg)

Enhancement of SC by proximity to an AF phase

#### BUT

- As ξ ~ ħv<sub>F</sub>/2πk<sub>B</sub>T<sub>c</sub> drops below 1-2 nm, so any generic lattice defects can locally suppress Δ(r)
- Precipitation of nonsuperconducting phase on grain boundaries and other materials defects

#### S. Davis et al

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![](_page_15_Figure_0.jpeg)

### Main players

- Cubic, or hexagonal low-T<sub>c</sub> superconductors
- Highly anisotropic layered high-T<sub>c</sub> superconductors
- T<sub>c</sub> seems to scale with chemical complexity
- Layered cuprates with large electron effective mass anisotropy ratio along the c axis and the CuO ab plane:

 $\Gamma = (m_c/m_{ab})^{1/2}$ 

 $Bi_2Sr_2Ca_2Cu_3O_x$  $T_c = 108K$ 

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### **New Iron age: Fe-based superconductors**

![](_page_16_Figure_1.jpeg)

**ReOFeAs** based (1111) ( $T_c = 55K$ ) (Re = Sm, Nd)  $Ba_{1-x}K_{x}Fe_{2}As_{2}$  based (122) (T<sub>c</sub> = 38 K)  $FeSe_{x}Te_{1-x}$  based (11) (T<sub>c</sub> = 18 K)

Poor metals, low  $E_F = 3-100$  meV, short  $\xi=1-2$  nm, huge  $B_{c2} > 100$  T Less anisotropic than cuprates

![](_page_16_Figure_4.jpeg)

0.10

0.12

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### Why does performance of LTS scale with T<sub>c</sub>?

- 1. Large coherence length  $\xi > 3$  nm, large Fermi energy and s-wave pairing result in weak suppression of T<sub>c</sub> by impurities and extended defects.
- 2. No magnetic states competing with superconductivity (structural transition in A15 may enhance T<sub>c</sub>)
- 3. Thermal fluctuations of vortices are weak. The Ginzburg number (squared ratio of thermal energy and condensation energy per Cooper pair) is very small:

![](_page_17_Figure_4.jpeg)

In LTS Gi  $\approx 10^{-10} - 10^{-5}$  but Gi is strongly increased by the large anisotropy parameter  $\Gamma = \xi/\xi_c$  and low carrier density

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### Hard lessons of 30 years of R&D of cuprates and iron pnictides

- Competing charge/spin/... orders + unconventional pairing + layered structure + low carrier density may provide higher T<sub>c</sub> but is <u>a bad</u> combination for applications
- Higher T<sub>c</sub> and non s-wave pairing  $\rightarrow$  small Cooper pairs  $\xi \sim \hbar v_F/2\pi k_B T_c \rightarrow$  sensitivity to benign (in LTS) materials defects
- Competing orders → precipitation of competing AF phase on grain boundaries → current blocking in polycrystals
- Crystalline anisotropy and low carrier density
  - → Strong enhancement of superconducting and vortex fluctuations → significant decrease of the T-H space where pinning of vortices can provide supercurrents
  - $\rightarrow$  Low irreversibility field H<sup>\*</sup> and strong thermally-activated flux creep
  - → Weaker charge screening aggravates current-blocking grain boundaries

Applications at 77K and higher T require addressing many conflicting physics and materials science problems which can be formulated even if we do not know microscopic mechanisms of T<sub>c</sub>

### **Thermal fluctuations of vortices**

Dispersive line tension of a vortex 

$$\varepsilon_l = \frac{\varepsilon_0}{\Gamma^2} \ln \frac{1}{k\xi_c}, \qquad \varepsilon_0 = \left(\frac{\phi_0}{4\pi\lambda}\right)^2 = \frac{\pi\hbar^2 n_s}{4m}$$

Brandt, Rep. Prog. Phys. 58, 1465 (1995); Blatter et al, RMP 66, 1125 (1994)

Electron mass anisotropy,  $\Gamma^2 = m_c/m_{ab} >> 1$  strongly reduces bending rigidity of the vortex:

 $\epsilon_{\ell} \sim 10^4 \text{ K/nm for LTS}$  $\rightarrow$  rigid rods  $\epsilon_{\ell} \sim 30 \text{ K/nm} \text{ (YBCO @ 0K)}$  $\rightarrow$  soft filaments

ε<sub>ℓ</sub> ~ 5 K/nm (YBCO @ 77K)

![](_page_19_Picture_6.jpeg)

Mostly limited by the superfluid density  $n_s(T) = (1 - T^2/T_c^2)n$ , low carrier density n and large mass anisotropy  $\Gamma^2$  = 30-50 in YBCO and  $\Gamma^2$  = 10<sup>3</sup> -10<sup>5</sup> in BSCCO.

### Melting and thermal depinning of solid vortex structure

- J<sub>c</sub> = 0 in the vortex liquid phase B > B<sub>m</sub> where B<sub>m</sub> ≈ the irreversibility field B\*(T)
- Lindemann criterion:  $\langle u^2(T,B_m) \rangle = c_L^2 \phi_0/B_m$ ,  $c_L \approx 0,1-0.3$  (Nelson et al; Blatter et al, Brandt et al; ...)
- Upper branch of the melting field B<sub>c1</sub> << B<sub>m</sub> << B<sub>c2</sub>:

$$H_m(T) = \frac{\pi^3 \phi_0 c_L^2}{4k_B^2} \left(\frac{\hbar^2 n_s}{m\Gamma}\right)^2 \left[\frac{1}{T} - \frac{1}{T_c}\right]^2 \quad \text{Independent of } \mathbf{H}_{c2}$$

For YBCO,  $B_m(77K)\approx 9T$ ,  $B_{c2}(77K)\approx 20T$ 

The melting field  $H_m$  (T) is determined by the ratio  $g = (n_s/m\Gamma)^2$  insensitive to pairing mechanisms

Blatter et al, RMP 66, 1125 (1994) Η H  $H_{c2}$ vortex liquid normal phase vortex solid  $H_{\rm m}$ H<sub>c1</sub> Meissner phase n  $\boldsymbol{T}$ T $H_{c2}(T)$ 10 **YBCO** H[T] 75 80 85 90 T[K]Calorimetric measurements of  $H_m(T)$ , Schilling et al PRL, 78, 4833 (1997); Nature, 382, 791 (1996)

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### Strong suppression of H\* by crystal anisotropy

20 Bi-2223 Ο  $H_{c2}$ YBCO Vortex fluctuations amplified by  $MgB_2$  $\diamond$ anisotropy cancel the benefits 15 of higher T<sub>c</sub> and  $H_{c2}$  ин Bi-2223 H<sub>irr</sub> (T) 10 At 77 K YBCO ( $T_c = 92K$ ) is much better than Bi-2223 ( $T_c = 108K$ ) 5 Nd(F,O)FeAs  $MgB_2$  (T<sub>c</sub> = 40K) or FBS (T<sub>c</sub> < 55K) can be as good as Bi-2223 at 20K < T < 35K, and B < 15T 0 20 40 60 80 100 0 Temperature (K)

### **H-T diagrams of high-field superconductors**

- Only YBCO can be used in magnets at 77 K
- Many choices at 4.2K: MgB<sub>2</sub>, PbMo<sub>6</sub>S<sub>8</sub> and FeSe<sub>05</sub>Te<sub>05</sub>, Bi-2212 can outperform Nb<sub>3</sub>Sn
- At 20-35 K, Ba<sub>06</sub>K<sub>04</sub>Fe<sub>2</sub>As<sub>2</sub> has the second highest H<sub>c2</sub> and H<sup>\*</sup> after YBCO
- For FBS, the difference between H<sub>c2</sub>(T) and H\*(T) is not as big as for cuprates
- Low H\*(T) above 77K can be mitigated by lower anisotropy or higher carrier density.
   Example: Pb doped Bi-2223.

![](_page_22_Figure_6.jpeg)

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### Can J<sub>c</sub> and H\* be improved if we pin every 5-10 nm of the vortex line?

McManus-Driscoll, Nature Materials 3, 439 (2004) (BZO); Harrington et al, SUST 22, 022001 (2009); Haugan et at, Nature 430, 867 (2004) Yamada et al, APL 87, 132502 (2005); Matsumoto et al, JJAP, 44, L246 (2005).; Gutierrez et al, Nature Materials, 6 367 (2007); Obradors et al, SUST 19, S1 (2006); Solovyev et al, SUST, 20, L20 (2007). Rupich et al, MRS Bull., 29, 572 (2004) Obradors and Puig, SUST 22, 044003 (2014)

Combination of nanoparticles and columnar pins

![](_page_23_Picture_3.jpeg)

![](_page_23_Picture_4.jpeg)

![](_page_23_Figure_5.jpeg)

Self assembled BZO nanoparticles

Kang et al, Science 311, 19111 (2006)

![](_page_23_Picture_8.jpeg)

- Splayed columnar defects reduce flux creep at high fields
- Weaker field dependence (reduced  $\alpha$  in J<sub>c</sub>  $\propto$  H<sup>- $\alpha$ </sup>)

#### IEEE-CSC, ESAS and CSSJ SUPERCONDUCTIVITY NEWS FORUM (global edition), Issue No. 57, Oct 2024. Plenary presentation given at ASC 2024, Sept 2024, Salt Lake City, Utah, USA.

### "Designer" APC nanoparticle structures to increase J<sub>c</sub>

Self-assembled chains of BZO nanoparticles

![](_page_24_Picture_3.jpeg)

![](_page_24_Figure_4.jpeg)

![](_page_24_Figure_5.jpeg)

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#### 8 nm YBa<sub>2</sub>CuO<sub>5</sub> nanoparticles

![](_page_24_Figure_8.jpeg)

25 Haugan, et al. Nature 430, 867 (2004)

### Improvement of Jc at high fields by designer pinning in YBCO

- J<sub>c</sub>(77K,B) is increased by several orders of magnitude at 5<B<14 T</li>
- Irreversibility field at 77 K only doubles, from 7 to 14.5 T

- Why does Nb<sub>3</sub>Sn at 4.2K outperform YBCO at 77 K and B > 8T, although they have the same B<sub>c2</sub> and pinning in Nb<sub>3</sub>Sn seems weaker than in YBCO?
- Why cannot pinning push B\*(77K) all the way to B<sub>c2</sub>(77K) = 30 T?

![](_page_25_Figure_5.jpeg)

**Obradors and Puig, SUST 22, 044003 (2014)** 

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### Pinning limit by dielectric nanoprecipitates/pores

- Elliptic critical vortex loops:  $L_{||}L_{\perp} = \ell^2$ ,  $L_{||} = \Gamma L_{\perp}$
- Analog of the Frank-Reed dislocation source with the effective loop width  $L_{\perp} \sim \ell \Gamma^{-1/2}$ ,  $\epsilon/R = \phi_0 J/c$
- Depinning due to reconnection of parallel vortex segments: the smaller the pin spacing the higher J<sub>c</sub>:

$$J_c \cong \frac{c\phi_0}{8\pi^2\lambda^2\Gamma^{1/2}\ell} \ln \frac{\ell}{\xi_c}$$

- $J_c(77K) \sim 9 \text{ MA/cm}^2$  in YBCO is obtained at average pin spacing  $\ell \sim 30 \text{ nm}$
- Too many pins cause T<sub>c</sub> suppression and current blocking

### **Reduction of current-carrying cross section by pins**

![](_page_27_Picture_1.jpeg)

 Effective medium theory for an anisotropic matrix with dielectric precipitates of volume fraction x

$$\rho = \rho_0 \frac{A}{A_{eff}}, \qquad A_{eff} = \left(1 - \frac{x}{x_c}\right)A$$

- The current-carrying cross section A<sub>eff</sub>(x) vanishes at the percolation threshold x<sub>c</sub>
- x<sub>c</sub> = 0.5 in 2D
- $x_c \approx 2/3$  in isotropic 3D

### **Optimum pin density: pinning vs current blocking**

![](_page_28_Figure_1.jpeg)

J<sub>c</sub> due to random insulating precipitates of radius r<sub>0</sub>

Optimum pin spacing and volume fraction:

 $\ell_m \approx 3 - 4r_0, \qquad x_m = \frac{4\pi r_0^3}{3\ell_m^3} \approx 8 - 12\%$ 

Optimum critical current density:

![](_page_28_Figure_5.jpeg)

Gurevich, SUST 20, S128 (2007); Annu. Rev. Cond. Mat. Phys. 5, 35 (2014) Kwok, et al. Prog. Phys. 79, 116501 (2016).

![](_page_28_Figure_7.jpeg)

For  $\Gamma$  = 7 in YBCO,  $J_{cmax} \approx 0.5J_d$  for  $r_0 = \xi$ , and  $J_{cmax} \approx 0.25J_d$  for  $r_0 = 3\xi$ 

#### Upper limit for small pins, no fluctuations and no T<sub>c</sub> suppression by pins.

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![](_page_29_Figure_0.jpeg)

### Why cannot pinning increase $B^*$ to $B_{c2}$ in cuprates?

Shorter soft vortex segments hop easier between pins. increasing pin density can accelerate flux creep

In BSCCO the maximum pinning energy of a short (s =1.5 nm) pancake vortex,  $U_0 = \phi_0^2 s / 8\pi \mu_0 \lambda_0^2 \simeq 350 K$ . E-J characteristic caused by uncorrelated hopping of vortex pancakes at B<< B<sub>c2</sub>:

![](_page_29_Figure_4.jpeg)

where  $J_1 = k_B T / \phi_0 s \ell$ . Irreversibility field at which E(J) turns ohmic at E<sub>c</sub> = 10<sup>-6</sup> V/cm. Case study: Bi-2212 at I = 30 nm.

![](_page_29_Figure_6.jpeg)

![](_page_29_Figure_7.jpeg)

![](_page_29_Figure_8.jpeg)

![](_page_29_Figure_9.jpeg)

30

![](_page_29_Figure_11.jpeg)

To find dopants which can increase the carrier density and reduce electronic anisotropy (increase coupling of ab planes) without suppressing Tc too much

#### **Bi-Pb-substitution in Bi-2212 and 2223**

Reduces anisotropy of  $B_{c2}$  and increases B<sup>\*</sup> at high T but not enough to enable magnet applications at 77K

 $(Cu,C)Ba_2Ca_3Cu_4O_{11+x}$ , T<sub>c</sub>= 116 K, B\*(77)= 12-14 T Zhang et al, Sci. Adv. 4, 0192 (2018).

 $Y \rightarrow RE$  (Ho, Sm, Nd, Eu, Co, Tb, Ce, Pr, La, Dy...) atomic substitutions in YBCO

Improves high-field  $J_c(B)$  and increases B\*(77K) up to 12-13 T, Jha and Matsumoto, Front. Phys. 7, 82 (2019)

Nanoprecipitates or reduced anisotropy?

SmFeAsO<sub>1-x</sub>H<sub>x</sub>,  $T_c = 45$  K H substitution + proton irradiation Miura et al, Nature Mater. (2024)

- Quadruples the depairing current density  $J_d \rightarrow 415 \text{ MA/cm}^2$  and  $J_c(4.2\text{K}, 0\text{T}) \rightarrow 140 \text{ MA/cm}^2$
- Quadruples the carrier density and reduces
   λ down to 120 nm
- Decreases the anisotropy to  $\Gamma\approx 4$
- Vortex line tension  $\epsilon_l(0)\simeq 80\,$  K/nm, better than YBCO

### The grain boundary problem

Ideal 16<sup>0</sup> [001] tilt grain boundary in YBCO

![](_page_31_Picture_2.jpeg)

![](_page_31_Figure_3.jpeg)

Partial grain connectivity In Sm-1111 Kametani et al,

SUST 21, 015010 (2009)

Dimos, Chaudhari and Mannhart, PRB 41, 4038 (1990) Hilgenkamp and Mannhart, APL 73, 265 (1998); RMP 74, 485 (2002)

![](_page_31_Figure_7.jpeg)

2 orders of magnitude drop in Jc with the misorientation angle

Similar in FBS

- Precipitation of AF phase at grain boundaries
- Charge and strain coupling of dislocation cores
- d-wave pairing symmetry

Gurevich and Pashitskii, PRB 57, 13875 (1998); Grazer et al, Nat. Phys. 6, 609 (2010)

### Magnetic granularity in HTS polycrystals

![](_page_32_Figure_1.jpeg)

![](_page_32_Figure_2.jpeg)

![](_page_32_Picture_3.jpeg)

- Only small currents can pass through GBs despite strong pinning of vortices caged in the grains
- Fragmentation of current flow into decoupled current loops in the grains, low transport critical current

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![](_page_32_Figure_7.jpeg)

### **GB weak links in FBS**

 $\theta_{\rm GB}$  (°)

5

10

- Degradation of J<sub>gb</sub> (θ) with the misorientation by 1-2 orders of magnitude
- larger critical angle in FBS might result from weaker pinning in the grains than in YBCO
- GBs in K-doped 122 polycrystals appear better connected than others FBS

#### Why are GBs weak links in cuprates and FBS so alike?

- Cannot be the pairing symmetry (d-wave in cuprates and multiband s-wave in FBS)

 $J_{\rm c}^{\rm GB}/J_{\rm c}^{\rm Grain}$ 

0.01

0

- Pairing mechanisms are different
- Phase diagram with competing AF states are similar
- Both are poor metals

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![](_page_33_Figure_11.jpeg)

SUST, 33, 043001 (2020)

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NdFeAs(O,F) (20 µm) [29]

NdFeAs(O,F) (30 µm) [29]

▲ NdFeAs(O,F) (40 μm) [29]

25

30

□ Co-doped Ba122 [7]

20

![](_page_33_Figure_12.jpeg)

![](_page_33_Figure_13.jpeg)

Si et al APL 106, 32602 (2015);

Sarnelli et al. APL 104, 162601<sup>4</sup>(2014)

### Why are GBs weak links in cupates and pnictides?

- Competing AF states, sensitivity of T<sub>c</sub> to small shifts of chemical potential due to local non-stoichiometry, strains or GB charges
- Small E<sub>F</sub> = 300-500 meV in cuprates and E<sub>F</sub> = 10-200 meV in FBS.
   If E<sub>F</sub> = 30 meV = 330K electron gas becomes classical at room temperatures.
- Poor screening, large TF screening length, l<sub>TF</sub> ≈ ξ ≈ 1-2 nm.
   GB charges revealed by electron holography Schofield, et al. PRL 92, 195502 (2004).

Suppression of SC gap on GB (Gurevich and Pashitskii, PRB Durrell et al, Rep. Prog. Phys. 74, 12451 (2011)

$$\frac{\Delta_0}{\Delta} = \frac{1}{\sqrt{1 + \Gamma^2} + \Gamma}, \qquad \Gamma = \frac{2^{3/2} \pi q e l_{TF}^2}{s b \xi_0 T_c \sqrt{\tau}} \left[ \frac{\partial T_c}{\partial \mu} \right] \sin \frac{\theta}{2}$$

![](_page_34_Figure_6.jpeg)

Clouds of impurities in strain and electric fields around GB

Song et al Nature, Mat. 4, 470 (2005)

Dislocation charge q = e (per ab plane) shifts  $\mu$  by  $\approx$  250 meV. Strong band bending effects.

 $I_{TF} \approx \xi_0 \approx 1-2$  nm, and  $dT_c/d\mu \approx 0.4-0.8$  K/meV result in  $\Gamma > 1$  at  $\theta > 10^\circ$  and 77K ( $\tau = 1 - T/T_c$ )

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### Low carrier density in cuprates and pnictides aggravates current-blockage by grain boundaries

Constant electrochemical potential in equilibrium:  $\mu_e = E_F[n(x)] + e\varphi(x) = const.$ 

![](_page_35_Figure_2.jpeg)

### Improving grain boundaries

- Add more holes to GBs by local overdoping
- Reduce local strains at GBs
- Doping and stress can reduce T<sub>c</sub> and B<sup>\*</sup>

#### YBCO

**Ca overdoping**: improves  $J_{gb}$  but reduces  $T_c$  and  $B^*$ . Up to 8-fold increase of  $J_{gb}$  at 4K, OT for a 24° tilt GB. Replace 30% of Y<sup>3+</sup> by Ca<sup>2+</sup> Schmehl et al., Europhys. Lett. 77, 110 (1999); Hammerl et al., Nature 407, 162 (2000)

30-40% increase of  $J_{gb}$  for a 5° GB at 44K at 0-3 T Daniels, Gurevich, Larbalestier, APL 77, 3251 (2000)

#### 10% $Nd^{3*}$ substitution for $Y^{3+}$

Strain management, critical angle increases to  $6^{\circ}$ , no T<sub>c</sub> degradation, modest increase of J<sub>c</sub> of a  $9^{\circ}$  GB Kim and Larbalestier, SUST 34, 025008 (2021)

#### Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> tapes

Improve intergrain connectivity by K overdoping at the expense of reduced  $T_c$ , (max  $T_c$  at x=0.497)

Cheng et al, Materials Today Phys. 28, 100848 (2022)

![](_page_36_Figure_12.jpeg)

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### **Conductor tape technology**

On recent progress, see Obradors et al., SUST 37, 053001 (2024)

YBCO coated conductor

On progress in FBS films, see Iida et al, SUST 36, 063001 (2023) BaK122 tapes and wires

![](_page_37_Picture_3.jpeg)

#### Molodyk and Larbalestier, Science, 380, 1220 (2023)

- Grow YBCO on biaxially-textured substrates to eliminate high-angle GBs
- High price of the weak-linked grain boundaries
- Complex, expensive, only a tiny fraction carries current, high ac losses
- J<sub>c</sub> of YBCO layer must be pushed to its limit
- Industry produces km long second generation YBCO coated conductors

![](_page_37_Picture_10.jpeg)

![](_page_37_Picture_11.jpeg)

Dong, Hu and Ma, Nat. Sci. Rev. 11, 122 (2024)

![](_page_37_Figure_13.jpeg)

### Making old/new superconductors useful

For applications at T = 4.2 K, the conventional materials optimization works for all SCs: NbTi, A15, PbMo<sub>6</sub>S<sub>8</sub>, MgB<sub>2</sub>, YBCO, Bi-2212, Bi-2223, ...

- High T<sub>c</sub> produce high H<sub>c2</sub> which can be further increased by impurities. Large field space  $0 < B < B^* \approx B_{c2} \lesssim 200 T$
- Designer APC nanostructures to produce high critical current densities
- Ameliorate current-blocking GBs (Nb<sub>3</sub>Sn, PbMo<sub>6</sub>S<sub>8</sub>, FBS, cuprates)

#### **Intermediate temperatures 5 < T < 60 K**

For not very anisotropic SCs with Tc < 55 K, (MgB<sub>2</sub>, FBS,...) B<sup>\*</sup> is not much lower  $B_{c2}$ , and the conventional optimization still works if the GB problem is addressed.

#### Materials wish list for applications at T = 77K and higher: only YBCO for now. In searching for new SCs with Tc > 90 K pay attentions to:

- 1. Carrier density: the higher the better
- 2. Electronic anisotropy: the smaller the better (less anisotropic than YBCO)
- 3. Thomas Fermi screening length: the smaller the better
- The higher the operation temperature, the less parameter space gets available to satisfy the constrains on carrier density and anisotropy
- The higher the operation temperature, the less relevant T<sub>c</sub> and H<sub>c2</sub> become, moderate increase of B<sup>\*</sup> by pinning
- Non-s-wave pairing and competing magnetic order complicate applications

### Superconductivity limited by thermal fluctuations

Ginzburg number:

$$Gi = \frac{\Gamma^2}{2} \left( \frac{k_B T_c}{H_c^2 \xi^3} \right)^2 \propto \left( \frac{T_c^2 m \Gamma}{v_F n_s} \right)^2$$

 $\Delta T \sim -T_c G i^{1/2} \propto T_c^3 m^* \Gamma n_s^{-4/3}$ 

- cuprates: Gi ~ 0.1-10<sup>-2</sup>,
- 122 pnictides and MgB<sub>2</sub>: Gi ~ 10<sup>-4</sup> 10<sup>-3</sup>
- 1111 pnictides: Gi ~ 10<sup>-2</sup>
- T<sub>c</sub> reduction by fluctuations goes up rapidly with T<sub>c</sub> and is increased by anisotropy and low carrier density

Low vortex line tension:  $\varepsilon_1 = 1-10 \text{ K/nm} \rightarrow \text{small energy barriers for vortex hopping between pins U = <math>|\varepsilon_0/\Gamma \cong 100 \text{ K}$ of soft vortex segments I  $\cong 10-100 \text{ nm} \rightarrow \text{reduction of H}^*$ , no matter how strong pinning is

$$B_m(T) = \frac{\pi^3 \phi_0 c_L^2}{4k_B} \left(\frac{\hbar^2 n}{m^* \Gamma}\right)^2 \left(\frac{1}{T} - \frac{1}{T_c}\right)^2 \qquad \epsilon_l \simeq \frac{\pi \hbar^2 n}{4m\Gamma^2} \left(1 - \frac{T}{T_c}\right) \qquad \text{Independent of } \mathsf{T_c} \text{ at low }$$

For a putative RTS with  $T_c = 400$  K and  $B_{c2} = 1000$  T and carrier density and anisotropy of YBCO, the irreversibility field may not exceed 15 T at 77K

Applications at 300 K and high fields would require a nearly isotropic RTS with T<sub>c</sub> > 350 K and carrier density of LTS

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## If a RTS with $T_c = 400K$ and $B_{c2} = 1000T$ has been discovered, can its full potential be realized at 300K?

- Tough materials requirements to enable room temperature superconducting magnets even for isotropic RTS.
- Much easier to satisfy these requirements at 77K
- Cheap and environmentally safe liquid nitrogen cooling enables cryostability and ac loss management of composite conductors
- An isotropic superconductor with T<sub>c</sub> = 240 K and LTS carrier density can be revolutionary for applications at 77K
- Chasing genuine RTS may not necessarily result in a magnet technology which can be used even at 77K
- Magnetic pressure In a magnet p = B<sup>2</sup>/2µ<sub>0</sub>: 1T produces 4 atm, 10 T = 400 Atm, 100 T = 40,000 atm. Mass of mechanical support M = CB<sup>2</sup>, big problems with mechanical stability

### **Challenges and opportunities**

### **Challenges of improving the existing HTS:**

- 1. Reduce strong thermal fluctuations of vortices
- 2. Increase the irreversibility field
- 3. Reduce pairbreaking by grain boundaries
- 4. Multiparameter materials optimization and design optimum pinning nanostructures are required

### **Quest for new materials**

- RTS with T<sub>c</sub> = 350K could be useful at 300K if they are:
  - nearly isotropic
  - exhibit no segregation of competing phases on grain boundaries
  - have a higher superfluid density, that is,  $\lambda \leq 100$  nm like in Nb<sub>3</sub>Sn or MgB<sub>2</sub>
  - preferably non-d-wave
- Moderately anisotropic RTS ( $\Gamma$  < 10) may be very useful for high field applications at 77K
- Nearly isotropic HTS 150 < T<sub>c</sub> < 300K or cubic superconductors with T<sub>c</sub> = 90-150K could revolutionize power/magnet applications at 77K
- The success of MgB<sub>2</sub> and weakly anisotropic pnictides should inspire search for moderately anisotropic "intermediate/high T<sub>c</sub>" superconductors

## History is full of grim predictions on superconductivity: no high-field, no high-T<sub>c</sub>, no applications, no RTS, no ...

![](_page_42_Picture_1.jpeg)

The pessimist sees difficulty in every opportunity. The optimist sees the opportunity in every difficulty.

**Winston Churchill** 

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