High-Field Intermetallic Superconductors

A proposal to the

DOE Center of Excellence for the Synthesis and Processing of Advanced Materials

From

Ames Laboratory
Argonne National Laboratory
Brookhaven National Laboratory
Idaho National Environmental and Engineering Laboratory
Los Alamos National Laboratory
National Renewable Energy Laboratory
Oak Ridge National Laboratory

Arizona State University
University of Kentucky
The Ohio State University
Pennsylvania State University
Princeton University
University of Wisconsin

Lance Cooley (BNL), contact

Project

2 Workshops – 3/24/04 at APS Montreal, 4/27/04 at ANL

<u>Objective:</u> Understand and control the materials science of high-field forms of MgB_2

- Understand physics of electron scattering in unique two-band framework
  - Opportunities to attain new limits are not available with any other superconductor
  - Other properties make breakthrough possible
- Understand materials science:
  - of homogeneously alloyed single phase MgB_2
  - of precipitates and nanostructures in unalloyed MgB_2
  - of precipitates in alloyed MgB_2

Motivation

- High Field = better than copper-iron electromagnet
- ***All HFSC are intermetallics*** except for Nb-Ti alloy
  - Why? T_c and H_c are high enough in intermetallics.
  - Excitement : Y_2C_3, B-diamond, $PuCoGa_5$, osmates, pyrochlores
- ***Few*** intermetallics can be ***made in useful forms***
  - Should be isotropic, too
- ***Alloying is essential*** to improve H_{c2}
  - But: intermetallics are much more susceptible to losing superconductivity upon alloying than metals are

MgB_2 satisfies all of these prerequisites, hence is best candidate for focused study

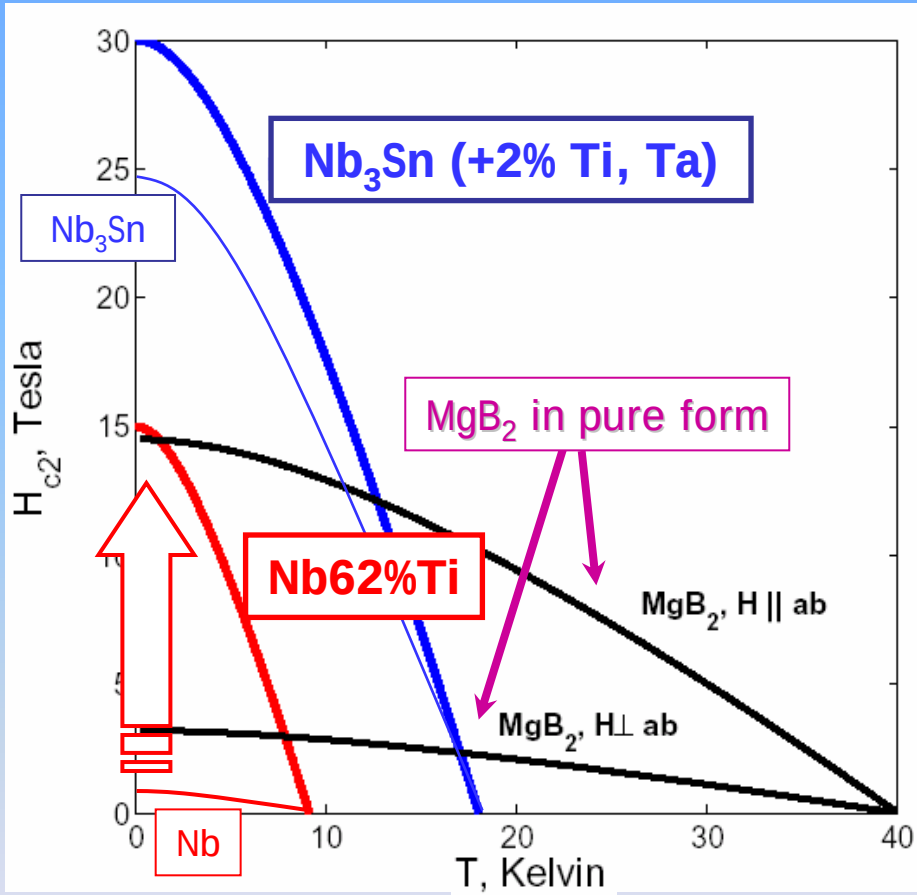
- Interesting physics, cheap raw materials, processing that scales to long length, high-field properties that are interesting

Science of HFSC

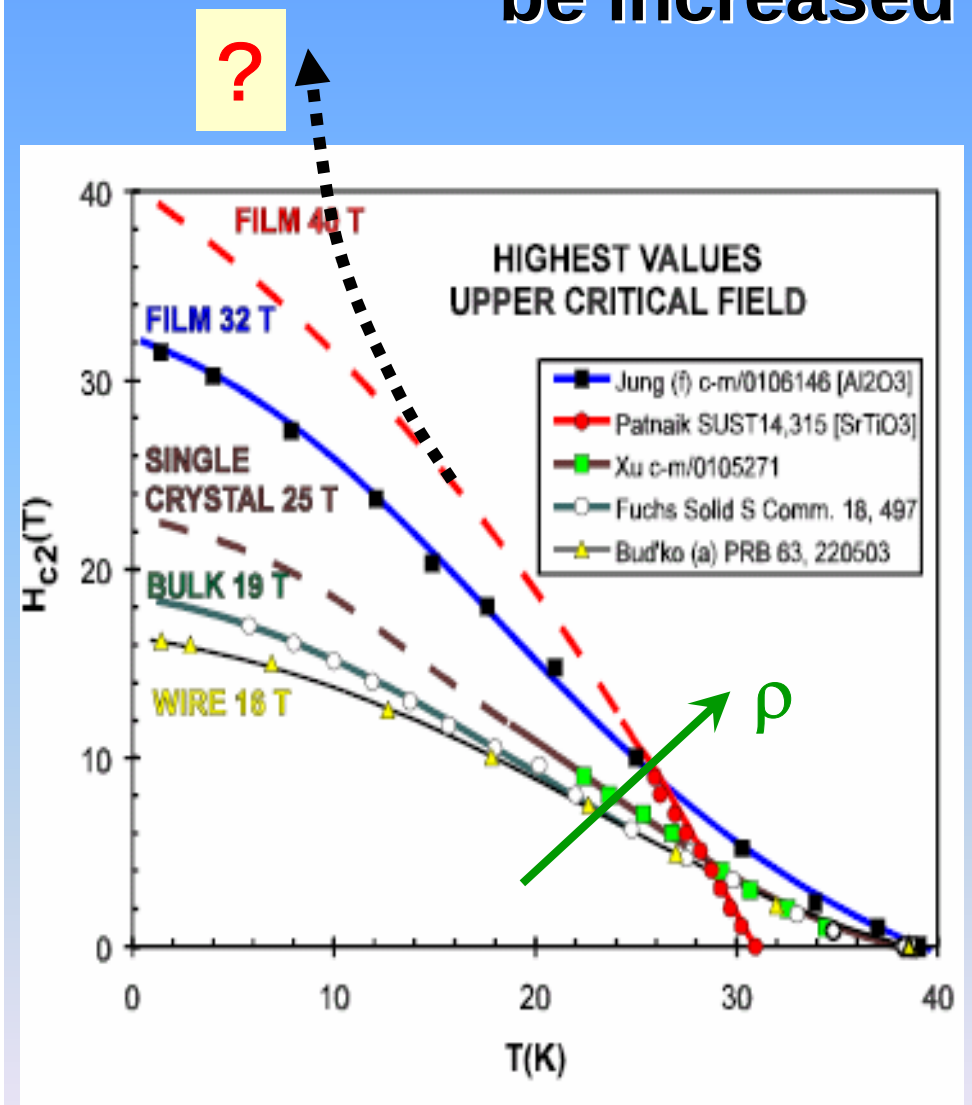
- Conventional SC (1-band, spherical Fermi surface):

  $\mu_0 H_{c2}(0) = 3110\ \rho_N\ \gamma\ T_c$ tesla

  - Alloying drives up H_{c2}
  - **Note 62% Ti in Nb-Ti vs. 2% Ti, Ta in Nb_3Sn**
- Improving $H_{c2}(0)$ without reducing T_c improves properties at ***all*** fields and temperatures
- H_{c2} determines *possibilities*, however *flux-pinning determines practical performance limits*



Late 2002: How far can H_{c2} of MgB_2 be increased by disorder?



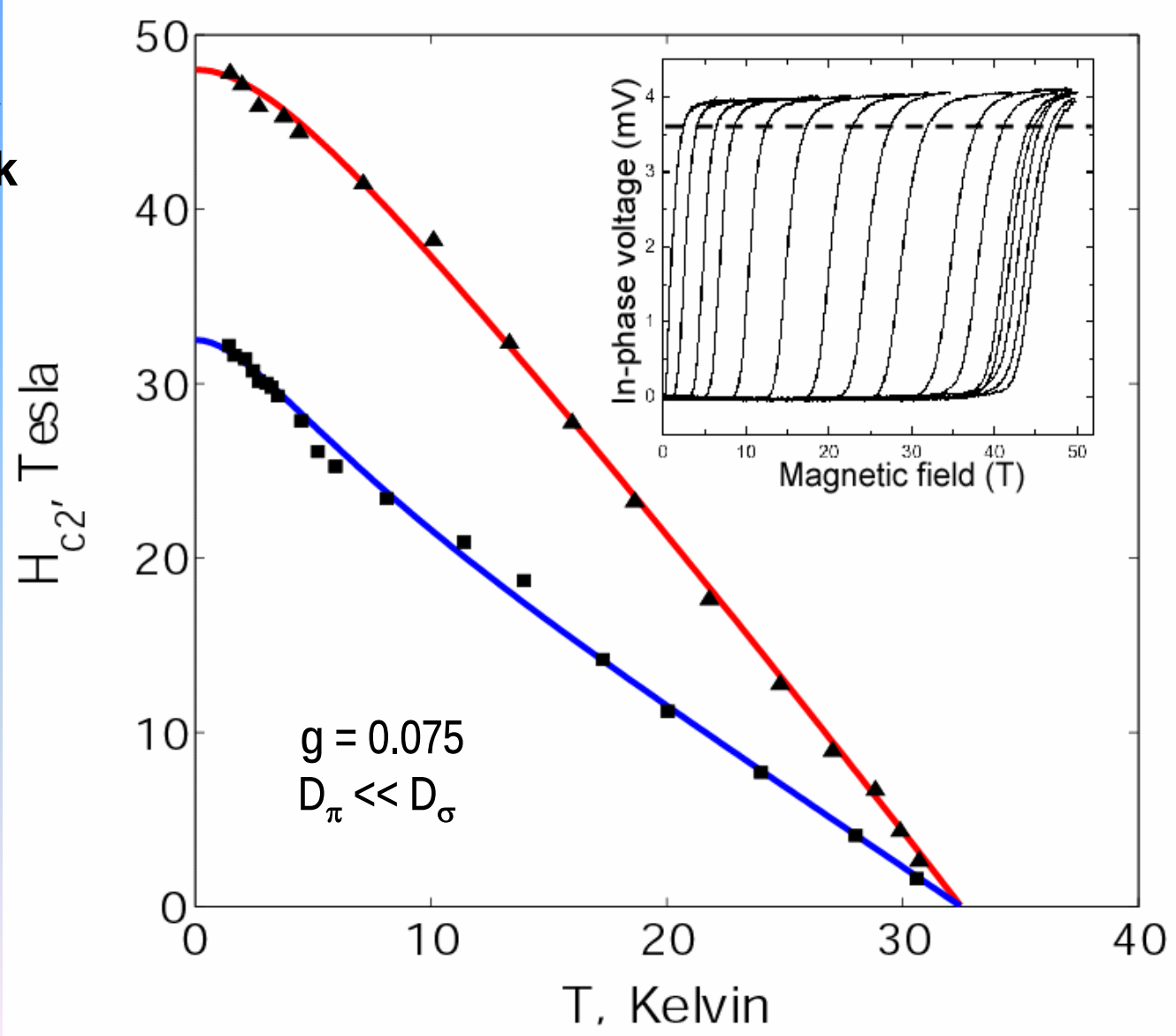
- Which way does $H_{c2}(T)$ go at low T ?
- What is $H_{c2}(0)$ for $(dH_{c2}/dT)|_{Tc} \approx$ -1 to -2 T/K ?
- Can MgB_2 compete with HTS at 20-30 K?
- **Does the one-gap theory work for MgB_2 in the dirty limit?**

$$H_{c2}(0) = 0.7\ T_c\ H_{c2}'$$

$$H_{c2}' = 4ec k_B \pi^{-1} N_F\ \rho_N$$

50 T result shown at preproposal

Measured previously at LANL by S. Patnaik
Film by C.B. Eom
ρ = 220 $\mu\Omega$cm



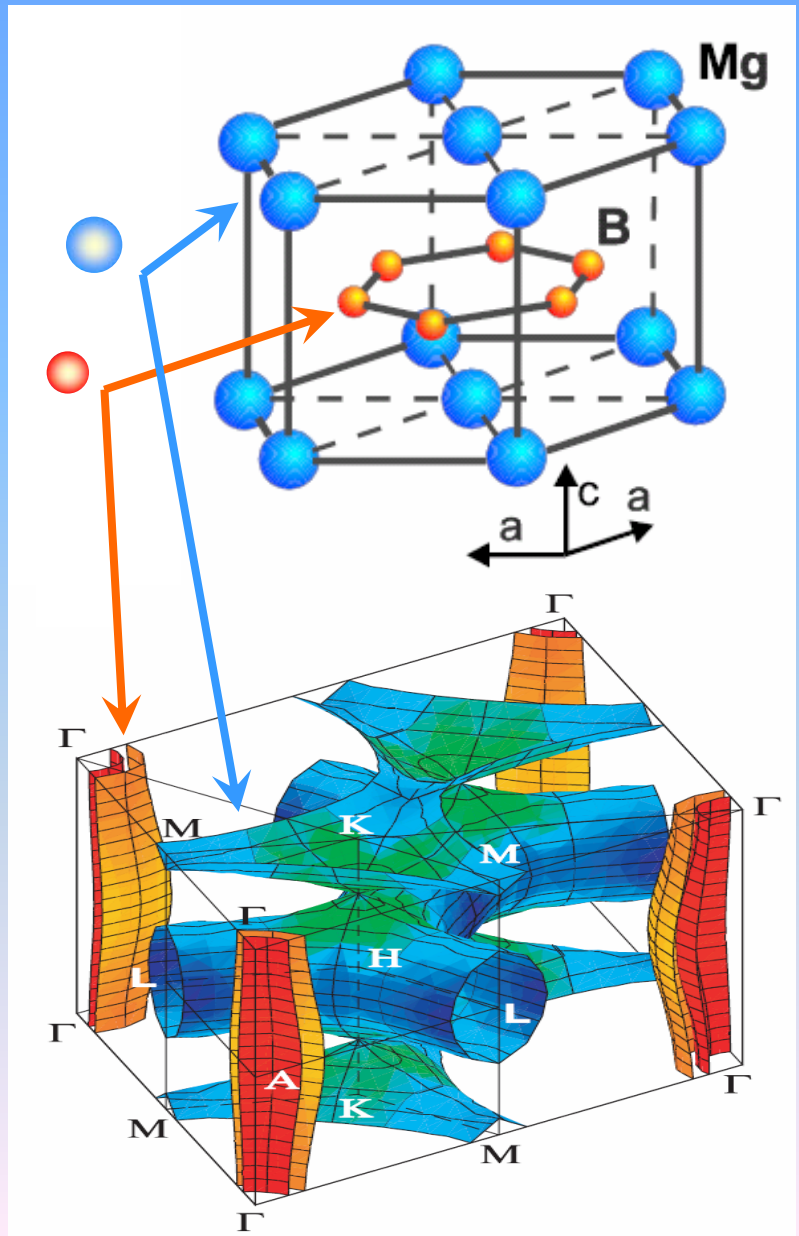
A. Gurevich et al.,
SuST, 17, 278 (2004)

Tunable impurity scattering

- Mg substitution introduces ***intra***band scattering in 3D π band
- B substitution: introduces ***intra***band scattering in 2D σ band
- ***Inter***band scattering is weak but not negligible
- Quenched impurity or vacancy structures also produce scattering
- Electron diffusivities are anisotropic:

$$D_{\sigma}^{(c)} << D_{\sigma}^{(ab)} \quad D_{\pi}^{(c)} \approx D_{\pi}^{(ab)}$$

The ratio $D_{\sigma}^{(ab)} / D_{\pi}^{(ab)}$ is a variable material parameter.



Dirty-Limit Theory of 2-gap SC

A Gurevich – Wisconsin PRB 67:148515 (2003);
Golubov (Twente) and Koshelev (ANL) – PRB and PRL 2003

Result 1: Critical Temperature

Interband *coupling* always increases T_c

Suhl, Mattias, Walker; Moskalenko (1959):

$$T_{c0} = 1.14\omega_D \exp[-(\lambda_+ - \lambda_0)/2w],$$

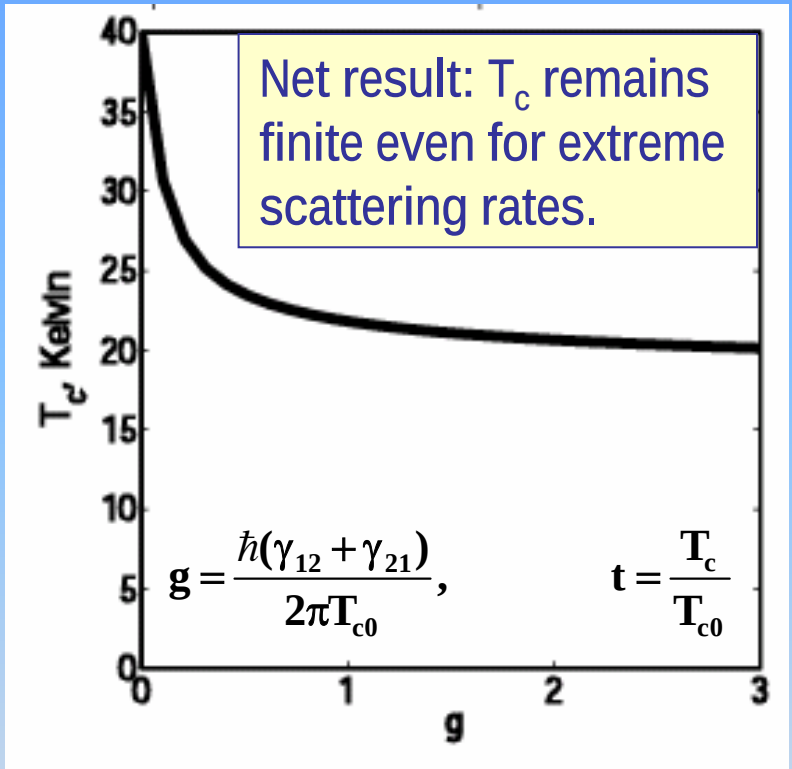
$$\lambda_\pm = \lambda_{11} \pm \lambda_{22}, \qquad \lambda_0 = \sqrt{\lambda_-^{\,2} + 4\lambda_{12}\lambda_{21}},$$

$$w = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21},$$

But: interband *scattering* breaks pairs, decreases T_c

$$\psi\left(\frac{1}{2} + \frac{g}{t}\right) - \psi\left(\frac{1}{2}\right) = -\frac{(\lambda_0 + w\ln t)\ln t}{p + w\ln t},$$

Moskalenko et al; Golubov and Mazin (1996); Gurevich (2003)



Interband scattering is weak (g << 1) due to orthogonality of σ and π orbitals (Mazin et al, 2002)

Does this remain valid with alloying?

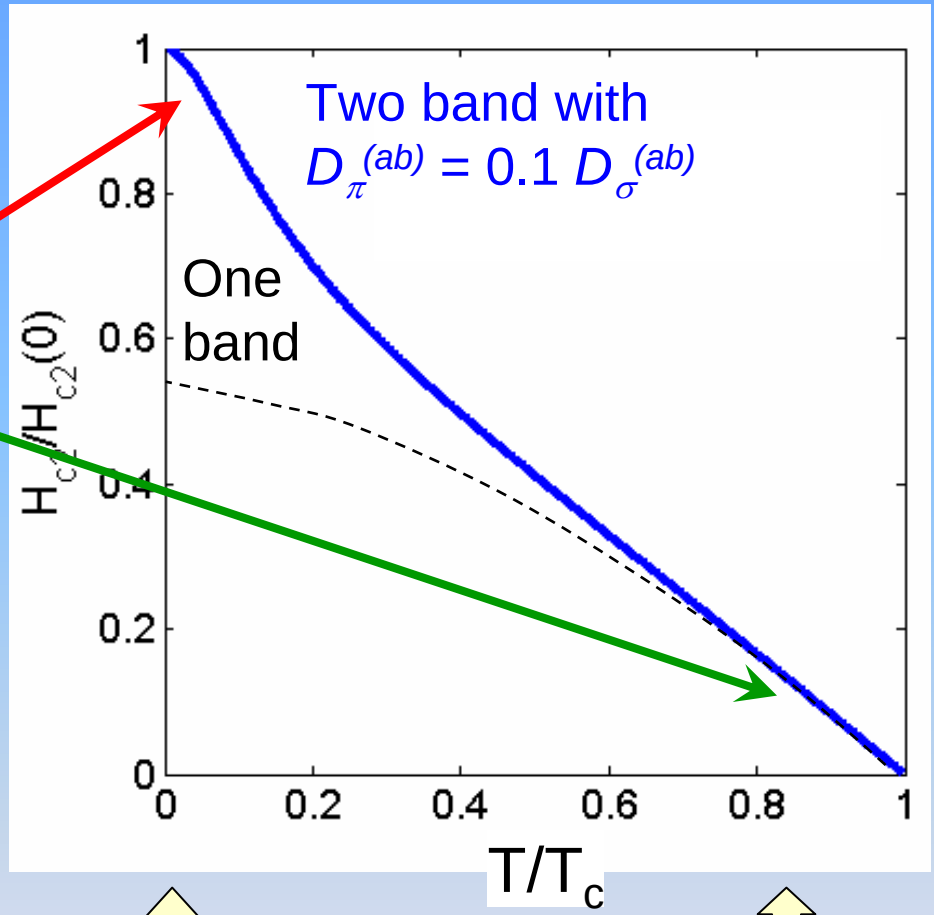
Dirty-Limit Theory of 2-gap SC

Result 2: The shape of $H_{c2}(T)$

- $H_{c2}(0)$ is determined by *minimum* diffusivity of either band, and can turn ***upward***
- $H_{c2}(T_c)$ is determined by *maximum* diffusivity of either band
- $H_{c2}(0)$ can be **much higher** than the one-band extrapolation
  - **50-80 tesla are possible**
- For $D_\pi << D_\sigma$, paramagnetic effects at very high field become important
  - One band:

    $\mu_0 H_p = 1.84\ T_c\,[K]$ tesla

    $\approx$ **70 tesla**



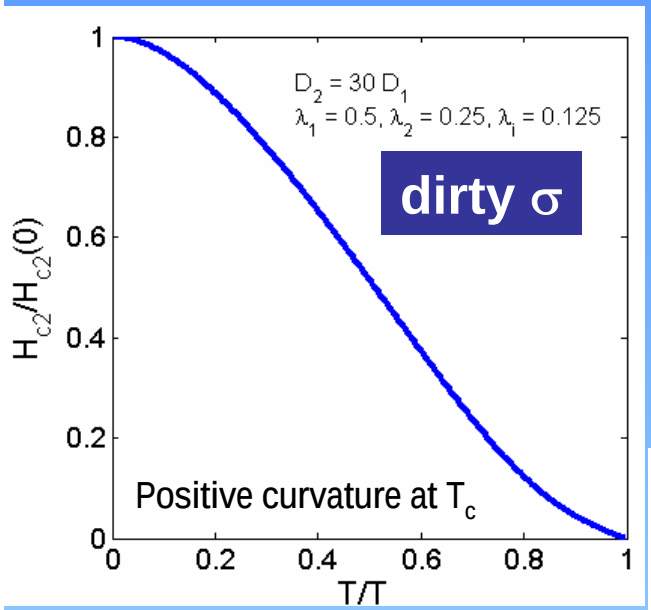
$$H_{c2}(0) = \frac{\phi_0 T_c}{2\gamma\sqrt{D_1 D_2}} \exp\left(\frac{g}{2}\right)$$

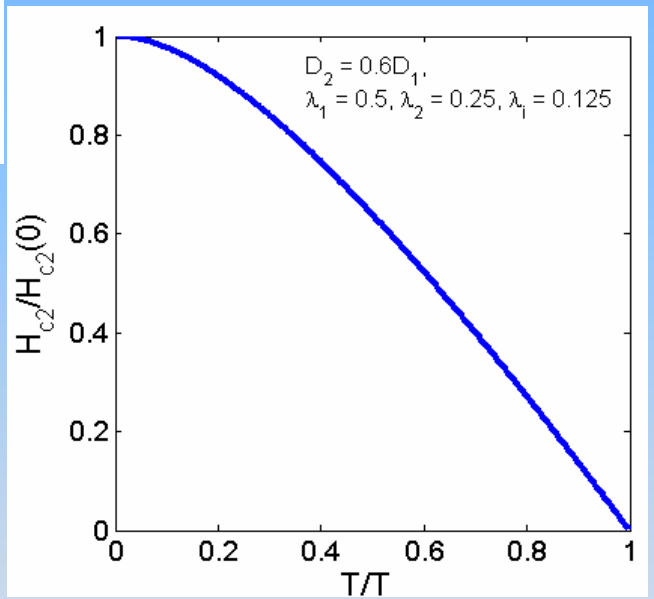
$$H_{c2} = \frac{8\phi_0 (T_c - T)}{\pi^2 (a_1 D_1 + a_2 D_2)}$$

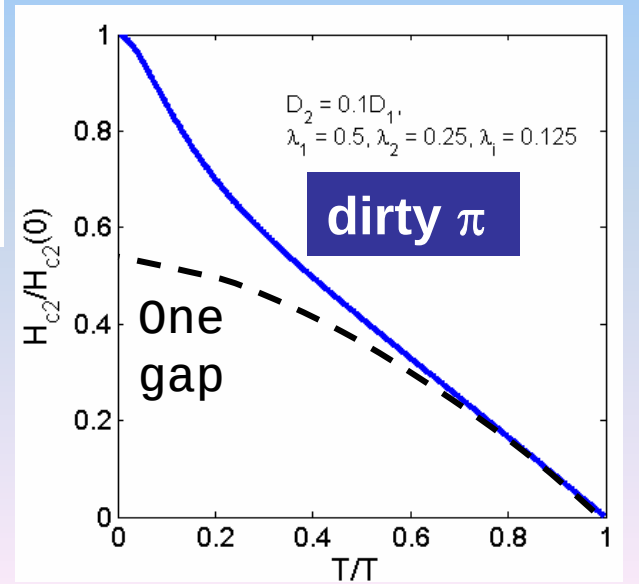
Interplay of different scattering channels

Result 3: Anisotropy can decrease

Strong H_{c2} enhancement by disordering Mg sublattice







Increase π scattering relative to σ scattering

Proposal Team

- 2 workshops
  - 3/24 at Montreal (APS)
  - 4/27 at Argonne
- 13 Labs

Ames: M. Angst, S. Bud'ko, P. Canfield*, D. Finnemore, V. Kogan, D. Wilke

ANL: D. Hinks, M. Iavarone, J. Jorgensen, A. Koshelev, U. Welp*

ASU: N. Newmann, J. Rowell

BNL: L. Carr, L. Cooley*, R. Klie, Q. Li, A. Moodenbaugh, C. Petrovic, D. Welch

INEEL: H. Farrell

Kentucky: G. Cao, K.-W. Ng

LANL: L. Civale*, M. Jaime, X. Liao, A. Serquis, Y. Zhu

ORNL: D. Christen, M. Paranthaman*, J. Thompson

NREL: Y. Yan

OSU: E. Collings, M. Sumption

PSU: A. Pogrebnyakov, J. Redmond, X. Xi

Princeton: R. Cava

Wisconsin: C.-B. Eom, A. Gurevich*, T. Heitmann, E. Hellstrom, D. Larbalestier, S. Patnaik, M. Rzchowski, B. Senkowicz, X. Song

* Steering committee

- Industry and End-Users:
  - General Electric
  - HyperTech Inc.
  - Superconductor Technology Inc.
  - Specialty Materials Corp
  - MIT Plasma Science and Fusion Center

Synthesis routes toward high-field MgB_2

These routes work

- Alloy with carbon
  - Best control so far
  - Bulk: high temperature
  - CVD: gas mix
- Irradiate with neutrons
- Force Mg-sublattice disorder
- Add oxygen interstitials
- Make nanostructured, use strain

- Alloying for Mg doesn't work?!

Overview of work planned

- Doped single crystals
  - *No US source right now!*
  - ANL, Ames
- Thin films
  - ASU, ORNL, Penn State, Wisconsin, Industry
- Dense, well-connected bulk, wires
  - Ames, BNL, LANL, ORNL, OSU, Princeton, Kentucky, Wisconsin, Industry
- Samples with controlled precipitates, pinning centers

Most important need:
Reference base of benchmark samples !!

Thin films are best so far; lack control

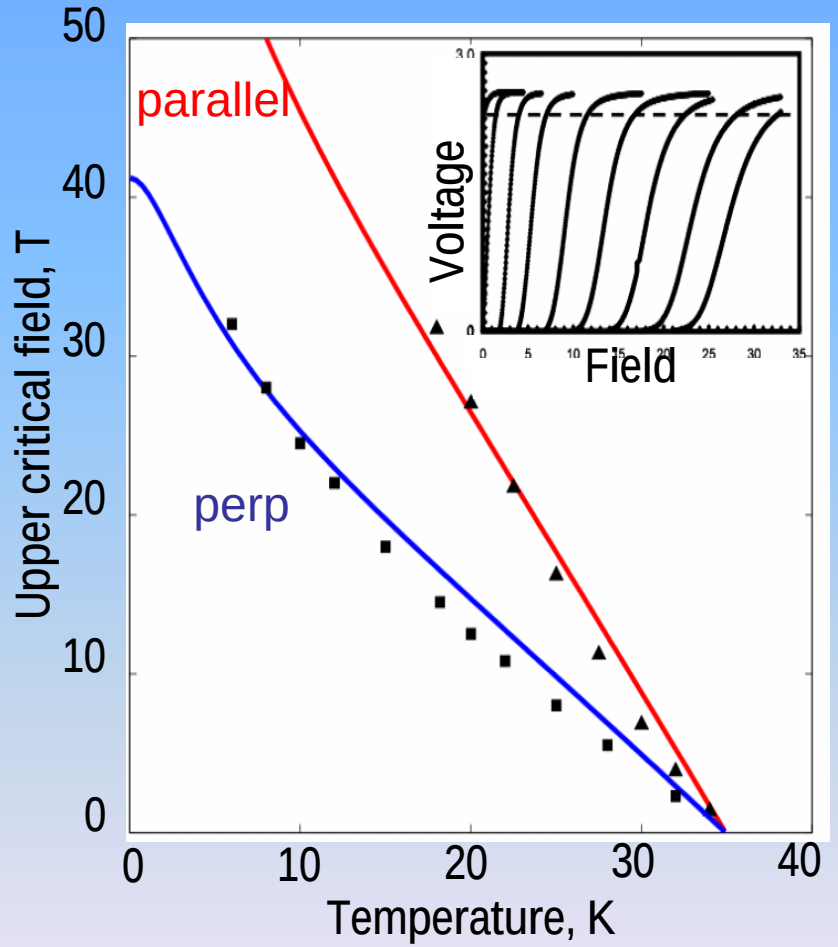
- 12 different alloyed and pure thin films
  - UW-ASU-PSU-Conductus-NHMFL-Genoa-Naples collaboration
- Numerous synthesis and characterization issues

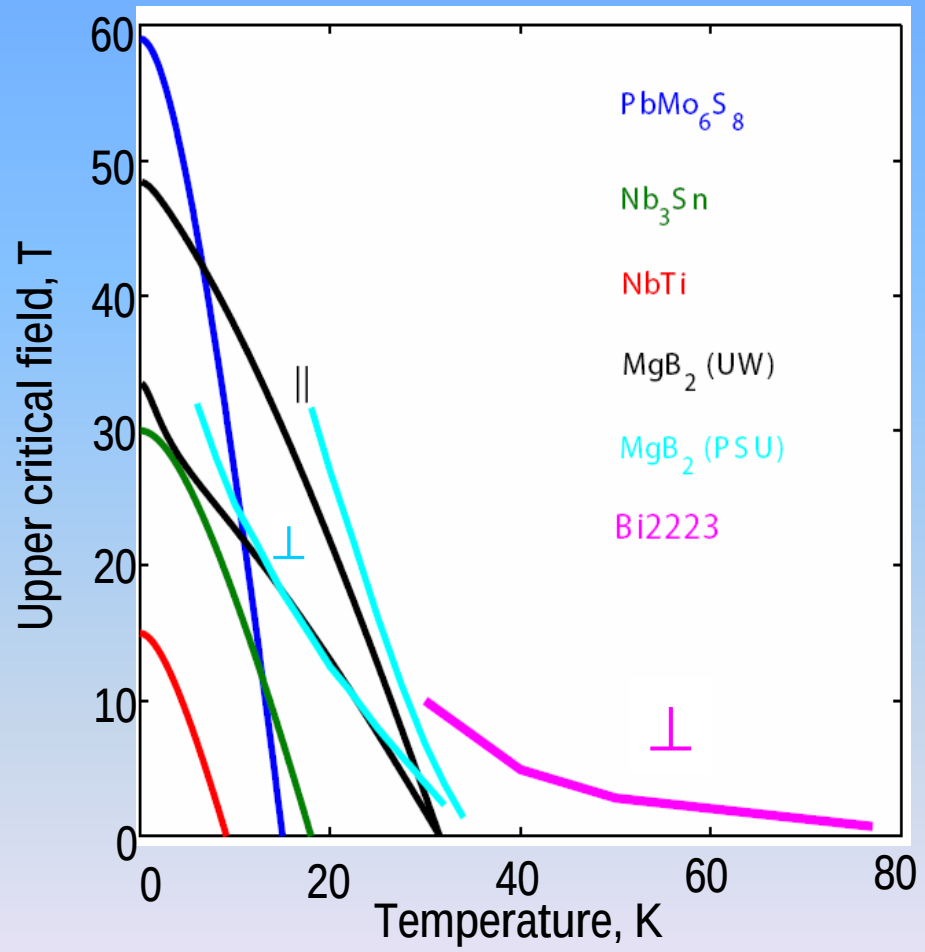
Samples	Substrate	T_c (K)	ρ_n(40K) ($\mu\Omega$cm)	$H_{c2}^{\perp}$ (T)	$H_{c2}^{\parallel}$ (T)	g	D_π/D_σ	c (Å)	a (Å)	Mg at%	B at%	C at%	O at%
A epitaxial [16]	(0001)Al_2O_3	35	9(4)	13.5	33	0.045	0.12	3.516	3.047	29	53	10	8
B fiber-textured [16]	(0001)Al_2O_3	23.7	86(56)	17	17	0.5	<<1	-	-	28	57	7	8
C epitaxial*[16]	(0001)Al_2O_3	34	7	20.5	30	0.06	<<1	3.52	3.08	-	-	-	-
D fiber-textured*[13]	(111)$SrTiO_3$	31	220	33	48	0.075	<<1	3.547	-	37	32	14	17
E epitaxial	SiC	41.5	1.6(0.4)	12	34.5	-	-	3.511	3.107	30	57	2	11
F fiber-textured [15]	SiC	35	564	40	>74	0.045	<<1	3.542	3.055	26	46	21	6
G fiber-textured [15]	SiC	35	250	28.2	55.5	0.045	0.065	3.536	3.057	25	42	26	6
H epitaxial [15]	SiC	38	10.5	10.5	30	0.025	0.06	3.519	3.107	31	63	4	1
I untextured [14]	(0001)Al_2O_3	32	567(290)	21.7	26.8	0.09	0.08	-	-	-	-	-	-
L no 00l textured [18]	r-cut Al_2O_3	39.4	9.9(2.8)	10.8	21.4	0.025	0.07	-	-	32	65	1	1
M epitaxial [12]	(111)MgO	33.5	47	14.6	38.1	0.095	0.1	3.533	3.036	24	41	28	6
N untextured [17]	(001)MgO	28.6	400	15.8	24.3	0.155	<<1	-	-	33	53	5	9

This is an example of how this CSP project can be effective!

The Champion: ~75 T (||), ~40 T (⊥)

- But: Nanostructured, intergrowths, strain, reproducibility

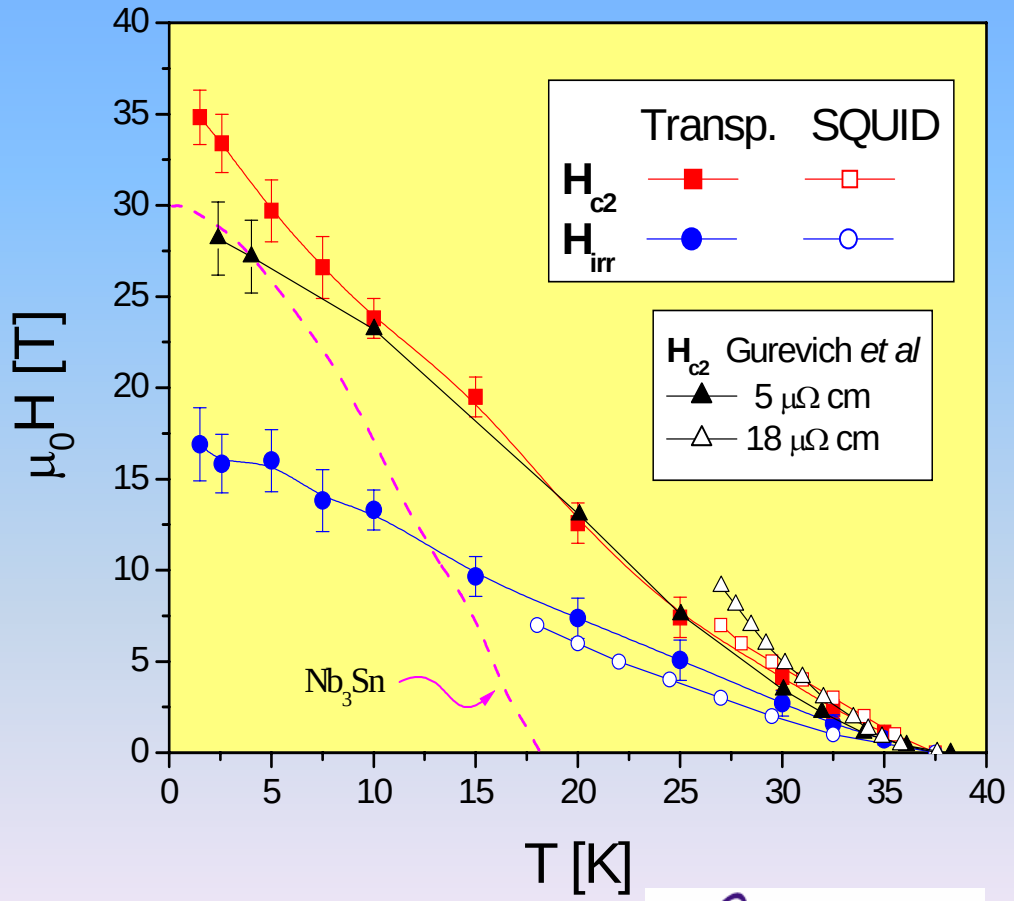




Bulk samples show improvement, too

- But no bulk sample has properties close to those in films – how to make alike?
- Good properties arise by accident
- Again, numerous synthesis, characterization issues

MgB_2 + 10% SiC, Dou *et al.* (Woolongong)
Plus hot-pressing at LANL/UC
Measurements at NHMFL-LANL

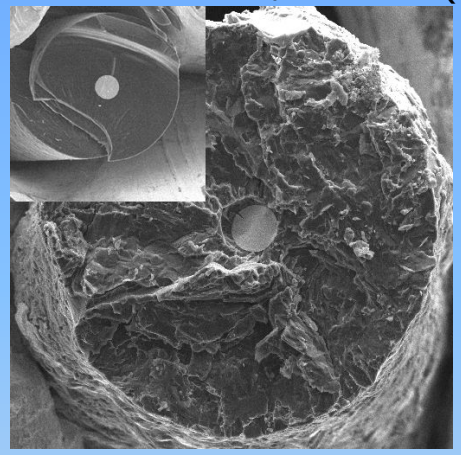


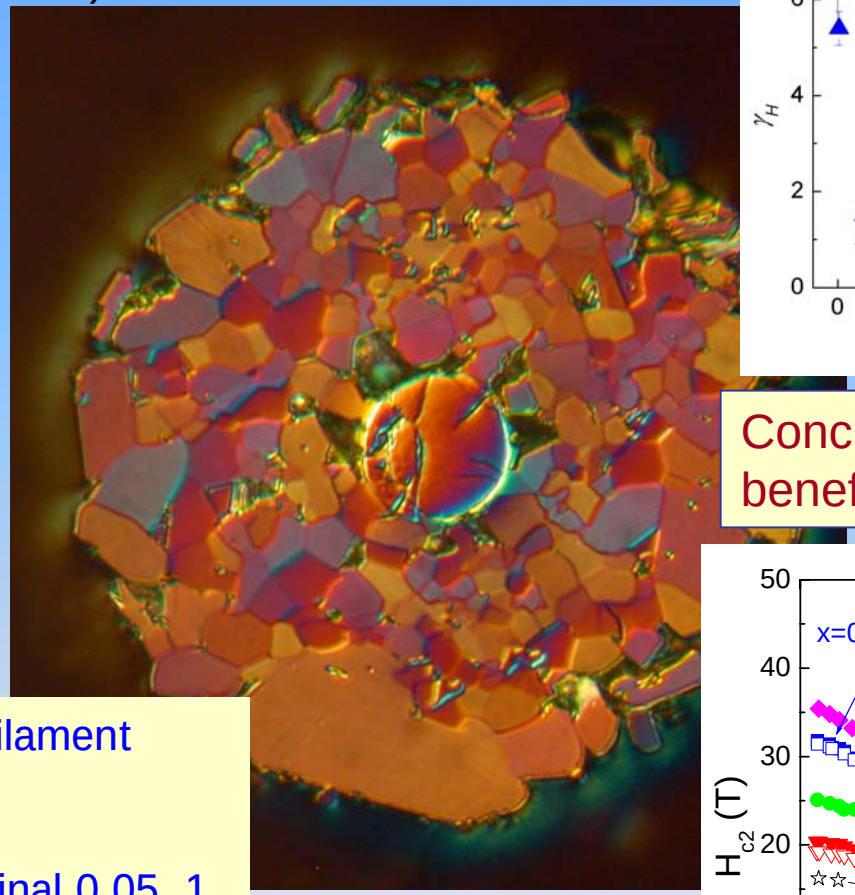
UC University of California

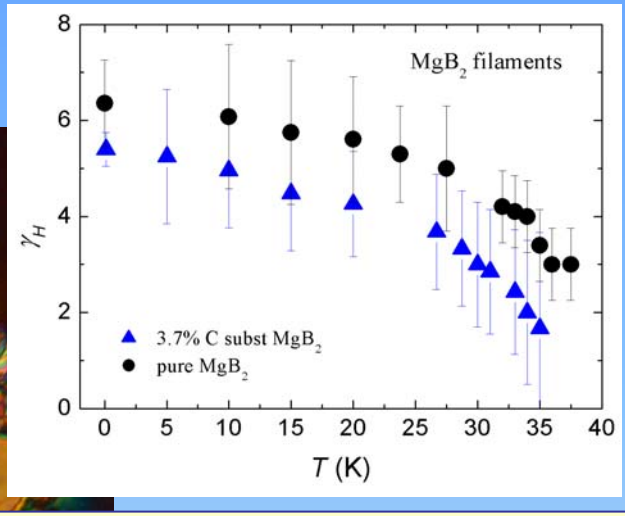
Los Alamos

Boron (+ carbon) filament, reaction in Mg vapor

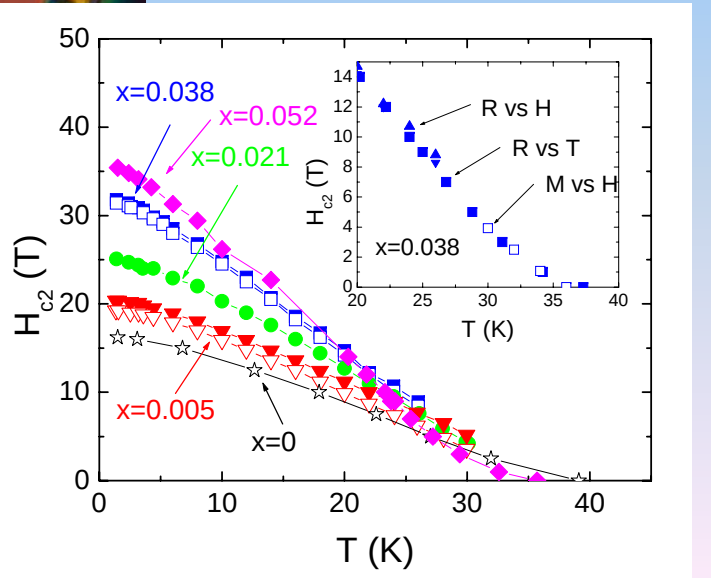
MgB_2 from boron filament: Ames and Specialty Materials Inc.
Canfield et al., PRL 86 (2001) 2423







Conclusion: carbon doping benefits are offset by Tc loss.



Nominal $Mg(B_{0.98}C_{0.02})_2$ filament
Diameter ~ 100 μm

So far samples with nominal 0.05, 1, 2, 3.3 %% carbon were synthesized
Wilke et al, PRL to appear

Characterizations of high-field MgB_2

Probes of 2-band properties, high-field behavior

- Spectroscopy
  - STM/STS – ANL, Wisconsin
  - ARPES, EELS – ANL, BNL
- Raman, IR, soft x-ray
  - ANL, BNL, ORNL
- Transport
  - All
  - *But: resistivity is not as useful as for 1-band SC*
- Heat capacity
  - ANL, LANL
- Magnetic – All labs
  - NHMFL-LANL

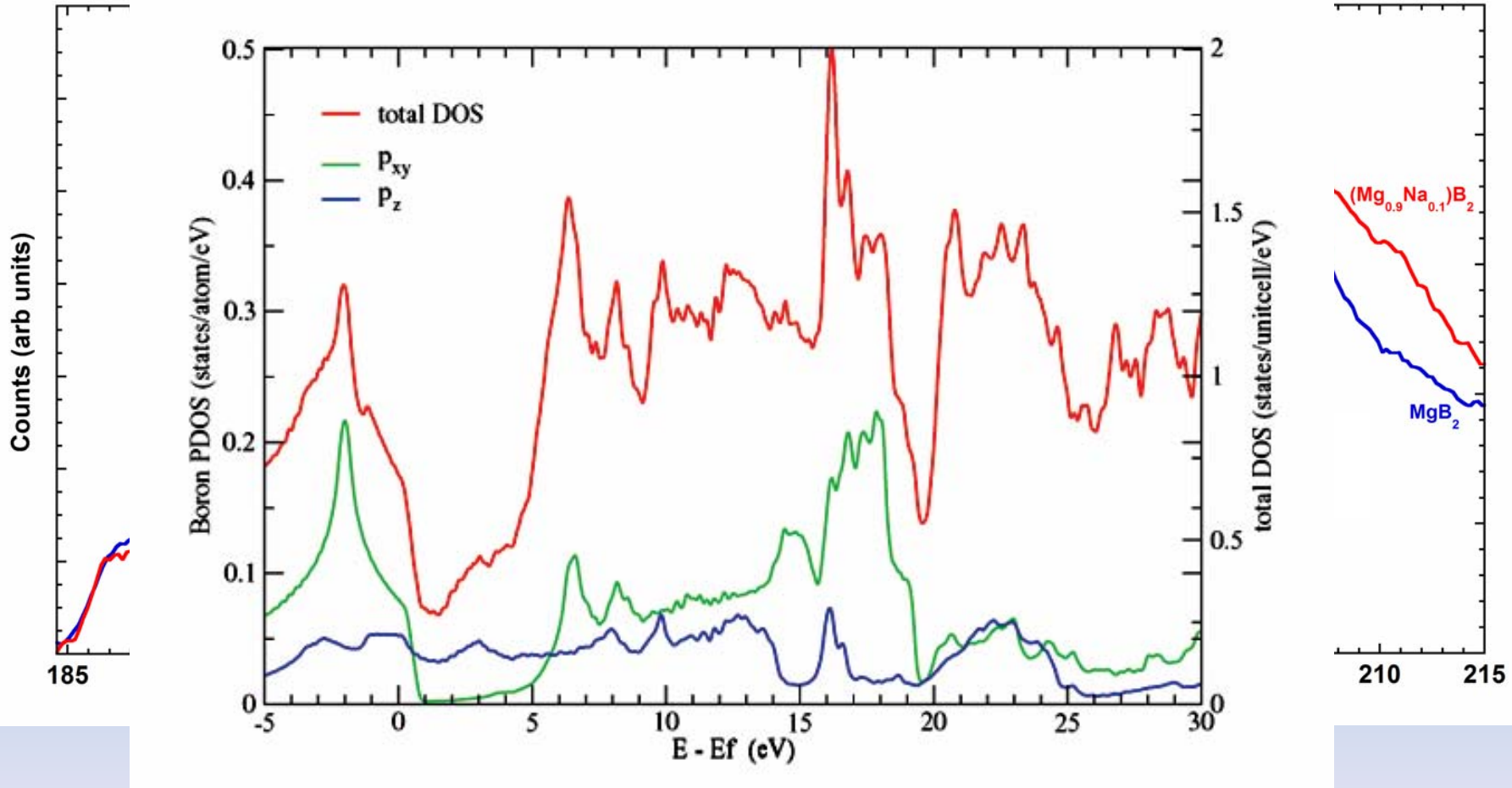
Other characterizations

- Screen boron supply
  - RBS, XRF, ICP – ASU, BNL, Ames
- Atomic-resolution structure, chemistry
  - STEM/EELS – BNL
  - HRTEM – Wisconsin, ANL, BNL
- Surface passivation
  - ASU, PSU, Wisconsin

Evidence for non-rigid-band filling

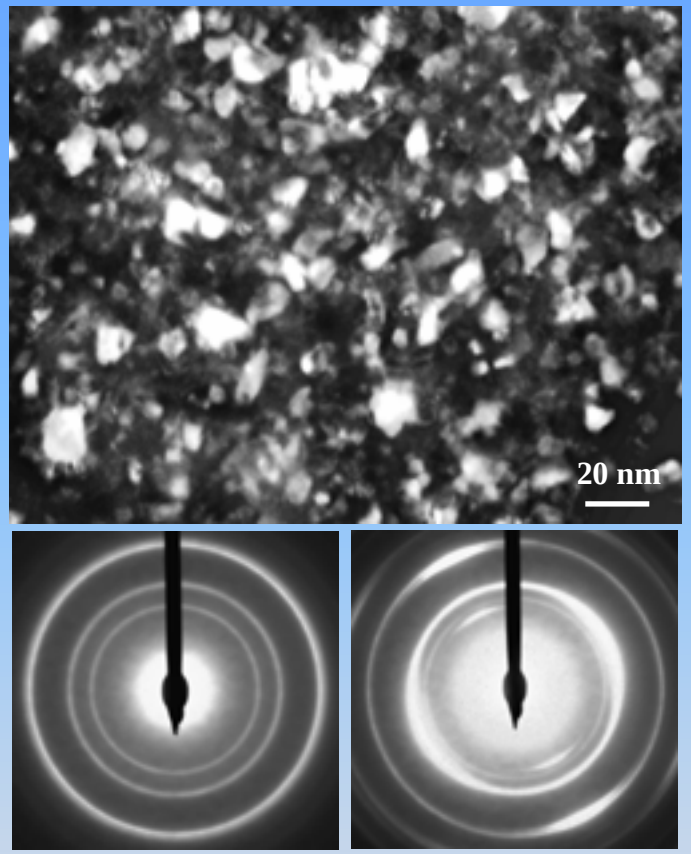
C doping adds electrons *only* to σ

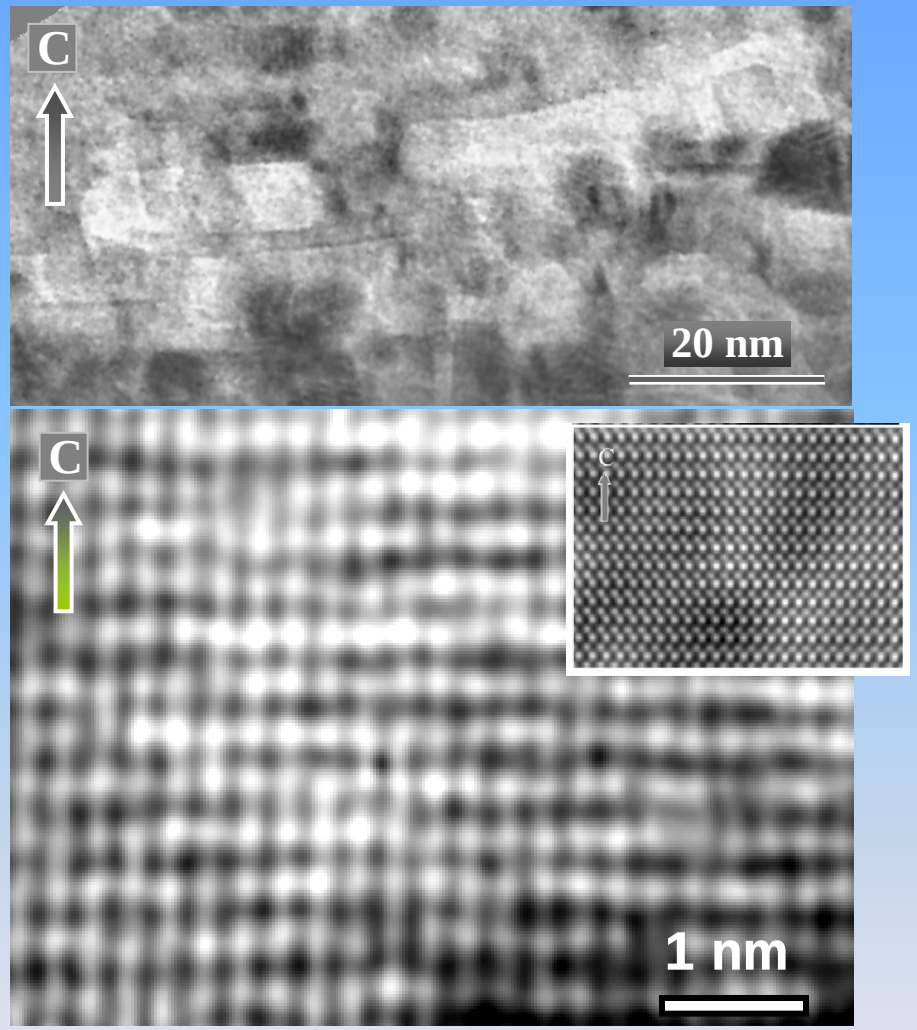
Na doping adds holes *only* to π



If carriers are added / removed, what are consequences for P_N?

High-field films are nanostructured, disordered



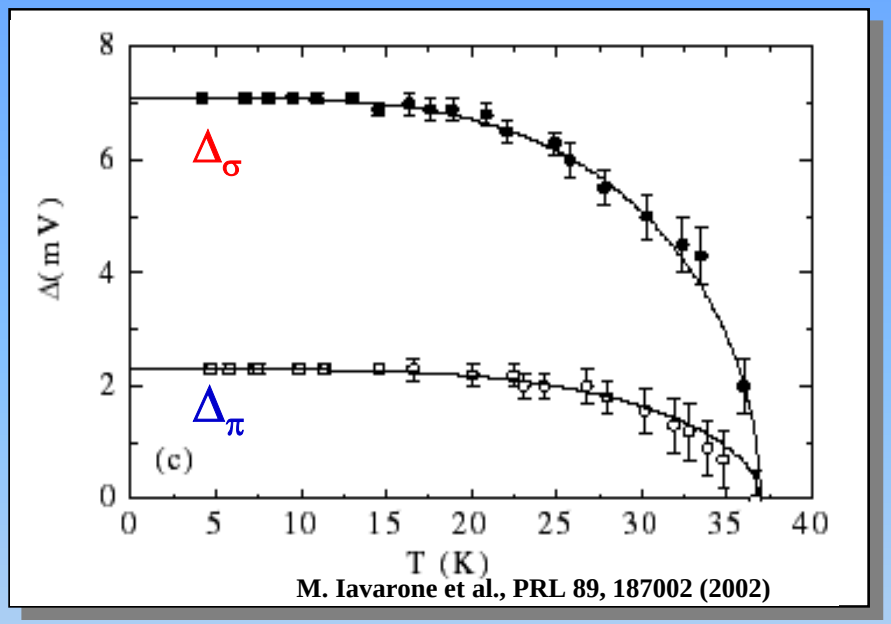


Expanded c-lattice parameter, buckling of Mg planes causes strong π scattering

Eom et al. Nature 411, 558 (2001)

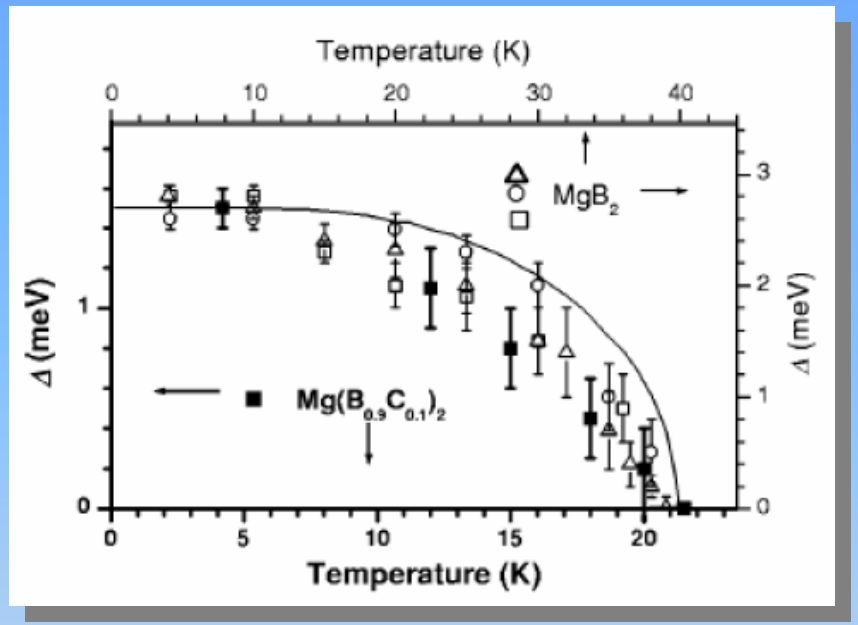
X. Song, UW

Probes of 2-gap SC

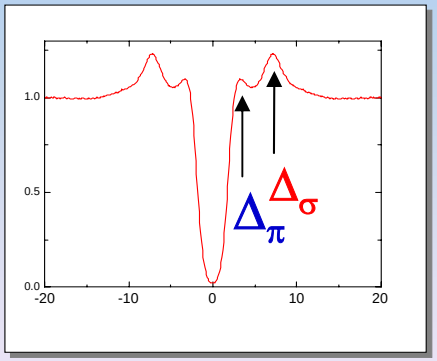


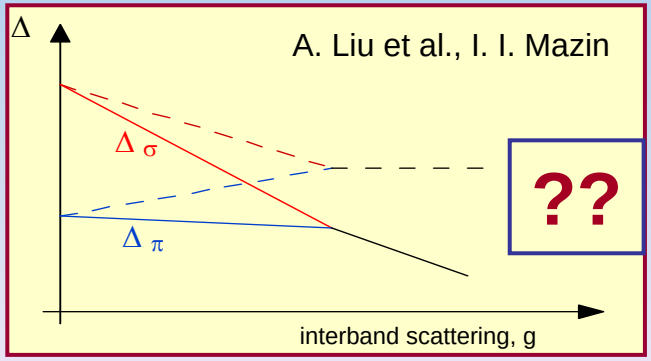
M. Iavarone et al., PRL 89, 187002 (2002)

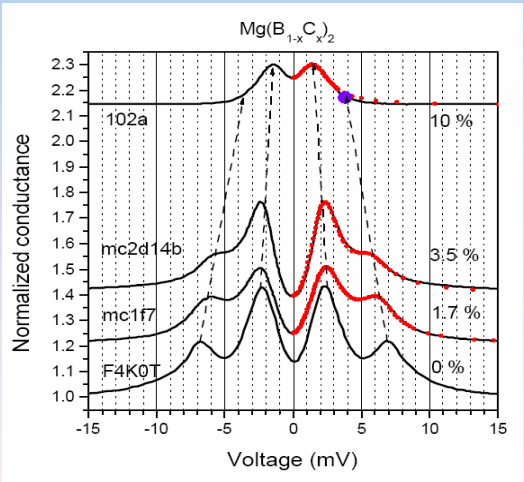
STM/STS of pure single crystals



STM/STS of C-doped bulk↑ and filament↓



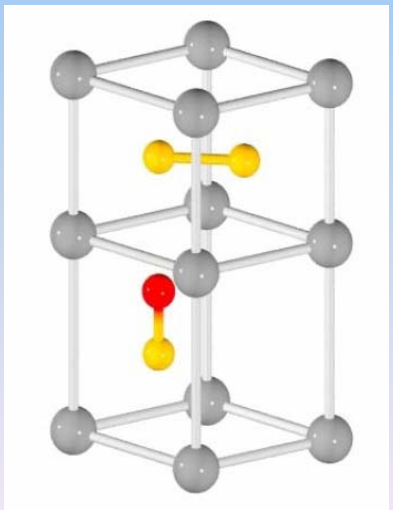


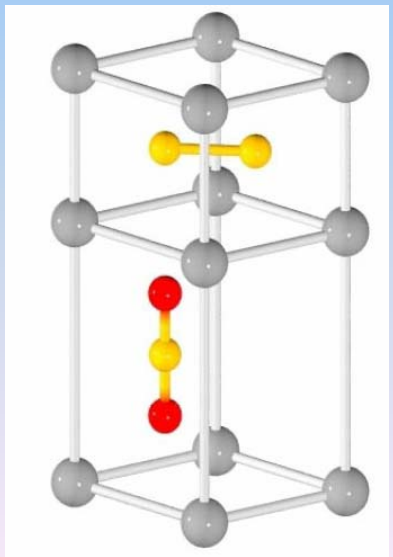


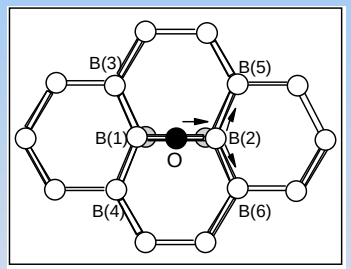
Pioneering Science and Technology

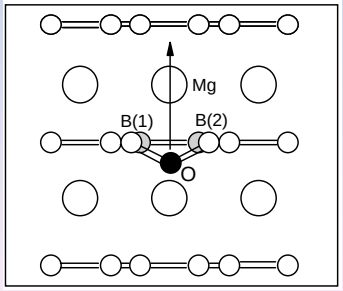
Modeling

- Further development of 2-band dirty-limit theory – ANL, Wisconsin, Ames
  - Composite vortex structures, flux pinning
- Electronic structure of doped MgB_2 –BNL, NREL, LBNL?
- Defect structures, strain – BNL, NREL
- New or related materials – INEEL

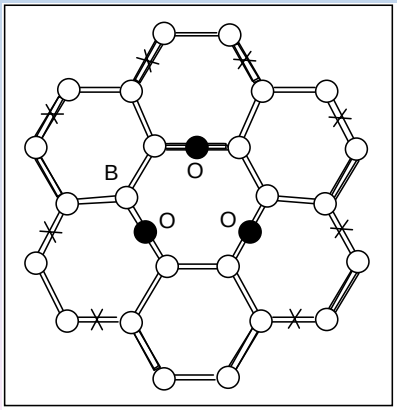








Models of oxygen interstitials – some are superconducting



Managing this CSP project

- **Steering committee**
  - Core BES labs + Wisconsin
  - Keep focus on project objectives
  - Help coordinate ideas, expertise, facilities
  - Make funding decisions; determine funding pathways
  - Identify and rank priorities
  - Make outside contacts
  - Organize workshops
    - Semi-annual meetings (committee)
    - Larger annual workshop (all)

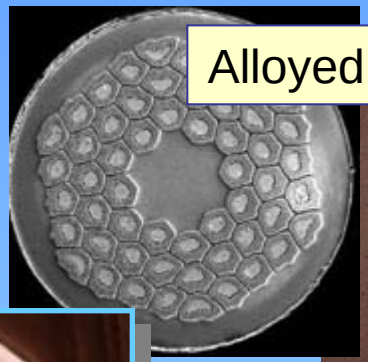
Year 1 example:

Priority – homogeneously doped single phase benchmark samples

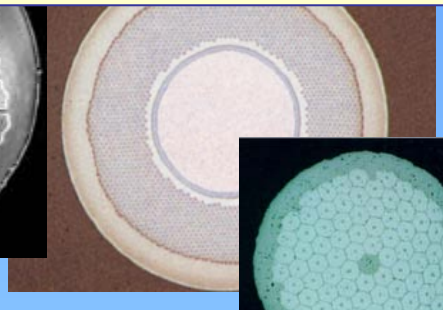
- Synthesis
  - Doped crystals ***Ames, ANL***
  - Doped thin films ***ASU, ORNL, PSU, Wisconsin, STI***
  - Doped bulk: ***Ames, BNL, Princeton, Ky, SMI***
- Characterization
  - STM/STS: ***ANL, Wisconsin***
  - H_{c2}: ***LANL-NHMFL***
  - Transport, magnetic: ***(Most labs)***
  - Calorimetry: ***ANL, LANL, UW***
  - Structure, spectroscopy: ***ANL, BNL, ORNL, Wisconsin***
- Models: ***UW, ANL, NREL, BNL***

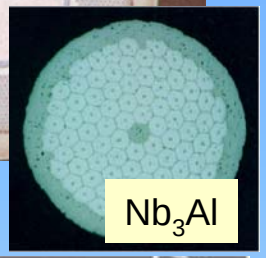
Cannot forget cost and complexity!

Nb-Ti ($150 / kg)



Alloyed Nb_3Sn ($1,060 / kg)

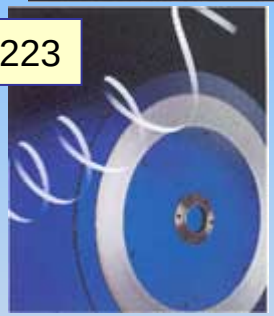




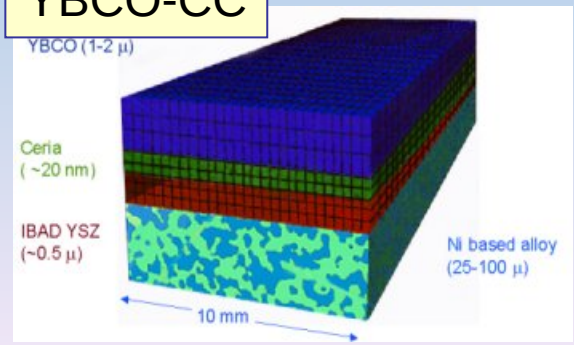
Nb_3Al

MgB_2 ($50 / kg)

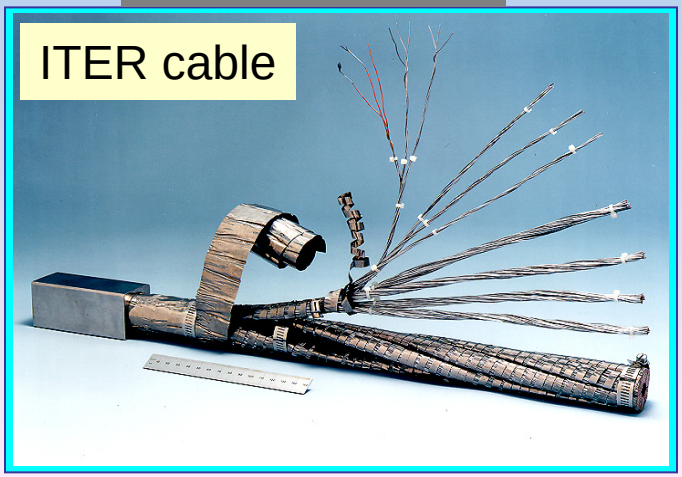
Bi-2223

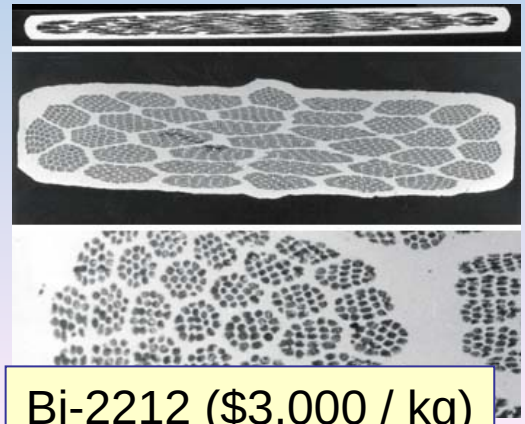


YBCO-CC



ITER cable





Bi-2212 ($3,000 / kg)

Relevance to DOE Mission and US Economy

- HFSC primarily used in medical imaging, ~$2B / year
- Electricity Transmission and Distribution:
  - Replacement of 3,500 km of underground lines at $1M / km
  - Infrastructure improvement (8/14/2003 blackout)
- High-Energy and Nuclear Physics:
  - Discovery tools, ~$0.5B annualized projects
- Fusion Energy:
  - Burning plasma science on Secy. Abraham's "little list"
  - Near term: dictated by ITER decision, $1B of HFSC
  - Far term: Burning plasma power plants work best at 20-30 K operation, $1B of HFSC *per power plant commissioned* (~$4B annual)
  - Worldwide: 1000 GW by 2050 at $1/watt

Interactions with DOE Technologies

- Industry and End-Users are integrated into project
  - Industry colleagues choose to participate at no cost to project
  - This area will be expanded
- Workshop format will follow excellent examples
  - DOE-HEP Low Temperature Superconductor Workshop
  - DOE-OETD Conductor Workshops
- Organizers of these workshops are part of this team

- OETD and others will be appraised of progress
- Attempts will be made to leverage DOE Technologies and Other Agency support
  - Attitude: “Ignition”, not “glue” money

Summary

- All of the ingredients for focused study are here
- Assembled team from 13 labs has established record of excellence
- US competence will maintain pace with Japan and EU
- Opportunities for technology transfer and program building are already being pursued
- New interactions are already forming because of planning workshops