

High- T_c superconducting materials for electric power applications

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Large-scale superconducting electric devices for power industry depend critically on wires with high critical current densities at temperatures where cryogenic losses are tolerable. This restricts choice to two high-temperature cuprate superconductors, $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ and $\text{YBa}_2\text{Cu}_3\text{O}_x$, and possibly to MgB_2 , recently discovered to superconduct at 39 K. Crystal structure and material anisotropy place fundamental restrictions on their properties, especially in polycrystalline form. So far, power applications have followed a largely empirical, twin-track approach of conductor development and construction of prototype devices. The feasibility of superconducting power cables, magnetic energy-storage devices, transformers, fault current limiters and motors, largely using $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ conductor, is proven. Widespread applications now depend significantly on cost-effective resolution of fundamental materials and fabrication issues, which control the production of low-cost, high-performance conductors of these remarkable compounds.

The potential applications of superconductors — Kamerlingh Onnes's name for materials that lose electric resistance on cooling below a transition temperature, T_c — were apparent to Onnes almost immediately. In 1913, just two years after his discovery, Onnes talked in Chicago about the design of very powerful magnets far exceeding the fields achievable by iron; these would cost as much as a battleship if made from copper and cooled with liquid air, but be affordable if made from superconducting wires. By that time he had already tested a Ni alloy coated with Pb-rich superconducting solder, but this lost superconductivity at fields of less than 50 mT. He ascribed this unexpected setback to bad places in the wire, a problem he anticipated would soon be fixed without difficulty!

In fact, applications had to wait 50 more years, particularly because the physics of superconductivity in magnetic fields were seriously misunderstood¹. This need not have been so, because London and Shubnikov made important breakthroughs in understanding the magnetic properties of superconductors in the 1930s. By careful alloying experiments, Shubnikov pointed out the vital distinction between type I superconductors, in which currents flow only at the surface and superconductivity is destroyed by weak fields, as in Onnes' 1913 experiment, and a new type of superconductor, now called a type II superconductor, capable of carrying bulk supercurrent at high fields. The key understanding that the behaviour of type II superconductors is due to quantized magnetic vortices was achieved by Abrikosov in the 1950s.

In spite of these theoretical insights, it was not until 1961 that Kunzler *et al.*² showed that high-field applications really were possible. Drawing Sn inside a Nb wire and reacting it to form the brittle intermetallic Nb_3Sn , they reached a current density $J > 10^5 \text{ A cm}^{-2}$ at 8.8 T and 4.2 K. This astounding result demonstrated that superconductivity could indeed exist in very high magnetic fields. In fact, Onnes's vision for a 10 T, high-field magnet has been abundantly fulfilled. Laboratory superconducting magnets exist by their thousands, some producing fields exceeding 20 T. Fermilab, Brookhaven, DESY and CERN all have accelerators

composed of kilometres of superconducting bending and focusing magnets (see, for example, the proceedings of the biennial Applied Superconductivity Conferences published in odd-year volumes of *IEEE Trans. Magn.* up to 1989, and *IEEE Trans. Appl. Supercond.* since 1991). The medical technique of magnetic resonance imaging was developed using very homogeneous, persistent-mode superconducting magnets, a business now exceeding US\$3 billion per year.

All of these applications are based on two low-temperature superconducting (LTS) materials, Nb-Ti alloy ($T_c = 9 \text{ K}$) or Nb_3Sn ($T_c = 18 \text{ K}$). For such applications, superconductivity is a true enabling technology and the use of liquid helium or helium-driven refrigerators is immaterial. But helium cooling tends to be too expensive for replacement of conventional electrotechnology based on Cu and Fe, two cheap and well understood materials. The first period of superconducting applications, from approximately 1962 to 1986, explored various power applications but found them too complex and expensive, thus relegating superconductivity to new high-technology and medical applications where no competitors existed. By 1986, the highest- T_c compounds possessed the A15 structure, the lightest of which, Nb_3Ge , has a T_c of 23 K (ref. 3), 5-K higher than for Nb_3Sn .

Prospects changed dramatically in late 1986, when Bednorz and Muller⁴ discovered superconductivity at 30 K in the layered cuprate, $\text{LaBa}_2\text{CuO}_{4-x}$. In early 1987, superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_x$ (YBCO) at 92 K was announced⁵, well above the boiling point of liquid nitrogen (77 K). Soon T_c rose to more than 130 K (ref. 6) in $\text{Hg}_x\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$. Widespread replacement of Cu and Fe by these new high-temperature superconductors (HTSs) was broadly discussed and significant public and private programmes to build electric utility superconducting devices were put in place in Japan, Europe and the United States⁷. Major components of the generation, transmission (power cables and devices for superconducting magnetic energy storage), distribution (transformers and fault current limiters) and end-use (motor) devices have been built, primarily using the $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Bi-2223) conductor⁷.

Superconductivity can have a significant role in deregulated electricity markets and in lessening CO_2 emissions and

other environmental impacts, but significant market penetration of HTS devices requires HTS wires that fully exploit their fundamental current-carrying capability. The announcement by Akimitsu in January 2001 that the binary compound MgB_2 superconducts at up to 39 K (ref. 8) has generated new interest in superconductors for power applications, owing to the cheap abundance of Mg and B and the potential of analogous compounds.

Conductor requirements for power technology

A conductor is more than just the superconductor. Conductors for power applications are multifilamentary wires or tapes in which many superconducting filaments are embedded in a matrix of a normal metal, such as Cu or Ag, which provides protection against magnetic flux jumps and thermal quenching⁹ (Fig. 1). Such wires must have sufficient strength to withstand the fabrication process, device winding, cool-down and electromagnetic stresses, and be capable of being made or cabled to sufficient size to carry operating currents from hundreds to thousands of amperes at costs comparable to Cu. Estimates of the acceptable cost range from about US\$10 to US\$100 per kA m, the scale being set at its lower end by the cost for Cu (~US\$15 per kA m) and at the upper end by higher-cost applications where superconductivity has advantages not possessed by present technology^{7,10}. Such higher-cost applications include high power density underground power cables in inner cities, environmentally friendly, oil-free HTS transformers, or superconducting magnetic energy-storage devices for power networks where low reactance and instantaneous redistribution of power is vital¹¹.

These requirements define a parameter set that restricts present choice to Bi-2223 or YBCO. The combination of critical current density J_c , field and operating temperature is summarized in Table 1. Target superconductor current densities must attain 10^4 – 10^5 A cm⁻² in fields of 0.1–10 T at temperatures of 20–77 K. The data in Table 1 represent an ongoing dialogue between the R&D and applied communities that tends to push up the operating temperature, current density and field so as to better separate a Cu and Fe technology limited to fields of 1–2 T from a higher-field, superconductor-based technology. Reliable and inexpensive refrigeration is a critical part of the HTS technology and significant advances have been made in this area too¹².

At present, the only HTS conductor in production is Ag-sheathed Bi-2223 with $T_c \sim 108$ K (its lower- T_c sibling $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ has only a small application in the power sector). Figure 1 shows a cross-section of a Bi-2223 conductor of the type being used for the Detroit Edison 100-MVA power cable, in which multiple strands are wound to make a fully transposed 6,000-A cable operating at 70–75 K. The individual strand is 0.2 mm by 4 mm with a critical current I_c of the order of 125 A in zero field at 77 K. Underpinning conductor R&D programmes worldwide is the conviction that such conductors have neither the magnetic field performance for applications at liquid nitrogen temperatures (~65–80 K), nor sufficiently low costs. The potential lower-cost alternative is a biaxially textured YBCO conductor or possibly MgB_2 or an analogous compound¹³. We now explore the underlying materials issues that define these beliefs.

Superconducting properties, crystal structure and anisotropy

Figure 2 presents the important magnetic field–temperature (H - T) phase diagram for the three actual (Nb-Ti, Nb_3Sn and Bi-2223) and

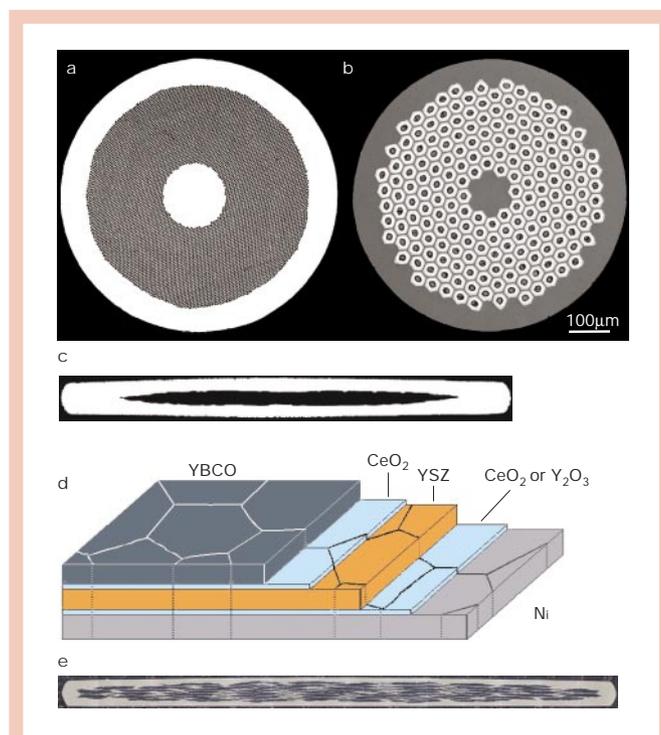


Figure 1 Conductor forms of practical superconductors. **a**, Conductor containing about 3,000 Nb47wt%Ti filaments embedded in a copper stabilizer, which protects the conductors during a transition (quench) to the normal state. The wire is ~0.8 mm in diameter and the filament diameter is 10 μm . **b**, Powder-in-tube Nb_3Sn conductor made by extruding and drawing a mixture of NbSn_2 and Cu inside the Nb tubes that form each of the 192 filaments. The conductor is ~1 mm in diameter and the filaments are ~20 μm in diameter. **c**, Cu-sheathed MgB_2 tape made by rolling MgB_2 powder in a copper tube. The tape is ~0.2 mm thick by 4 mm wide⁸⁰. **d**, Structure of a deformation-textured coated conductor. Typical thicknesses of the Ni, $\text{CeO}_2/\text{Y}_2\text{O}_3$, YSZ, CeO_2 and $\text{YBa}_2\text{Cu}_3\text{O}_x$ (YBCO) layers are (respectively) 50–100 μm , 10 nm, 300 nm, 10 nm and 0.3–2 μm . **e**, Cross-section of a silver filament ($\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Bi-2223)-power cable tape encased in a silver matrix, which acts as stabilizer. The tape is ~0.2 × 4 mm wide. All conductors except the coated conductor were made by conventional extrusion, wire drawing and/or rolling techniques.

two potential (YBCO and MgB_2) conductor materials. Their different phase diagrams result from their distinctly different physical parameters and crystal structures, as shown in Fig. 3 and Table 2. All five are type II superconductors for which bulk superconductivity exists up to an upper critical field $H_{c2}(T)$, which can exceed 100 T for Bi-2223 and YBCO. In fact, applications are limited by a lower characteristic field, the irreversibility field $H^*(T)$ at which J_c vanishes. For the isotropic cubic superconductors, Nb-Ti and Nb_3Sn , $H^*(T)$ is about $0.85H_{c2}(T)$ (refs 14,15). All three higher- T_c compounds have anisotropic layered structures, which result in significant anisotropy of the upper critical field, $\eta = H_{c2}^{\parallel}(T)/H_{c2}^{\perp}(T)$, parallel and perpendicular to the superconducting layers. Here η has values of 1 for Nb-Ti and Nb_3Sn , 2–3 for MgB_2 (refs 16,17), 5–7 for YBCO, and

Table 1 Industry consensus wire performance requirements for various utility device applications

Application	J_c (A cm ⁻²)	Field (T)	Temperature (K)	I_c (A)	Wire length (m)	Strain (%)	Bend radius (m)	Cost (US\$ per kA m)
Fault current limiter	10^4 – 10^5	0.1–3	20–77	10^3 – 10^4	1,000	0.2	0.1	10–100
Large motor	10^3	4–5	20–77	500	1,000	0.2–0.3	0.05	10
Generator	10^3	4–5	20–50	>1,000	1,000	0.2	0.1	10
SMES*	10^3	5–10	20–77	$\sim 10^4$	1,000	0.2	1	10
Transmission cable	10^4 – 10^5	<0.2	65–77	100 per strand	100	0.4	2 (cable)	10–100
Transformer	10^3	0.1–0.5	65–77	10^2 – 10^3	1,000	0.2	1	<10

*SMES, superconducting magnetic-energy storage.

Data supplied by R. Blaugher.

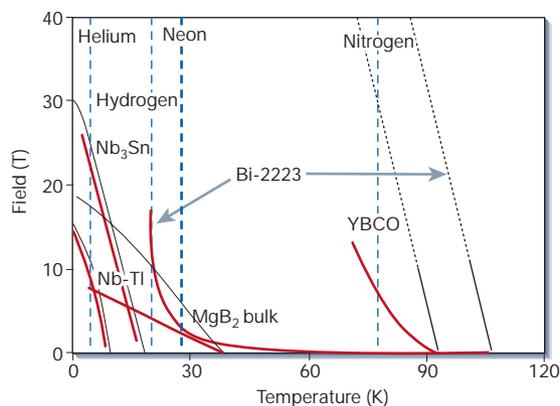


Figure 2 Magnetic field–temperature diagram for Nb47wt%Ti, Nb₃Sn, MgB₂, Bi-2223 and YBCO. The upper critical field H_{c2} at which bulk superconductivity is destroyed is indicated in black, while the irreversibility field H^* at which the bulk critical current density goes to zero is indicated in red. It can be seen that $H^*(T)$ is close to $H_{c2}(T)$ for Nb-Ti and Nb₃Sn, about half of $H_{c2}(T)$ for MgB₂, and much lower than $H^*(T)$ for YBCO and Bi-2223. There is not much scope for using Bi-2223 at 77 K because its irreversibility field (~0.3 T) is so low. As a result, applications at 77 K are restricted mostly to power cables for which the self-field is well below $H^*(77\text{K})$.

50–200 for Bi-2223 (ref. 18). In all cases, H_{c2} and H^* are lower for field applied parallel to the c -axis and perpendicular to the a - b planes. Figure 2 shows that strongly anisotropic Bi-2223 exhibits an enormous suppression of $H^*(77\text{K})$ to the very low value of ~0.2 T, well below $H_{c2}(77\text{K})$, which is of order 50 T for H parallel to the c -axis¹⁹. This low irreversibility field prevents use of Bi-2223 at 77 K in any significant field (although power cables are possible) and provides one of the key arguments for developing a second-generation HTS technology based on YBCO, for which $H^*(77\text{K}) \sim 7\text{T}$.

The structural origin of these important superconducting anisotropies is illustrated in Fig. 3 and Table 2. Nb-Ti, normally Nb47-50wt%Ti, is a body-centred cubic, solid-solution alloy with short electron mean-free-path, high resistivity and coherence length $\xi(4\text{K}) = 5\text{ nm}$. Nb₃Sn has $\xi(4\text{K}) = 3\text{ nm}$ and possesses the cubic A15 crystal structure, in which three orthogonal chains of Nb atoms separate the Sn atoms lying at cube corners and cube centre. MgB₂ has the hexagonal space group $P6/mmm$ composed of alternating B and Mg sheets, the B being nested in the Mg interstices⁸. It can form as a low-resistivity ($\rho(40\text{K}) = 0.4\ \mu\Omega\text{ cm}$), perhaps perfectly stoichiometric compound²⁰ for which $\xi(4\text{K})^{\perp} = 6.5\text{ nm}$ and $\xi(4\text{K})^{\parallel} = 2.5\text{ nm}$, where the perpendicular direction along the c -axis is taken with respect to the B planes in MgB₂ and the CuO₂ planes in HTSs. YBCO is a complex, layered perovskite centred on a Y layer, around which are stacked the CuO₂ plane of strong superconductivity and a double charge-reservoir layer of O-Ba-O and O-Cu. In Bi-2223, the charge-reservoir layer consists of a double (Bi,Pb)-O and its neighbouring Sr-O layers, the superconducting layers being a stack of three CuO₂ layers interleaved by two Ca layers. The anisotropy of Bi-2223 is so

large that $H^*(77\text{K})$ is only 0.2–0.3 T, restricting 77-K applications to low self-field uses such as power cables, even though $H_{c2}(77\text{K})$ exceeds 10 T.

Flux pinning and the critical current density

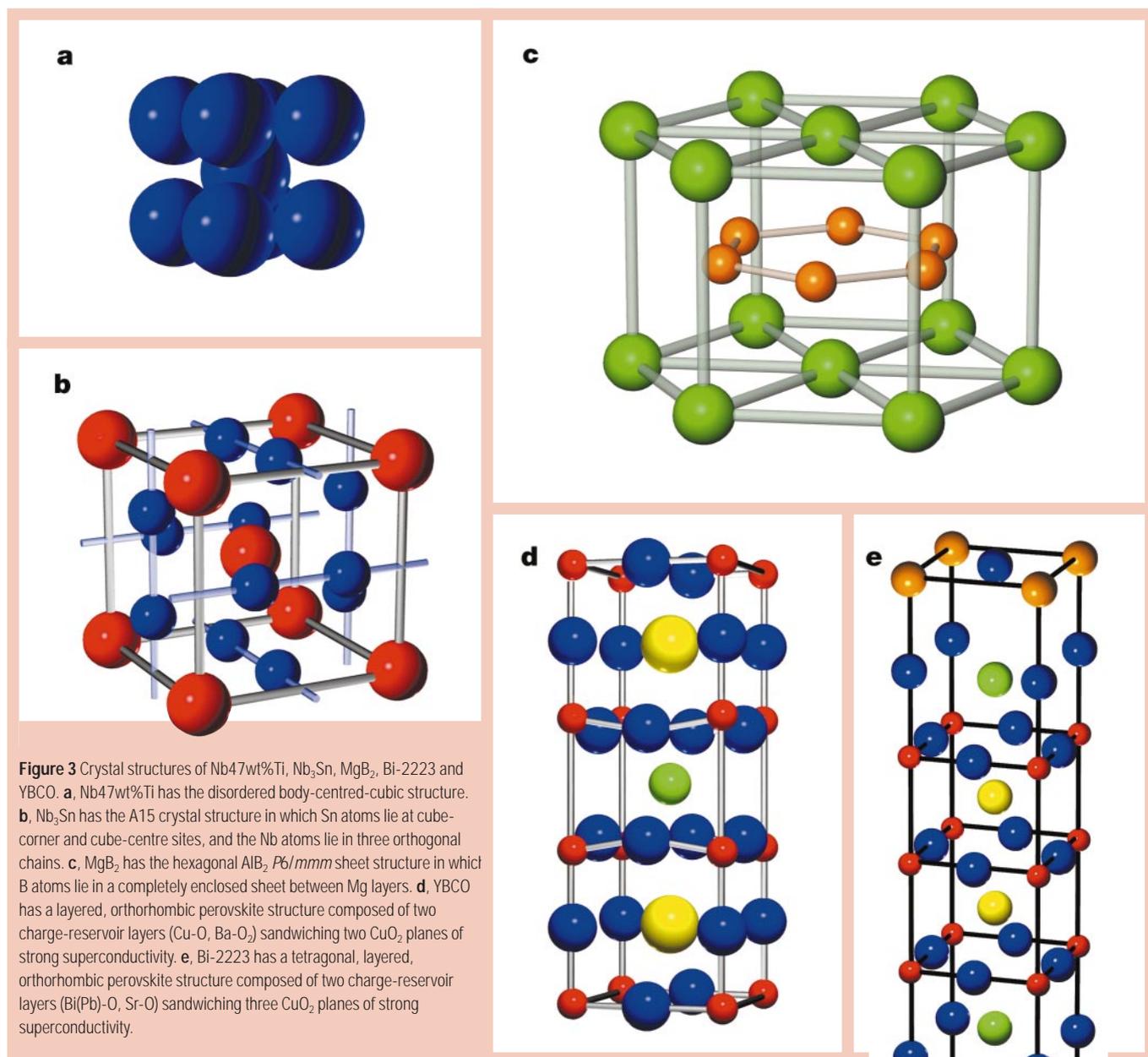
Under equilibrium conditions, magnetic flux penetrates the bulk of a type II superconductor above the lower critical field H_{c1} , which is of order 10–20 mT for the materials under consideration. Over most of the available H - T space, $H > H_{c1}$, this magnetic flux exists as a lattice of quantized line vortices or fluxons^{18,21}. Each fluxon is a tube of radius of the London penetration depth $\lambda(T)$, in which superconducting screening currents circulate around a small non-superconducting core of radius $\xi(T)$, where $\xi(T)$ is the superconducting coherence length. The flux carried by the screening currents in each fluxon equals the flux quantum $\phi_0 = 2 \times 10^{-15}\text{ Wb}$. Bulk superconductivity is destroyed when the normal cores overlap at the upper critical field, $H_{c2}(T) = \phi_0 / 2\pi\mu_0\xi(T)^2$. In isotropic materials such as Nb-Ti and Nb₃Sn, vortex lines are continuous, but the weak superconductivity of the blocking layers of HTS compounds produces a stack of weakly coupled ‘pancake’ vortices whose circulating screening currents are mostly confined within the superconducting CuO₂ planes¹⁸.

Superconductors can carry bulk current density only if there is a macroscopic fluxon density gradient²², defined by the Maxwell equation $\nabla \times \mathbf{B} = \mu_0\mathbf{J}$. This gradient can be sustained only by pinning the vortices (flux pinning) at microstructural defects. Increasing T and H weaken the potential wells at which vortices are pinned, thus lessening $H^*(T)$ and $J_c(T, H)$. Flux pinning is determined by spatial perturbations of the free energy of the vortex lines due to local interactions of their normal cores and screening currents with these microstructural imperfections²². In addition, the fluxon structure is subjected to the Lorentz force $\mathbf{F}_L = \mathbf{J} \times \mathbf{B}$ of the macroscopic current. The critical current density $J_c(T, H)$ is then defined by the balance of the pinning and Lorentz forces, $F_p = F_L$, where F_p is the volume summation over all microstructural pinning defects in the strongly interacting network of flux lines. Ideally, a type II superconductor can carry any non-dissipative current density J smaller than J_c . When J exceeds J_c , a superconductor switches into a dissipative, vortex-flow state driven by the Lorentz force.

This description of flux pinning immediately suggests tailoring the defect structure of the conductor to maximize J_c . In fact, this does not correspond well to the essentially empirical way in which conductors have been developed, especially those of Bi-2223 and YBCO, for which the important flux-pinning defects are largely unknown. In practice, it is the critical current I_c of a wire of cross-section A that is measured, normally at the finite electric field of $1\ \mu\text{V cm}^{-1}$. The physics of vortex dynamics and pinning is enormously interesting^{18,22}, but equally important in practical terms is the fact that the actual cross-section over which transport currents flow in HTS conductors is much less than the whole²³. What has greatly held back the development of Bi-2223 conductors in particular is uncertainty about whether the long-range utilization of the current-carrying cross-section of state-of-the-art conductors is 5, 25 or 50%. It is certain that it is not 100%, for reasons discussed later. Irrespective of such percolative current-flow effects, the upper limit to the critical current is set by

Table 2 Basic material and critical current density relevant parameters for practical superconductors

Material	Crystal structure	Anisotropy	T_c (K)	H_{c2}	H^*	In-plane coherence length $\xi(0)$ (nm)	In-plane penetration depth $\lambda(0)$ (nm)	Depairing current density (A cm ⁻²), 4.2 K	Critical current density (A cm ⁻²)	$\rho(T_c)$ ($\mu\Omega\text{ cm}$)
Nb47wt%Ti	Body-centred cubic	Negligible	9	12 T (4 K)	10.5 T (4 K)	4	240	3.6×10^7	4×10^8 (5 T)	60
Nb ₃ Sn	A15 cubic	Negligible	18	27 T (4 K)	24 T (4 K)	3	65	7.7×10^8	~10 ⁶	5
MgB ₂	$P6/mmm$ hexagonal	2–2.7	39	15 T (4 K)	8 T (4 K)	6.5	140	7.7×10^7	~10 ⁶	0.4
YBCO	Orthorhombic layered perovskite	7	92	>100 T (4 K)	5–7 T (77 K)	1.5	150	3×10^8	~10 ⁷	~40–60
Bi-2223	Tetragonal layered perovskite	~50–100	108	>100 T (4 K)	~0.2 T (77 K)	1.5	150	3×10^8	~10 ⁶	~150–800



the flux-pinning current density, which cannot exceed the depairing current density $J_d = \phi_0 / [3/(\sqrt{3})\pi\mu_0\lambda^2(T)\xi(T)]$, the maximum supercurrent density circulating near the vortex cores.

The best information that we possess about the limits to flux pinning in practical superconductors has come from extensive studies of Nb-Ti²⁴. Exceptionally strong pinning by a dense, ~20–25-vol% lamellar structure of normal (that is, non-superconducting) α -Ti ribbons, about 1 nm (0.2ξ) thick and aligned parallel to the wire transport current, can produce a J_c that approaches 5–10% of J_d at zero field and 4.2 K. The flux-pinning mechanism is also known for Nb₃Sn, in which J_c is determined by the magnetic interaction of the fluxon currents with grain boundaries^{25,26}. In this case, $J_c(T, H)$ increases with decreasing grain size, thus encouraging low-temperature methods of phase formation that produce a fine grain size²⁷. There are also indications of grain-boundary pinning in MgB₂ (refs 28–30). All three materials possess zero-field J_c values, which can exceed 1 MA cm⁻² at 4.2 K.

The important flux-pinning interactions in conductor forms of YBCO and Bi-2223 are largely unknown, partly because the coherence length is so small that even atomic-sized point defects can pin fluxons. The critical temperature of HTSs is extremely sensitive to the carrier

(hole) density, which is in turn determined by local oxygen (and other) non-stoichiometries, so that even a weak hole-depletion at crystalline defects can locally drive an HTS into an antiferromagnetic insulator. The proximity of the superconducting state to the metal-insulator transition, the d-wave pairing symmetry, the very short in-plane coherence length ($\xi(0) = 1.5\text{--}2$ nm) and the much longer Debye screening length ($l_D \sim \xi(0)$) all combine to produce significant suppression of the superconducting order parameter $\Delta(\mathbf{r})$ near crystal defects such as impurities, dislocations and grain boundaries³¹, thus making them effective pinning centres. In fact, recent scanning tunnelling microscopy of Bi-2212 single crystals³² has revealed significant variations of $\Delta(\mathbf{r})$ near single-atom impurities (Zn and Ni), as well as striking intrinsic variations of $\Delta(\mathbf{r})$ on scales of 1–2 nm even in pure crystals³³.

This extreme sensitivity of HTSs to nanoscale defects, along with the intrinsic spatial fluctuations of $\Delta(\mathbf{r})$ from 25 meV to 60 meV on the scale of ξ or less³³, provide effective core pinning of vortices. There is strong experimental evidence that point defects such as Zn and Ni impurities³² or oxygen vacancies³⁴, or line defects such as edge³⁵ and

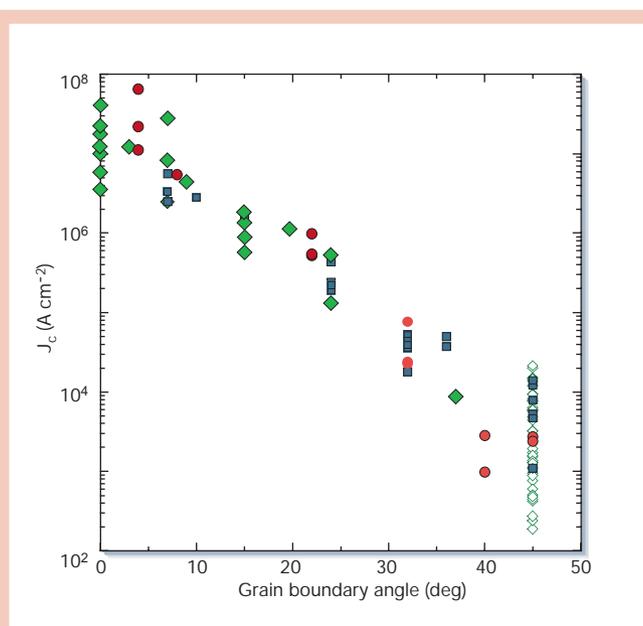


Figure 4 Transport critical current density at 4.2 K measured in thin films of YBCO grown on [001] tilt bicrystal substrates of SrTiO₃ (squares³⁷ and filled diamonds⁴⁴), Y₂O₃-stabilized ZrO₂ (circles⁹⁷), and bi-epitaxial junctions (open diamonds⁹⁶) of varying misorientation angle θ . Data of refs 97 and 44 were taken at 77 K and have been multiplied by a factor of 10.9 to make them comparable with the data at 4.2 K. Despite a significant scattering, $J_c(\theta)$ exhibits a universal exponential dependence on θ . Data compilation courtesy of J. Mannhart³⁷.

screw³⁶ dislocations, can effectively pin pancake vortices in layered HTSs. Thus, all forms of HTSs possess crystalline defects of different kinds, which provide strong flux pinning at low temperatures. Consistent with this, YBCO and Bi-2223 films^{37,38} can exhibit $J_c(4K)$ values of more than 10 MA cm⁻² and single crystals of YBCO (ref. 39) can exhibit values up to 1 MA cm⁻². Although these J_c values are about an order of magnitude lower at 77 K, they are more than sufficient for power applications (Table 1). Moreover, they are still much lower than the depairing limit: $J_d \approx 300$ MA cm⁻² at 4 K for characteristic values $\lambda(0) \approx 150$ nm and $\xi(0) \approx 1.5$ nm. Thus, unlike Nb-Ti where the material is rather close to its performance limit, there is enormous untapped potential for improved HTS material performance.

The main obstacle for HTS conductors is that their current-carrying capability deteriorates rapidly with increasing temperature and magnetic field. As shown in Fig. 2, J_c vanishes at the irreversibility field $H^*(T)$, which is far below $H_{c2}(T)$ at 77 K. An independent, significant limit on J_c is current blockage by grain boundaries, as described in the next section. But the underlying cause of the suppression of $H^*(T)$ at higher temperatures is due to the layered structure of HTSs, which greatly facilitates depinning of stacks of weakly coupled pancake vortices by thermal fluctuations^{15,18}. For $H < H^*(T)$, vortex fluctuations cause thermally activated creep of magnetic flux⁴⁰, producing measurable dissipation well below J_c defined at $1 \mu\text{V cm}^{-1}$. Anisotropy strongly enhances the effect of thermal vortex fluctuations¹⁸, thus suppressing H^* and enhancing flux creep in Bi-2223 much more than in YBCO. Both H^* and J_c can be significantly improved by irradiation, which creates effective columnar pinning tracks⁴¹. For example, neutron irradiation of U-doped Bi-2223 tapes fissions the U and produces heavy-ion tracks that raise $H^*(77K)$ to over 1 T, while also reducing the anisotropy of J_c (ref. 42).

Grain boundaries

Large devices need kilometres of polycrystalline conductors and the influence of grain boundaries is therefore critical. The most unambiguous evidence that grain boundaries are strong barriers to current

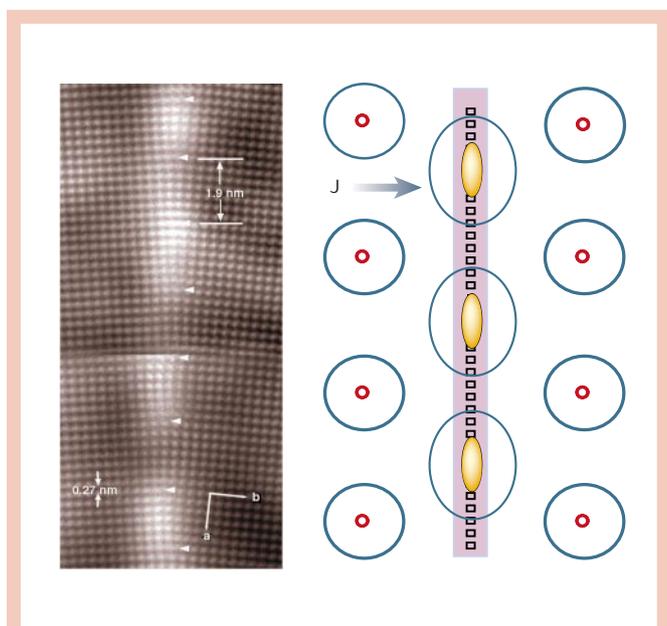


Figure 5 Grain boundary structure and its effect on vortex properties. The left panel is a Fourier-filtered transmission electron micrograph of an 8° [001] tilt Bi₂Sr₂CaCu₂O_x bicrystal, showing the edge dislocation structure at the grain boundary and the channels of good conduction that lie between their cores. Stacking faults are indicated by the white regions between alternate pairs of dislocations. Strong strains are found at the core of each dislocation. On the right is a schematic illustration of the consequences of the dislocation structure for vortices at the boundary. Abrikosov vortices are indicated within the grains, but their cores become elongated when they sit at the boundary, owing to the reduced current transparency of the boundary. The grain-boundary dislocations are indicated by the black squares and the carrier-depleted zone by the pink-shaded region. Vortices at the grain boundary acquire a mixed Abrikosov–Josephson character, which strongly reduces their pinning force along the boundary.

flow in HTSs comes from detailed studies of [001] tilt YBCO bicrystals on SrTiO₃ or Y₂O₃-stabilized ZrO₂ (YSZ) substrates. These have shown that the critical current density J_b across the grain boundary drops exponentially below that of the grains, $J_b = J_0 \exp(-\theta/\theta_c)$, as a function of the misorientation angle θ between the neighbouring crystallites^{37,43–45}, where $\theta_c \approx 2–5^\circ$, depending on the value of the intragrain J_c (Fig. 4). This extreme sensitivity to misorientation, coupled with the intrinsic anisotropy of the HTS compounds, dictates the need to texture conductors into the tape forms shown in Fig. 1, so as to shift the grain-boundary misorientation distribution to as small a value of θ as possible. For Bi-2223, the rolling deformation used to make the tape produces a marked uniaxial, c -axis texture, whereas for YBCO-coated conductors, the essential goal is to make both in- and out-of-plane texture so that the tape behaves as a quasi-single crystal, even though the YBCO grain size is $< 1 \mu\text{m}$ in diameter^{46,47}.

This behaviour of HTS materials is in strong contrast to metallic LTS and MgB₂, in which grain boundaries are not only transparent to current, but also significantly contribute to flux pinning, increasing the overall J_c as the grain size decreases^{26,28,48}. Although Table 2 might suggest that HTS grain boundaries are weak links because of their short coherence length, in fact a value of $\xi(77K)$ of ~ 4 nm for YBCO is actually longer than for Nb₃Sn at 4 K. Moreover, low carrier density makes grain boundaries in (Ba,Pb)BiO₃ also weak-linked, even though $\xi(4K) \sim 7$ nm for this compound^{49,50}. In fact, the weak-link (that is, current-obstructing) behaviour of grain boundaries in HTSs results from the same factors, which otherwise enhance the pinning of vortices by point defects, namely the strong dependence of T_c on hole concentration and low carrier density. A tilt grain boundary is built of a chain of edge dislocations spaced by $d = b/2\sin(\theta/2)$, where

$b \approx 0.4$ nm is the Burgers vector. As the misorientation angle θ increases, the spacing between the insulating dislocation cores decreases, becoming of the order of the coherence length ξ at $\theta \approx 5\text{--}7^\circ$. For higher angles, a grain boundary becomes a Josephson weak link, because of the local suppression of the order parameter in the current channels between the dislocations, and J_b decreases exponentially with increasing θ ⁵¹. Extensive electron microscopy has established that the hole content is depressed in the vicinity of grain boundaries and that there is only partial occupancy of cations in the structural units that form the boundary^{51,52}.

Grain boundaries possess extra ionic charge, whose magnitude increases with increasing θ and causes hole depletion in a layer of thickness given by the Thomas-Fermi screening length $l_D \approx \xi(0)$ (Fig. 5). As a result, the grain boundary is in a locally underdoped state, the local order parameter suppression increasing as θ increases^{31,52}. That is why local overdoping by partially substituting Ca for Y in YBCO (refs 53,54) is effective. Adding extra carriers to the grain boundary with Ca-doping and reducing local strains can significantly increase J_b (by a factor of eight for a 24° [001] tilt boundary at 4.2 K and 0 T (ref. 53), and by a factor of three for a 7° -bicrystal at 55 K and 3 T (ref. 55)). Although overdoping ameliorates, rather than removes, the problem of current obstruction by grain boundaries in HTSs, it may certainly be useful for conductors^{43–45,56,57}, because the critical angle θ_c is only $2\text{--}5^\circ$.

Performance of HTS conductors in a magnetic field is determined by the nature of the vortices at the grain boundaries and their pinning interaction with structural defects, and by their magnetic interactions with strongly pinned vortices within the grains (Fig. 5). Because of the extended core of grain-boundary vortices, they are generally pinned more weakly than vortices in the grains (ref. 58, and Gurevich *et al.*, submitted), and grain boundaries become barriers to current flow because their critical current density $J_b(T,H)$ is smaller than $J_c(T,H)$ in the grains. This behaviour is clearly visible in the magneto-optical images of Fig. 6, which shows preferential flux penetration along a network of coated-conductor grain boundaries⁵⁶ having $\theta > 4^\circ$.

The influence of misorientation is less certain for compounds other than YBCO, as all other HTS compounds are harder to grow as good films on bicrystal substrates. To first approximation, thin films of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ and Bi-2223 show similar exponential decrease of $J_b(\theta)$ (refs 59,60) to that seen for YBCO, suggesting a common mechanism for suppression of critical current on grain boundaries. Experiments on float-zone $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ bicrystals indicate much weaker dependence of J_b on θ than for thin films⁶¹, but the intragranular critical current densities are so small that they might not reach the critical current density of the grain boundaries. An extensive magneto-optical study of [001] tilt, thin-film YBCO bicrystals⁶² has shown that the temperature dependences of J_c and J_b are significantly different.

Current percolation in polycrystals

Any conductor must be polycrystalline. All conductor materials except Nb-Ti are brittle, making cracks endemic in Nb_3Sn , MgB_2 , Bi-2223 and YBCO. In addition, the last three compounds all are prone to porosity. The net result is that current percolates through a polycrystalline network containing many obstructions, some of which partially block the current while others result in a total block²³. Thus the local fraction of current-carrying cross-section is significantly less than unity. Magneto-optical imaging is an effective method of visualizing non-uniformities of current flow in polycrystalline conductor forms, as shown in Fig. 7 for MgB_2 , Bi-2223 and YBCO. These images show the 'roof' pattern of the perpendicular magnetic field, resulting from screening currents induced by an external field. For MgB_2 the finest, visible-scale variation seems to be controlled by $\sim 10\%$ porosity⁶³ and not by grain boundaries^{17,20,64}. For the Bi-2223 tape, a transverse feathering of the image is caused principally by quasiperiodic fluctuations of the superconductor

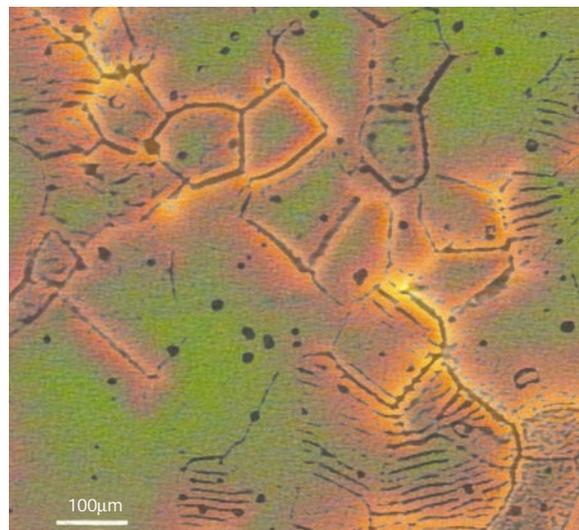


Figure 6 Magneto-optical image of the flux penetrating into a typical deformation-textured YBCO coated conductor overlaid on a light-microscope image of the underlying Ni substrate. Darker (green) areas are regions that are well connected electromagnetically, whereas the lighter (orange) flux network indicates where the local current density is reduced. Some grain boundaries in the Ni substrate (black) do not appear in the magneto-optical image. However, all grain boundaries with misorientations greater than 4° do appear⁵⁶.

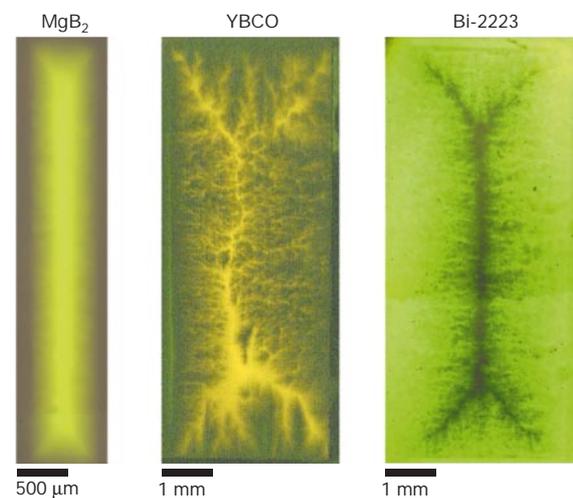
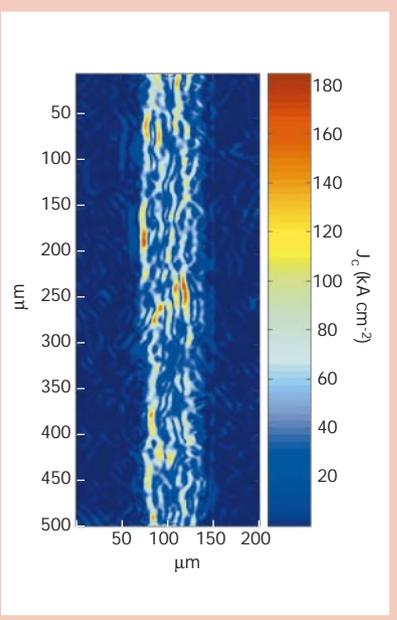


Figure 7 Magneto-optical images of (left to right) a sintered MgB_2 slab⁶³, a YBCO IBAD-coated conductor, and a Bi-2223 monocore tape²³. Each tape shows a global roof-top pattern of the perpendicular component of magnetic field above the superconductor surface. This 'roof-top' pattern arises from induced magnetization currents circulating through the entire sample. The MgB_2 slab shows the most uniform current flow, even though it is imaged in an applied field of zero after cooling in 120 mT to 37 K, only 2 K below T_c . Almost no spatial variation of J_c is visible. The coated conductor was imaged after field-cooling in 60 mT to 11 K, well below its T_c of 90 K, then reducing the applied field to zero. The Bi-2223 tape was imaged at 11 K after cooling without field and then applying a field of 120 mT. Some feathering of the image transverse to the tape length indicates a longitudinal variation of the current, probably associated with rolling defects.

Figure 8 Colour map of the spatial distribution of the local critical current density in the same monocore Bi-2223 tape imaged in Fig. 7. The tape was imaged in a slab geometry produced by sectioning the tape along a centre line parallel to the tape length. The image from which the current reconstruction was performed was made by cooling the slab to 77 K in zero field, then applying a field of 36 mT parallel to the half width of the slab. The transport critical current density $J_c(77\text{K},0\text{T})$ of the tape was 35 kA cm^{-2} , about the median of the distribution seen in the J_c map. Peak values of the J_c exceeded 180 kA cm^{-2} .



thickness and residual rolling damage²³. For the YBCO-coated conductor, there are multiple lines emanating from the central roof pattern, indicating local, higher current density loops superimposed upon the long-range current. Thus magneto-optical examination shows explicitly that there are multiple scales of current loops flowing in polycrystalline HTS samples. Such images pose the question: by how much is the average current density — defined by $J_c = I_c/A$, where A is the total cross-section, rather than the active cross-section $A_{\text{effective}}$ — reduced below the flux-pinning critical current density established within each grain?

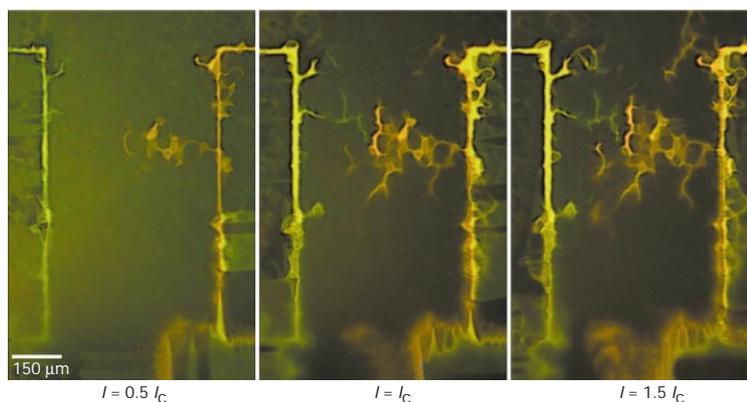
To answer such a question requires good understanding of the intragrain critical current densities. In fact, the magnitude of the intragrain J_c of Bi-2223 remains unclear, because few high-quality bulk or thin-film single crystals have been made. Several thin-film results^{38,60} suggest that $J_c(77\text{K},0\text{T})$ can achieve 1 MA cm^{-2} , whereas a single-crystal study⁶⁵ gave J_c values significantly less than 10^5 A cm^{-2} . At 77 K and 0 T, Bi-2223 conductors can achieve up to $\sim 75\text{ kA cm}^{-2}$ for small current conductors⁶⁶ and $40\text{--}50\text{ kA cm}^{-2}$ for high current conductors having $I_c \sim 150\text{ A}$ (refs 19,67). Figure 8 shows the results of a recent magneto-optical current reconstruction²³ on a high- J_c monofilament conductor with $J_c(77\text{K},0\text{T}) = 35\text{ kA cm}^{-2}$. Remarkably, J_c can achieve local values of 180 kA cm^{-2} , up to five times higher

than the transport J_c . Although there is a tendency for the highest- J_c regions to be located at the Ag–superconductor interface, in fact very-high- J_c regions are located throughout the tape. Such local variability is not surprising for a seven-component system, because it is hard to control the phase-conversion reaction of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ and other constituents to Bi-2223. Images such as Fig. 8 offer the possibility of directly correlating the local microstructure to the local J_c . It is interesting to note that regions of J_c exceeding 100 kA cm^{-2} are $50\text{--}100\text{ }\mu\text{m}$ long, several times the $\sim 20\text{-}\mu\text{m}$ grain length. Although earlier work⁶⁸ suggested that high- J_c regions were confined mainly to the Ag interface region, more recent results show that better processing is producing much more uniform J_c distribution across Bi-2223/Ag tapes, resulting in significant gains in tape performance.

State-of-the-art Bi-2223 conductors contain grains $\sim 20\text{ }\mu\text{m}$ long by $\sim 1\text{ }\mu\text{m}$ thick, distributed in $30\text{--}60$ filaments, each of which is of the order of $5\text{--}10\text{ }\mu\text{m}$ thick by $200\text{--}300\text{ }\mu\text{m}$ wide. The current percolates in a three-dimensional manner through a polycrystalline array, which possesses a significant c -axis texture, of the order of $10\text{--}12^\circ$ full-width at half-maximum (FWHM), but has little in-plane texture. By contrast, YBCO-coated conductors rely on epitaxy and are controlled mostly by the two-dimensional percolation^{69,70}. Figure 9 images the local field produced by current flowing along a track in a representative coated conductor. An extended planar array of misoriented grains obstructs the current, forcing it to flow non-uniformly and causing local dissipation, even at $0.5I_c$, where, as usual, I_c is defined at the electric field criterion of $1\text{ }\mu\text{V cm}^{-1}$ established across the whole track. As I increases, the current-obstructing network expands, eventually becoming continuous across the whole conductor. Such images show that I_c is determined locally, with many regions of the sample showing no signs of dissipation even at $I = 1.5I_c$. A similar conductor had a global $J_c(77\text{K},0\text{T})$ of 1.2 MA cm^{-2} , but individual clusters of YBCO grains had J_c values⁵⁷ exceeding 5 MA cm^{-2} . Thus, even in the biaxially textured coated conductors for which both in- and out-of-plane texture FWHM values are significantly below 10° , there is abundant evidence for current percolation around a network of obstructing grain boundaries and other defects.

Present $J_c(77\text{K},0\text{T})$ values in 1–2-cm-long coated conductor prototypes^{71–74} typically fall in the range $1\text{--}3\text{ MA cm}^{-2}$. Further performance improvement may be impeded by electromagnetic and thermal instabilities, such as flux jumping, hot-spot formation and quench propagation, the same factors which limit the current-carrying capability of low- T_c superconductors^{9,75}. Whatever future microstructure improvements occur, current-obstructing obstacles are inevitable in any practical long-length forms. Because the electromagnetic and thermal stability of the current-carrying state is influenced by hot spots near obstacles, the architecture of coated

Figure 9 Magneto-optical images at 77 K of the self-field produced by an applied transport current in a RABiTS coated conductor sample for which the full-width $J_c(77\text{K},0\text{T})$ was 0.7 MA cm^{-2} . A laser was used to cut a link restricting the current flow to a region $0.5 \times 1.1\text{ mm}$, which had a $J_c(77\text{K},0\text{T})$ of 0.6 MA cm^{-2} . Images shown are at applied currents for the link of (left to right) $0.5I_c$, I_c , and $1.5I_c$, where I_c is defined at $1\text{ }\mu\text{V cm}^{-1}$. A cluster of YBCO grain boundaries is visible in all the images, indicating where in the link J_c is limited well before the onset of bulk dissipation. As the current is increased, percolation near the visible cluster of grain boundaries becomes more pronounced, but above and below this cluster current flows more uniformly, and preferentially near the edges of the link. Even at $1.5I_c$, much of the link is supporting a current less than its local J_c . Patterning of bridges has shown that the intragrain J_c can reach 5 MA cm^{-2} , several times the J_c of this bridge. It is clear that current is percolating through a network of grain boundaries, which also result in localized dissipation in the track.



conductors needs to be optimized so as to reduce local dissipation by current redistribution to the Ag stabilizer, and enhance heat transfer from the YBCO to the coolant. An important question is what is the acceptable size and concentration of obstructive hot spots, which provides stable conductor operation.

Because obstructions initiate dissipation, the spatial distribution of the electric field $E(x,y)$ near such obstructions controls the J_c of the conductor. But unlike current distributions, which can be extracted from magneto-optical images, the electric field distribution is much harder to measure. Recently, analytical methods for the calculation of $E(x,y)$, taking account of the highly nonlinear E - J characteristic of a superconductor, have been developed^{76,77}. For $J < J_c$, the E - J relation is determined by thermally activated flux creep, which is conventionally approximated as $E = E_c (J/J_c)^n$, where $E_c \sim 1 \mu\text{V cm}^{-1}$, and the exponent $n(T,H)$ varies from 20–30 for $H \ll H^*$ to $n \approx 1$ at $H = H^*$. Calculations for a planar defect perpendicular to current flow show that the nonlinearity of $E(J)$ strongly enhances the electric field in extended regions of length $L_\perp \sim na$ perpendicular to the current, on a scale much larger than the defect size a . This behaviour is in stark contrast to ohmic conductors, in which current flow becomes essentially uniform at a distance $\sim a$ away from the defect. These long-range disturbances of $E(x,y)$ become particularly important in a coated conductor, because even a small defect can produce a large hot spot that spans the entire conductor if $a > d/n$, where d is the sample width. If this rather weak condition is satisfied (typically, $n \sim 20$ – 30 in HTSs), then even small defects produce significant local excess dissipation. Figure 10 shows the calculated distribution of the electric field near a planar defect of length $a = 0.1d$ and $n = 20$. Even for such a small defect, the disturbance of $E(x,y)$ is much stronger than the applied field E_0 and extends all the way to the opposite side of the film⁷⁷.

Materials fabrication considerations

One of our fundamental conclusions is that both Bi-2223 and YBCO fulfil the essential requirements for wide applications of superconductivity in the electric utility network. Conductors of Bi-2223 (and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$) are available today from several companies for service in 'real-world' utility sites. Cost and performance are not yet as good as are needed for widespread substitution of copper and iron, but significant special applications exist even at much higher costs. A key question for a cost-sensitive market for HTSs is to understand how much residual J_c performance capability there is in the materials themselves. As we have emphasized, the intragrain J_c capability of both Bi-2223 and YBCO is several times higher than realized in today's conductors. Material defects control the overall critical current density of present conductors and these defects are intimately tied to their particular fabrication processes. Although the specifics of particular fabrication processes are beyond the scope of this article, certain general points can be made to emphasize the decisive role that materials engineering now must play in developing this technology.

As Figure 1 shows, four of the five conductors are made by conventional composite metal-working techniques. A merit of conventional metal working is that there is little conceptual barrier to scale-up. Making longer wires is mostly a question of starting with larger pieces and using larger equipment. Both MgB_2 and Bi-2223 wires or tapes use essentially the same technology as that used for Nb_3Sn powder-in-tube composites. In fact, there is now no fundamental barrier to the production of a kilometre-long MgB_2 wire, and many groups have already demonstrated powder-in-tube conductors^{30,78–82}. What is less clear is how much of a niche there is for MgB_2 to fill, because even in its high resistivity form which maximizes the upper critical field¹⁷ it seems that the perpendicular irreversibility field is less than for Nb_3Sn . This is likely to restrict MgB_2 to applications of lower field, say 2–3 T, where the ability to operate at ~ 20 K is decisive economically¹³. By contrast, it has proven hard to scale up the YBCO-coated conductor to a genuinely continuous process, even

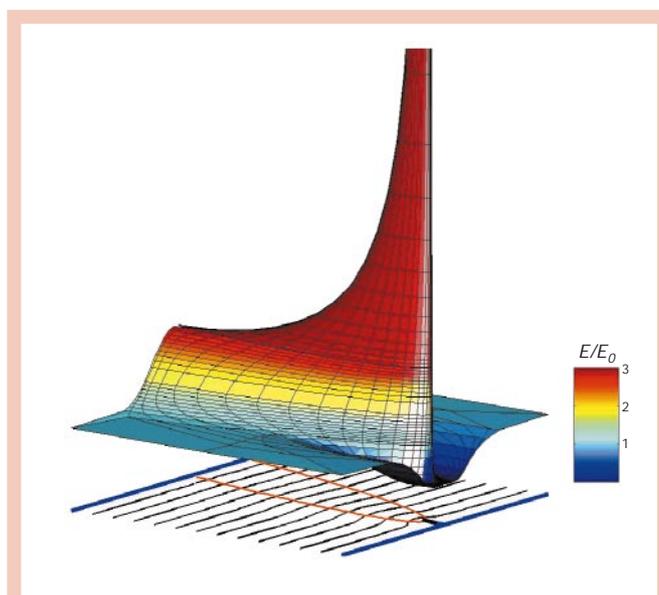


Figure 10 Spatial distribution of the electric field $E(x,y)$ for the power-law E - J characteristic, $E = E_c (J/J_c)^n$, near a planar defect of length a in a film of width d , $n = 20$ and $a = 0.1d$, as calculated in refs 76,77. The lower part of the figure shows the current streamlines (black) and the boundaries (red) of the hot spot of strongly enhanced electric field lying in the film plane. It is clear that even a small defect produces the extended hot spot that effectively blocks the entire cross-section and can result in thermal instabilities⁷⁵.

after multiple proofs of principle have shown that an appropriate substrate–buffer–YBCO architecture can be made by physical vapour deposition and/or chemical methods^{71–74,83}. The key issue here is to understand the defect populations that are characteristic of each fabrication method, and the capability and cost associated with reducing them to tolerable levels.

In assessing progress on HTS conductors, it is particularly relevant to recall that Nb_3Sn is now celebrating 40 years as a conductor and is still improving^{84,85}. Bi-2223 has had 10 years of industrial development and is also still developing strongly in attaining higher current density, reducing cost and in becoming more mechanically robust^{86–88}. The complexity of its microstructure and the interactions between fabrication and basic materials issues is slowly becoming clear⁸⁹, not least because multiple companies worldwide are selling the conductor in competition with each other. As Fig. 8 shows, the key issue is to increase the connectivity of the conductor. The prospect of two- to fivefold improvements in the performance of the present process, coupled to cost reductions triggered by significant scale-up and alternative routes such as overpressure processing⁹⁰ or melt processing⁹¹, could accelerate the early penetration of superconducting technology into the utility network. At the same time, this would provide time and rationale for solving the complex issues associated with scale-up of the YBCO coated conductors.

Any manufacturing method for YBCO conductors relies on the epitaxial deposition of YBCO onto a textured template of one or more oxide buffer layers and a normal metal substrate. This template is usually created through introduction of texture either into the buffer layers by ion beam-assisted deposition (IBAD)⁴⁷, or into the metal substrate by deformation texturing using the rolling-assisted biaxially textured substrate (RABiTS)⁴⁶ approach. The IBAD method enables the use of strong, non-magnetic Ni-superalloy substrates on which an IBAD of aligned YSZ is deposited. This process can achieve a high degree of texture with a layer of YSZ that is $\sim 0.5 \mu\text{m}$ thick. But such a process is widely considered to be too slow to be commercial. IBAD of MgO produces good texture within the first 1–2 nm and thus may be rapid enough to be commercial⁹², but the degree of texture in

present IBAD-MgO-coated conductors is not yet as good as in IBAD-YSZ variants⁹³. The inclined substrate deposition (ISD)⁹⁴ process is much quicker than IBAD, but the texture is significantly worse⁹⁵. Ion-texturing processes for buffer-layer deposition⁹⁶ are receiving renewed attention owing to their simplicity, but their success depends on producing sufficient texture so to compete with the slower and more expensive IBAD process.

All of these methods of producing a textured buffer layer involve physical vapour-deposition processes, which some believe are inherently too expensive for copper and iron replacement⁷¹. An advantage of the RABiTS approach is that all buffer-layer and YBCO-coating steps can be performed with non-vacuum processes. A strong [100] cube texture is introduced into the substrate by conventional rolling and recrystallization⁴⁶. Although the RABiTS approach has been developed mainly with pure Ni, alloyed substrate materials are being developed to increase the strength and reduce the magnetism of the Ni.

At this stage it is not at all clear which route to a coated conductor will win out. The design of a coated conductor is such that a substrate thickness of ~50 μm is needed to support 1–5 μm of YBCO, giving a superconductor fraction of ~5–10%, compared with the 25–50% for conductors made by metal-working techniques. Short samples of both IBAD and RABiTS conductors can both exceed 2 MA cm⁻² for YBCO layers up to ~0.5 μm . But thicker YBCO layers tend to exhibit less epitaxy, more porosity and thus smaller J_c . Degradation of the J_c of the YBCO layer as it grows is a serious and poorly understood problem. The principal cause seems to be loss of epitaxy once the layer gets thicker than ~0.5 μm . A proper understanding is required, as virtually all growth methods except liquid-phase epitaxy seem to share this degradation.

Summary

The fundamental crystal and electronic structure of superconductors determines their structural anisotropy and superconducting properties. Generally, the higher the value of T_c , the higher the anisotropy, and the greater the upper critical field and sensitivity to nanoscale variations of the superconducting properties. This variation is positive in the case of point and line defects, because it can lead to strong flux-pinning and high critical current density. The low carrier density, short coherence length, d-wave pairing symmetry and proximity to the metal-insulator transition of HTSs makes interfaces such as grain boundaries significant barriers to current flow, except at very small misorientations. In the higher carrier density LTS materials such as Nb-Ti, Nb₃Sn, and MgB₂, grain boundaries act as beneficial flux-pinning sites without being barriers to current flow.

Anisotropy exerts two additional, important influences on conductor design, and causes significant suppression of H^* as compared to H_{c2} . MgB₂, YBCO and Bi-2223 all exhibit higher critical current density, irreversibility field and upper critical field when the magnetic field is applied parallel to their layered structure. But in practice, either because of the inevitable misalignments that are present in any real wire or because the magnetic field cannot always be arranged to be parallel to the layers, it is the poorer perpendicular properties that generally control performance. An additional burden for YBCO and Bi-2223 is that the grain-to-grain misalignment must be held to values of 5° or less if obstruction of current across the grain boundaries is to be minimized. Thus planar conductor forms, which confer texture, are preferred.

These fundamental materials issues restrict the fabrication technologies available for conductors. So far all three industrially available conductors, Nb-Ti, Nb₃Sn and Bi-2223, use composite metal-working techniques, which are relatively easy to scale up from laboratory to factory. It is already clear that this technique will work too for MgB₂. But it does not work for YBCO, for which a multilayer epitaxial technique is required. Many techniques using a variety of chemical and physical vapour-deposition techniques can be used to make short samples that demonstrate the concept of the coated

conductor. At present the coated conductor R&D community is attempting to work out which fabrication techniques work cost-effectively in continuous processes. Broad and significant applications exist and await only the resolution of the vital materials issues that control development of the cheap, high-performance conductor-fabrication technology that underpins all superconducting applications. □

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