Next Generation Superconductors for High Field Applications

R. Flükiger and **B. Seeber** of the Department of Condensed Matter Physics and the Institute of Applied Physics, University of Geneva, describe the status of oxide and sulphide superconducting tapes and wires.

The economic generation of steady state magnetic fields by means of superconductors is presently limited to about 20 T. The main reason for this restriction is related to the intrinsic upper critical field (B_{c2}) of today's commercially available superconductors based on Nb_3Sn (see Fig. 1). For higher fields at 4.2 K, a new generation of superconducting materials must be developed, prime candidates being $PbMo_6S_8$ (PMS) crystallizing as the Chevrel phase and high temperature superconducting (HTSC) copper oxide compounds with a layered structure. The former has a B_{c2} at 4.2 K above 50 T which is nearly isotropic owing to the rhombohedral crystal structure with a rhombohedral angle close to 90°. The two-dimensional character of HTSC's implies a very strong anisotropy: for example, the compound $YBa_2Cu_3O_7$ has a B_{c2} at 4.2 K reaching 100 T perpendicular to the c-axis, and 25-60 T in the parallel direction.

Another important parameter for applications is the transport critical current density J_c at the required magnetic field. While supporting the generated magnetic field, the superconductor used to wind a magnetic solenoid must be in the form of a long wire with a reasonably high and constant J_c (typically 5×10^8 A/m^2) over several hundred metres. Obtaining such characteristics at 77 K using liquid nitrogen as the cryogen instead of at 4.2 K with liquid helium presents obvious economic advantages. However, the strong temperature dependence of J_c and B_{c2} implies that for many high field applications, the main operation range will be limited to 4.2 K for the foreseeable future. Nonetheless, achieving more modest fields (a few tesla) at 77 K offers important opportunities.

We review here briefly the status of the development of HTSC tapes and PMS wires for high field applications at 4.2 K. In the case of HTSC's, one is usually considering highly textured, monofilamentary or multifilamentary layers. For the Chevrel compounds, a wire of circular cross-section can be employed.

HTSC Tapes

From reported data on HTSC's, three candidates have been retained with applications at 4.2 K and 77 K in mind, namely $(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ — so-called Bi(2223), the Tl substituted version — so-called Tl(2223), and $YBa_2Cu_3O_7$.

Melt-textured samples of $YBa_2Cu_3O_7$ prepared by adjusting the solidification conditions of the molten compound to create the appropriate crystallographic anisotropy and crystal structure exhibit fairly high J_c's, *e.g.*, 3×10^8 A/m^2 at 77 K under an external field of 1 T [1]. Unfortunately, the poor mechanical properties of this compound render its deformation very difficult. This explains why, in contrast to melt textured samples, silver-sheathed tapes have so far exhibited disappointingly low J_c's owing to very low degrees of crystal texturing and to the presence of superconducting weak links at grain boundaries.

The situation is different for the two (2223) superconductors which are mechanically softer than the Y-based oxide and can be easily brought into a textured configuration by choosing the correct sequence of pressing and annealing steps for the powder-in-tube preparation technique, where a metallic can containing the HTCS powder is thermo-mechanically processed. The mechanical and physical properties of both (2223) phases are very similar, but a substantial difference resides in the formation mechanism. The Tl based phase can be formed by a melting process while Bi(2223) requires a reaction of the solid Bi(2223) phase with a liquid phase mixture and is stabilized by the addition of Pb [2]. It is known that Ag lowers the melting point of Bi(2223) but for the formation of this phase in Ag-sheathed ribbons, the important feature is the lowering of the melting point of the liquid phases, thus promoting Bi(2223) grain growth. In spite of the more complex formation conditions in Bi(2223) with respect to Tl(2223), most publications are restricted to the former as the toxicity of Tl calls for much higher safety standards.

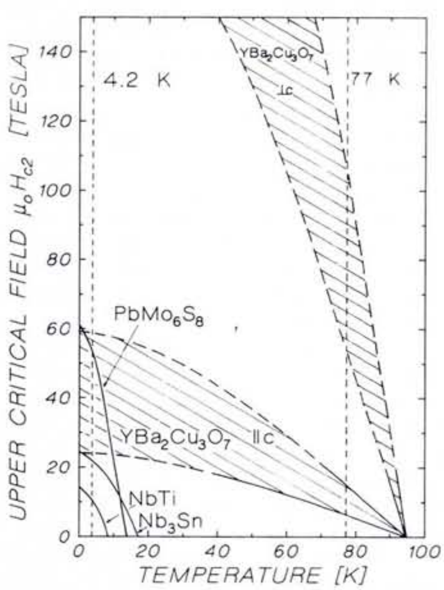

Fig. 1 — Upper critical field B_{c2} as a function of temperature for the commercial superconductors NbTi and Nb_3Sn. Data for the $PbMo_6S_8$ Chevrel phase are also compared with those for the high temperature oxide superconductor $YBa_2Cu_3O_7$ with the magnetic field both parallel and perpendicular to the the c-axis. The hatched areas correspond to uncertainties related to the criterion used to define B_{c2} and to the extrapolation scheme.

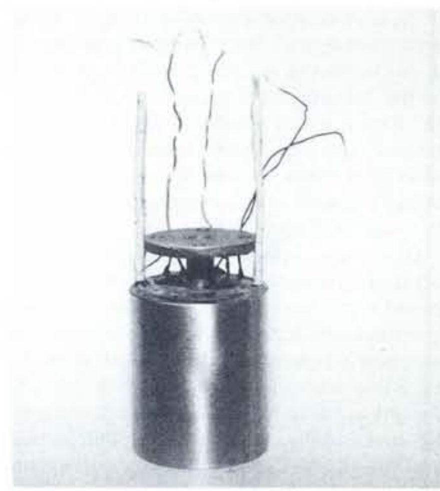

Fig. 2 — An 80 mm diameter coil made from a HTSC wire. It generated 23.225 T at 4.2 K with a 23.0 T backup field meaning that, with sufficient material, a 23.225 T magnet could be built (courtesy of Sumitomo Electric Industries, Osaka, Japan).

Relatively high J_c values have been obtained at 77 K for Tl and Bi(2223) tapes produced by a special type of powder-in-can process, namely the press-and-anneal method. Powders, which may comprise mainly the (2223) phase or a blend of CuO and some other oxides, are packed into a Ag tube which is swaged, drawn and rolled to roughly 0.1 mm thick tape. The tapes are then subjected to one or more pressing/heat treatment cycles. For Ag-sheathed Tl(2223) tapes, J_c (77 K, 0 T) values of 1.4×10^8 A/m^2 have been reported [3] while even higher values were found for Pb-stabilized Bi(2223) tapes (4.7×10^8 A/m^2 [4], 3.3×10^8 A/m^2 [5]); values up to 4.0×10^8 A/m^2 have been achieved in our laboratory. It must be noted, however, that these high values of J_c have so far only been found for tapes less that 40 mm in length where pressures as large as 50 kbar can be applied.

Attempts to reach these values by rolling steps alone (without intermediate pressing) have been unsuccessful. J_c (77 K, 0 T) values for tapes longer than the 10 m required to wind small laboratory magnets have to-date reached on the order of 1×10^8 A/m^2. Nevertheless, small superconducting magnets producing 0.2 T at 4.2 K have already been produced in Japan (Fig. 2).

The variation of J_c with the applied magnetic field B perpendicular to the c-axis (as is the situation in a wound magnet solenoid) is indicated in Fig. 3 for Bi(2223) tapes: there is a fairly strong dependence up to about 1 T. However, J_c is strongly dependent on the orientation of the magnetic field (Fig. 4). It follows that the limiting factor for the production of fields above 0.2 T at 77 K is the occurrence of magnetic field components parallel to the c-axis which arise at both ends of a wound coil. Particularly elaborate configurations have to be developed to solve this problem. Nonetheless, considerable effort is being undertaken in order to further enhance J_c values of HTSC's and to improve the mechanical properties of Ag-sheathed Bi(2223) tapes with applications both at 77 K and 4.2 K in view.

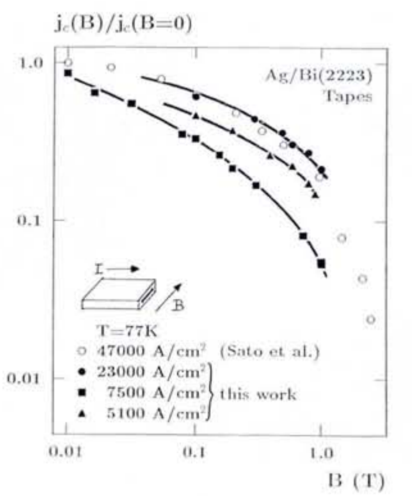

Fig. 3 — Normalized critical current density J_c at a temperature T = 77 K versus the applied magnetic field B for Ag-sheathed Bi (2223) high temperature oxide superconductor tapes with B perpendicular to the c-axis [7]. Densities at zero field are given at the lower left-hand side.

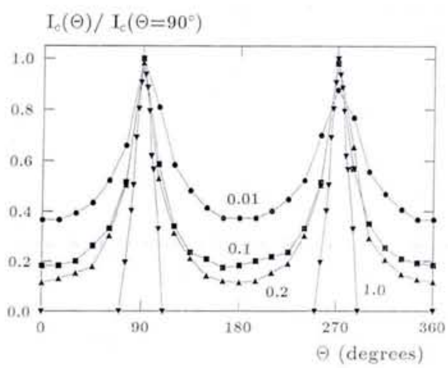

Fig. 4 — Normalized critical current density as a function of the orientation θ of the magnetic field for a Ag sheathed Bi(2223) tape [7]. An angle θ = 90° corresponds to a field B perpendicular to the c-axis.

PMS Wires

Chevrel phase superconductors have been under intensive investigation over the the past 10 years or so. A multinational EUREKA project was launched in 1986 to develop an industrial-scale production capability for PMS-based superconducting wire capable of generating fields above 20 T. Monofilamentary wires of up to one kilometre in length have been produced and coils wound from the conductor demonstrated. The best results reported to-date by the EUREKA project team are for Cu/Nb sheathed material produced using the conventional powder-in-tube method [8]. Other matrix materials such as Mo/stainless steel and Ta/Cu/stainless steel sheaths gave similar results [9, 10]. The Mo, Nb or Ta acts as a diffusion barrier while the steel ensures matching of the thermal expansion between the core and the sheath to suppress thermally induced stresses on cooling from heat treatment temperatures: copper acts as a thermal stabiliser. Starting powders are either a blend of metals and sulphide compounds or pre-reacted PMS, and heat treatments after wire fabrication by swaging, extrusion and drawing are needed to react the powders or to recover superconducting properties.

Critical current densities at 4.2 K exceeding those for Nb_3Sn superconductors have been achieved for short lengths (25 m) of Nb/Cu sheathed monofilamentary PMS material at 20 T (Fig. 5). Important advantages with respect to Nb_3Sn include almost no field dependence of J_c under uniaxial strain in the 8-24 T range (see Fig. 6) and a 30% higher intrinsic elastic limit. Calculations have also shown that the superconducting properties in monofilamentary PMS wire with today's usual filament diameters are intrinsically stable above 20 T at 4.2 K.

D. Cattani [12] using flux profile measurements has demonstrated that J_c within PMS grains in a bulk sample is about ten times larger than J_c for the best wires, and that it increases with an increase in B^*_{c2}, the upper critical field derived from AC susceptibility measurements. Depending on the preparation, B^*_{c2} is also significantly smaller than the virtually sample independent B_{c2} values obtained using calorimetry. It is believed that the low B^*_{c2} value reflects the upper critical field of the superconductor at the grain boundaries so, at present, the most important obstacle in improving J_c appears to reside in the local properties at grain boundaries. The coherence length of PMS is approximately the same as in the *ab*-plane of HTSC's (and much larger than for the *c*-direction) so the problems of grain boundary limited J_c's in the two materials show some similarity. Extensive work is now in progress to investigate the grain boundary aspects.

Conclusions

Based on available B_{c2} and J_c data (Figs 1 and 5) one can conclude that both classes of superconductors, HTSC and Chevrel phases, could be used to produce fields above 20 T at 4.2 K. From the present results, the advantage of HTSC materials is the weaker decrease of J_c above 20 T and

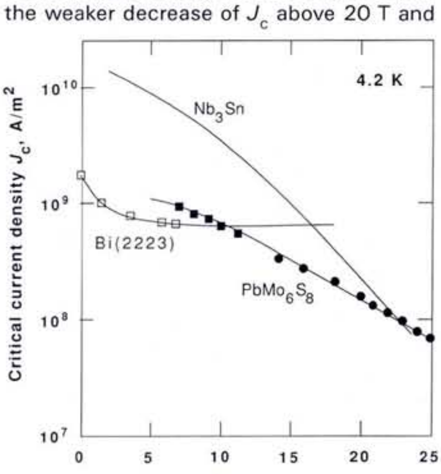

Fig. 5 — Comparison of the critical current density J_c at 4.2 K as a function of the magnetic field for a Nb/Cu sheathed monofilamentary $PbMo_6S_8$ Chevrel phase wire [8, 10], a multifilamentary Nb_3Sn wire and Ag-sheathed Bi(2223) tape [7]. J_c was determined for the superconducting cross-section.

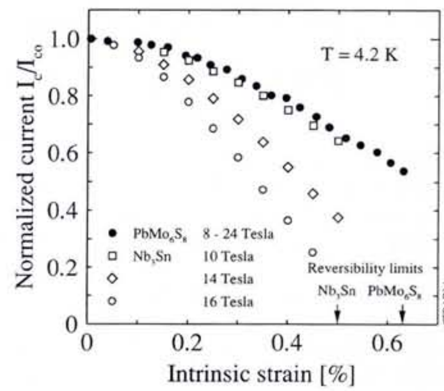

Fig. 6 — Critical current I_c normalized with respect to I_{co}, the critical current at zero strain, versus intrinsic strain at 4.2 K for monofilamentary Ta/Cu/stainless steel sheathed $PbMo_6S_8$ Chevrel phase and Nb_3Sn multifilamentary superconducting wires. The reversibility limits may be shifted to higher or lower values depending on the thermally induced pre-stress [10, 13].

the larger temperature difference between the critical temperature T_c and the operation temperature. However, the isotropic properties of PMS allow the use of round wires which presents some advantages for industrial fabrication and possibly for particular applications. From the data obtained on small test coils [8, 12], and the agreement between J_c in these coils and in short samples, it follows that a high degree of homogeneity can now be obtained in PMS wires, while long Bi(2223) tapes with high J_c still show a considerable variation with the length.

The development of HTSC tapes is presently directed towards high current leads at 77 K (*e.g.* between liquid N_2 and He) and towards solenoids with moderate magnetic fields, the medium term goal being to achieve field values comparable to those produced by conventional superconducting magnets.

REFERENCES

[1] Murakami M. *et al.*, *IEEE Trans. Magn.* **MAG-27** (1991) 1479.
[2] Sunshine S.A. *et al.*, *Phys. Rev. B* **38** (1988) 893.
[3] Okada M., *Jpn. J. Appl. Phys.* **29** (1990) 2732.
[4] Sato K. *et al.*, *IEEE Tras. Magn.* **MAG-27** (1991) 1231.
[5] Heine K. *et al.*, *Adv. Cryo. Engng.* **38** (1991), to be published.
[6] Yamada Y., Obst B. and Flükiger R., *Superconductor Sci. & Tech.* **4** (1991) 165.
[7] Flükiger R. *et al.*, VIth. Int. Workshop on Critical Currents, Cambridge, UK (1991); to be published in *Superconductor Sci. & Tech.*
[8] Genevey P., Vallier J.C. *et al.* (private communication).
[9] Seeber B. *et al.*, Proc. MRS Meeting on Advanced Materials **6** (1989) 83.
[10] Rimikis G. *et al.*, *IEEE Trans. Magn.* **MAG-27** (1991) 1116.
[11] Ekin J.W., Yamashita T. and Hamasaki K., *IEEE Trans. Magn.* **MAG-21** (1985) 474.
[12] Cattani D. *et al.*, *IEEE Trans. Magn.* **MAG-27** (1991) 950.
[13] Goldacker W. *et al.*, Proc. ICMAS (1991).