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Future Directions for Cuprate Conductors

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***Abstract*—Superconducting cuprate conductors present several challenges and research opportunities related to the areas of performance and cost. In order to meet these challenges, a combination of radical new approaches is required. This paper proposes several novel ideas and demonstrates early results in some of these areas.**

***Index Terms*—Crystal anisotropy engineering, Cuprate superconductors, Flux pinning, Liquid assisted growth, T_c enhancement, Uniaxial strain.**

I. Introduction

IT is a quarter of a century since the discovery of high superconducting transition temperatures (T_c) in the cuprates. It is therefore an opportune time to take a step back and consider how far we have progressed towards the goal of creating materials suitable for applications, to openly address the critical challenges that remain in spite of decades of concerted effort and progress, and thence to propose radical new approaches which can be taken to overcome these challenges.

Conductors based on the cuprate superconducting material $YBa_2Cu_3O_{7-\delta}$ (or other rare earth equivalents, herein termed YBCO) have reached a very sophisticated stage of technological development. This success can be attributed to three main factors: templating methodologies for creating highly aligned crystallites and hence low angle grain boundaries; incorporation of artificial vortex core pinning centres to provide strong flux pinning; and careful processing methodologies to ensure the reproducible production of highly crystalline, well aligned material.

With ~1 km lengths of conductor presently being fabricated which carry >100 A currents, one might wonder whether we have already surpassed application requirements and have little left to do. In fact, there is still much to do. First, we need higher current conductors, and second we need to understand and overcome the fact that absolute critical current (I_c) does not generally scale linearly with J_c. Finally, in order to make utility companies interested in widespread uptake and use of coated conductors, close to an order of magnitude decrease in cost is probably necessary.

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Fortunately, there are many unexplored opportunities for advancing cuprate-based technology for conductor applications. For high field conductors, these fall into the four areas of **improving flux pinning** in order to enhance J_c, **increasing T_c** to achieve better properties at operational temperatures, **engineering the crystal morphology** so as to attain grain connectivities as good as in the high interlayer distance compounds, e.g. BSCCO, and **achieving faster growth rates** to reduce production costs. Herein we demonstrate that an intimate understanding of the complex materials science of the cuprates is necessary for both the inception and fulfillment of novel ideas and hence is absolutely fundamental to future progress.

II. Novel Pinning Approaches: Improving Flux Pinning by Designing the Pinning Landscape

Firstly, we consider opportunities in the area of flux pinning. This research takes two forms: core pinning and magnetic pinning. For core pinning, there has been substantial progress over the past five years, yielding order of magnitude increases in the achievable J_c. However, it should be noted that while most pinning additives will improve pinning in some respect, there has been no substantive work in the area of designing the pinning landscape. As a consequence, we remain more than a factor of five away from the theoretical (pair-breaking) limit to J_c (~30 $MAcm^{-2}$ at 77 K, self field [1]), and even further removed at elevated fields.

Core pinning by design requires an understanding of how the nanocomposite (superconductor + pinning phase) materials self-assemble into forms other than simple heteroepitaxial films (Fig. 1). The critical parameter is whether the two phases are immiscible, partially miscible or completely miscible. For the case of complete miscibility it is necessary also to consider the possibility of a lowering of miscibility at lower temperatures which would mean there is a tendency towards clustering and hence spinodal decomposition [2]. Other considerations include the degree of strain between the two phases (which determines the critical size of the second-phase nanoparticles, as described below for tantalate and niobate additions); whether the pinning phase incorporates all of the cations present in the superconductor phase (in which case there is a greater tendency for horizontal plate-like structures to form rather than vertical rod-like structures); and the growth rate resulting from the flux of incoming material (slower growth rates favouring vertical columnar growth and faster growth rates favouring the formation of randomly oriented nanoparticles [3]).

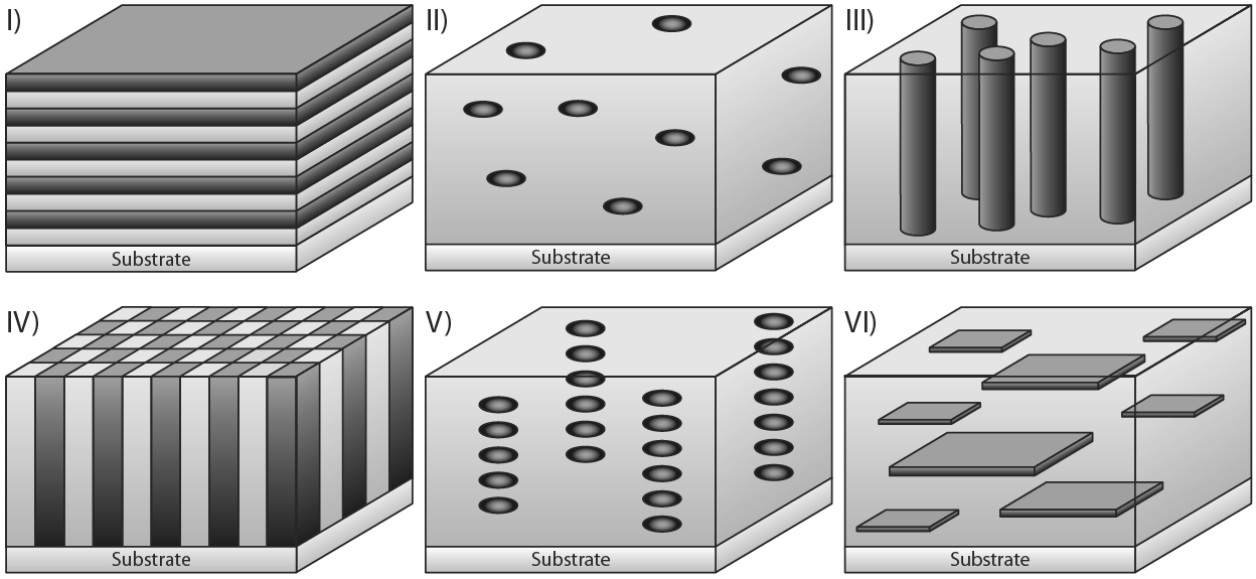

Fig. 1. Possible nanocomposite structures that can form in heteroepitaxial oxide films: I) multilayers, II) nanoparticles, III) nanorods, IV) vertical heteroepitaxial nanocomposite, V) nanocolumns, and VI) platelets.

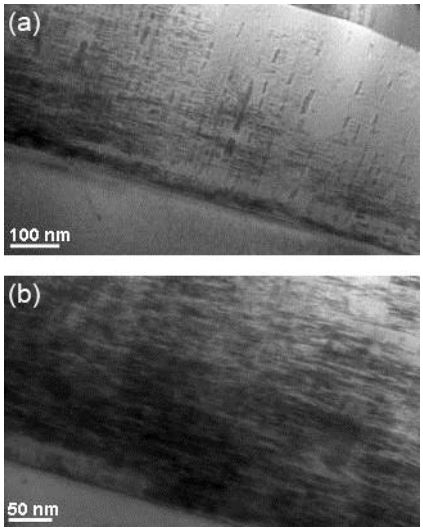

Fig. 2. Columnar pinning structures obtained in 0.3 μm thick films of YBCO + 5 at.% (total) of Gd_3TaO_7 + YBa_2NbO_6. a) Lower magnification cross-sectional TEM image revealing the coarser (~20 nm diameter maximum) nanocolumns present. b) Higher magnification cross-sectional TEM image of the same sample revealing the fine (~5 nm diameter) nanocolumns present. Samples grown by G. Ercolano, University of Cambridge; TEM images by H. Wang and J. H. Lee, Texas A&M University.

Some of our recent work has been targeted towards demonstrating that it is possible to design pinning structures with mixed pinning landscapes. Addition of a low lattice mismatch phase, the tantalate Gd_3TaO_7, and a high mismatch phase, the niobate YBa_2NbO_6, were made to YBCO powder between 5 and 10 at. % in total. Films were grown by pulsed laser deposition from the composite target materials. Fig. 2 shows that a moderately coarse, truncated nanocolumnar structure (Fig. 2a) characteristic of the niobate phase and a very fine nanocolumnar structure (higher magnification image, Fig. 2b) characteristic of the tantalate phase [4] are *both* present in the same sample. There are advantages of achieving the more discontinuous columns for inhibiting flux creep and advantages of achieving highly dense, fine columnar structures for achieving pinning at higher fields. Hence, the tailored combination of both through the incorporation of multiple pinning additions is highly beneficial.

Fig. 3 shows J_c versus applied field parallel to *c* for the films of Fig. 2 compared to other 'best-reported' data for pure YBCO and $BaZrO_3$-doped YBCO [5]. The superior performance of the double pinning additive is clear. Of particular note is the wide plateau in J_c out to ~2 T which is suggestive of a matching field effect consistent with the observed nanocolumn spacing of ~30 nm.

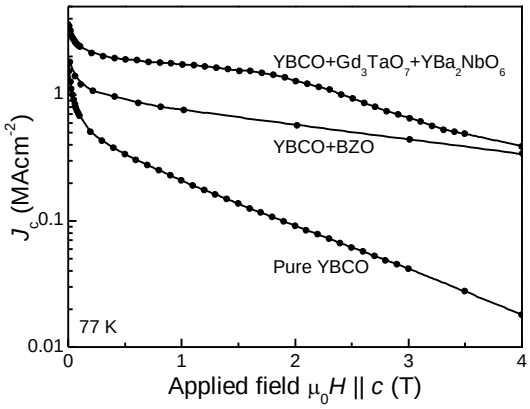

Fig. 3. J_c versus applied field parallel to *c* at 77 K for 0.3 μm thick films of YBCO + 5 at. % (total) of Gd_3TaO_7 + YBa_2NbO_6 [6] compared to pure YBCO and best-reported data for BZO-doped YBCO taken from [5].

Spinodal decomposition is an area which has been studied widely for enhancing pinning in bulk YBCO-related [7] and BSCCO [8] superconductors, but not for films. However, strongly enhanced high field properties which have been observed in mixed RE-123 films are more likely to result from 5–10 nm fine modulations of composition as a result of spinodal effects rather than from rare earth oxide nanoparticles

which often form in such films [9]. Spinodal decomposition in mixed RE-123 (both doped and undoped) is a very profitable future avenue of study for achieving high field pinning effects at 77 K because of the highly controlled, very fine (<10 nm), ordered structures which can be induced [10].

Another underexplored avenue for strongly enhancing J_c is via magnetic pinning. Ferromagnetic additions are proposed to produce a pinning force that is additive to that provided by core pinning through the additional magnetic interaction between the flux vortices and the pinning material [11]. We have proposed a model [12] for this interaction based around a reduction in the Lorentz force acting on a vortex due to its flux being diverted through the magnetic inclusion, which we believe gives the potential for order of magnitude increases in J_c, also at self field, ultimately enabling the depairing current to be reached [13]. Several groups have attempted to demonstrate magnetic pinning effects in YBCO [14]. Until now, however, all practical magnetic pinning additives have resulted in a depression of the superconducting properties [15], generally caused by a reduction in T_c, which is particularly susceptible to 'poisoning' by the magnetic constituent [16]. Hence, materials compatibility of the ferromagnetic addition is the first key hurdle to overcome before any practical pinning approach can be realised. Oxide compounds containing Fe, Ni, or Co (giving a strong likelihood of a high magnetic moment) together with one or more constituents already present in the cuprate phase are most likely to yield a compatible additive phase. We have demonstrated the compatibility of $YFeO_3$ [17], [18], [19] and are currently studying other anisotropic ferrite compounds. There are also promising signs of ferromagnetic pinning in epitaxial layered films [20]. However, further work is required to unambiguously establish the magnetic contribution to the pinning and the precise mechanism.

III. Increasing T_C

For high field applications cooled by liquid nitrogen, YBCO is the only system under serious study. YBCO is classified as a single layer cuprate since only the CuO_2 planes carry the superconducting current, with the Cu-O chain layers acting mainly as charge reservoir layers. The maximum T_c of the single layer cuprate systems is ~100 K. Compared to the standard YBCO-based superconductors being used at present, superconductors with a T_c of 100 K would provide strongly enhanced properties at liquid nitrogen application temperatures since they would be operating at just 0.77 T_c rather than 0.86 T_c — equivalent to operating a 90 K superconductor at 69 K — and so potentially yielding more than a factor two increase in J_c. Hence, it would be worth investigating whether it is possible to achieve a 100 K T_c in a straightforward manner. The potential for achieving higher T_c's has been demonstrated in the A_2CuO_4 system using a combination of chemical and hydrostatic pressure. Chemical pressure was induced by partial substitution of the larger Ba^{2+} ion for Sr^{2+} on the *A* site ($Sr_{2-x}Ba_xCuO_{4-y}$) enabled by using high hydrostatic pressure to force the Ba^{2+} ion to remain on the Sr^{2+} site [21]. A 98 K superconductor was achieved, which is remarkable considering that the more widely studied $La_{2-x}Sr_xCuO_4$ compound has a T_c of just 38 K. In fact, it happened somewhat by chance that the $Sr_{2-x}Ba_xCuO_{4-y}$ compound met the five basic requirements for enhancing T_c, which are:

A1) Optimise the carrier concentration in the CuO_2 plane.
A2) Make the CuO_2 planes flat, square (i.e. cubic or tetragonal crystal structures), and of optimal size.
A3) Eliminate defects in the CuO_2 planes.
A4) Optimise the CuO_2 interplanar distance. Note that for the highest T_c compounds one needs three CuO_2 planes (not more since then defects become prevalent) which lie close to one another.
A5) Have a large Cu-O apical distance since this allows the system to be more 2D-like ensuring carriers are localised in the CuO_2 planes.

The higher T_c in $Sr_{2-x}Ba_xCuO_{4-y}$ compared to $La_{2-x}Sr_xCuO_4$ relates in particular to its increased *a* lattice parameter of 3.88 Å compared to 3.76 Å. The importance of the *a* lattice parameter (requirement *A2*) was understood many years ago [22], as is illustrated in Fig. 4.

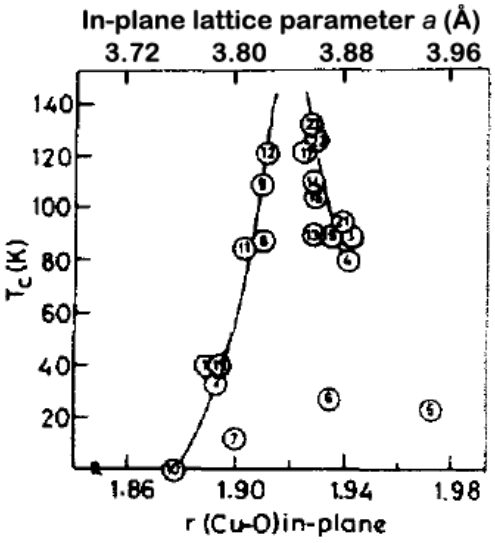

Fig. 4. Maximum T_c values observed (at optimal doping) in the cuprate superconductors as a function of the Cu-O in-plane distance / *a* lattice parameter, after [22].

In fact, it turns out that $Sr_{2-x}Ba_xCuO_{4-y}$ has similar optimum structural features to another single layer cuprate, Hg-1201, which has the same T_c, and very similar *a* (3.874 Å cf. 3.88 Å).

From a practical materials engineering approach, the means to increase T_c of the cuprates is by **decoupling of the doping–*a*–*c* relationship**. This is not trivial since *a* and *c* tend to scale with one another, e.g. when ionic substitutions are made the lattice size and doping both change, with increased doping reducing both *a* and *c*.

Examples which have clearly demonstrated the benefits of controlling and optimising *a* and *c independently* can be seen

in $(La,Sr)_2CuO_4$ single crystals and films. Nakamura *et al.* showed that decreasing *a* and increasing *c* in uniaxially stressed $(La,Sr)_2CuO_4$ led to a T_c increase from 38 K to 52 K [23]. Similar T_c increases were also observed in single layer $(La,Sr)_2CuO_4$ thin films grown on metallic layers in order to induce Madelung strain into the film. This produced an increased *c* (and increased Cu-O apical distance) *without* changing *a* [24]. If it were possible to increase *a* in this system (where it is sub-optimal according to Fig. 4, at 3.76 Å), we predict that it should be possible to increase T_c still further in this compound. However, since the La^{3+} and Sr^{2+} ions (effective ionic radii 1.16Å and 1.26Å, respectively) in the structure do not exert enough chemical pressure to allow *a* to be increased (as do Ba^{2+} ions in $Sr_{2-x}Ba_xCuO_{4-y}$), the T_c may remain limited to below 60 K.

The above results naturally lead to the notion that chemical plus uniaxial pressure rather than chemical plus hydrostatic pressure are required for T_c optimisation / enhancement. Following this approach, it should be possible to create a starting lattice having near-optimal CuO_2 plane size by chemical doping, fulfilling requirement *A2*, and then to achieve an enhanced Cu-O apical distance by straining along the *c* axis thus fulfilling requirement *A5*. Since uniaxial tension along *c* leads to a reduction in *a* (the precise amount depending on Poisson's ratio), it is, therefore, necessary to precompensate for the reduction in *a* by undertaking initial chemical doping to yield a slightly larger-than-optimal CuO_2 plane size, i.e. *a* axis larger than ~3.84 Å. Considering the various experimental difficulties associated with achieving a T_c increase to 100 K in the single layer cuprates, increasing T_c in the double layer or triple layer cuprate systems (where baseline T_c's are already ~130 K) may be a more productive strategy.

Our suggested guidelines for achieving enhanced T_c using combined chemical and uniaxial pressure are therefore:

B1) Study a double or triple layer cuprate system.
B2) Choose compounds which are close to stoichiometric in oxygen since those which are non-stoichiometric may accommodate strain by oxygen loss which will adversely modify doping [25].
B3) Choose a system where doping of the CuO_2 planes is not highly sensitive to external pressure.
B4) Use co-doping to modify the CuO_2 plane size (and hence the a lattice parameter) while allowing optimal doping to be maintained.
B5) Undertake uniaxial strain experiments by thin film growth techniques.

For the last point, the means to study and possibly practically implement uniaxial strain is by growing epitaxial thin films, either as single layers of <50 nm thickness or as multilayers separated by thin strain-inducing interlayers [26], or by using radically new strain controlling methods such as growth of vertical nanocomposite films of much larger thickness than standard films (type IV of Fig. 1) [27].

Let us consider where we could start. For the F-doped triple layer cuprate Hg-1223F under hydrostatic pressure, the T_c (164 K) is the highest measured to date [28]. However, it is questionable whether this system has indeed attained its highest possible T_c since *a* and *c* have not been independently controlled and so it is unlikely that *both* values are optimal.

Fig. 5 can be used to demonstrate the potential for increasing T_c by simultaneous optimisation of both *a* and *c*. We see from the figure that there is a general trend across all cuprate superconductors of increasing T_c with increasing Cu-O apical distance (indicated by the large arrow with positive gradient) in accordance with requirement *A5*. However we also see that, within a given cuprate series, T_c apparently increases with *decreasing* Cu-O apical distance as the number of cuprate layers, *n*, increases, shown by the lines with negative gradient joining the data points. However, this is a misleading correlation since the primary reason for the negative gradients is that the *a* parameter is decreasing at the same time as the Cu-O apical distance is decreasing. Thus it is **not** fundamentally the case that the decreasing Cu-O distance is increasing T_c but rather that the *a* lattice parameter is decreasing towards its optimal value (requirement *A2*). The dashed arrow with positive slope indicates what might be achievable if both *a* and the Cu-O apical distance were able to be independently controlled in order to fully optimise T_c.

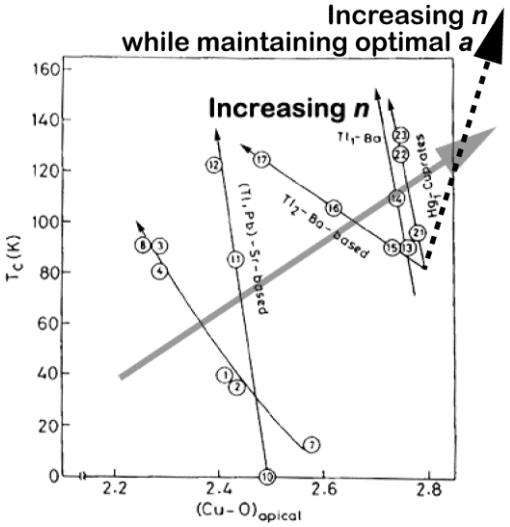

Fig. 5. T_c's in various cuprate superconductor systems as a function of the Cu-O apical distance, after [22], with solid grey arrow superimposed to indicate the general trend of increasing T_c with increasing Cu-O apical distance. The dashed arrow indicates the potential effect of breaking the interdependence of *a* and *c*, enabling the full optimisation of T_c.

Returning to Hg-1223F again, not only are *a* and *c* not independently controlled under the action of hydrostatic pressure but a very large degree of buckling (5°) of the CuO_2 planes also results [29], violating requirement *A2*. Application of uniaxial pressure in this system to reduce buckling effects, combined with light co-doping to moderately expand the CuO_2 plane size, represents a practical approach to increasing T_c beyond 164 K, perhaps even as far as 200 K, without requiring the discovery of any new materials systems, which is an inherently high-risk approach.

IV. Engineering the Crystal Morphology

There exists an unfortunate situation in the high T_c cuprates that systems having high irreversibility field, H_{irr}, also form blocky three-dimensional crystals. Such blocky crystals yield poorly connected grains when they are combined (in either thin film or bulk form). Fig. 6 shows a plot of H_{irr} versus blocking layer separation (the blocking layer being the insulating layer separating the superconducting CuO_2 planes). The diagram can be divided into two regimes, highlighting the divide between the two opposing properties. A large blocking layer separation also implies a large *c*/*a* ratio, which means it is favourable for crystals to grow more rapidly along the short repeat direction, i.e. along *a*, as is the case for Bi-2223 crystals. This growth rate anisotropy was predicted by Bravais in 1866 [30].

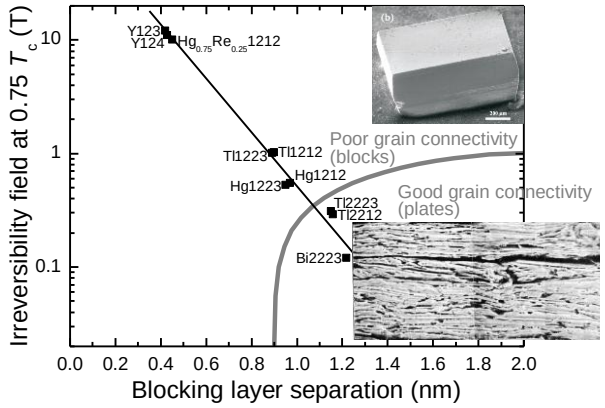

Fig. 6. Irreversibility field at 0.75 T_c versus blocking layer separation for a number of cuprate superconductors. Images of typical blocky (YBCO) and platey (BSCCO) crystals originating in the two regimes are superimposed on the plot.

State-of-the-art coated conductors utilise substrate templating methodologies to achieve highly biaxially textured films of YBCO. Although very costly they are required because it is not possible to achieve well connected grain boundaries in YBCO by powder in tube techniques because of the lack of natural texturing from the blocky crystals which occur in YBCO powders. On the other hand, it is possible to achieve adequately connected grains in BSCCO conductors by such methods, because low angle colony boundaries naturally form from the platey BSCCO crystals [31].

If it were possible to make crystals of YBCO of similar morphology to BSCCO crystals, namely highly anistotropic platey crystals which couple better [32], then it would be possible to create a much more cost-effective conductor processing route. The question then is how to make low blocking layer separation crystals form with plate-like morphologies just as the high blocking layer separation crystals do.

We believe there are real possibilities for controlling crystal morphology independently of intrinsic crystallography based on recent advances in crystal morphology engineering in other systems, e.g. in polyol synthesis of Ag nanowires [33] and in the biological synthesis of oxides [34]. Fig. 7 shows a cross-sectional image of biosynthesised crystals of YBCO prepared from a nitrate solution precursor with the initial addition of the biopolymer dextran to control the morphology during crystallisation and subsequent addition of NaCl to engineer the plate-like crystal growth upon calcining [35]. We obtain a reasonable alignment of plates approaching the low angle grain boundary microstructure observed in BSCCO (Fig. 6, lower inset). A schematic of a possible current path through the material is also shown. The magnetisation J_c values (calculated using the grain size as the current length scale) for the biotemplated material of this work compared with commercial YBCO powder show a substantial increase. This suggests either good connectivity between the platelets and/or improved flux pinning within them. Preliminary studies suggest that both effects play a role. Further work in applying biotemplated crystal growth to YBCO is therefore very promising.

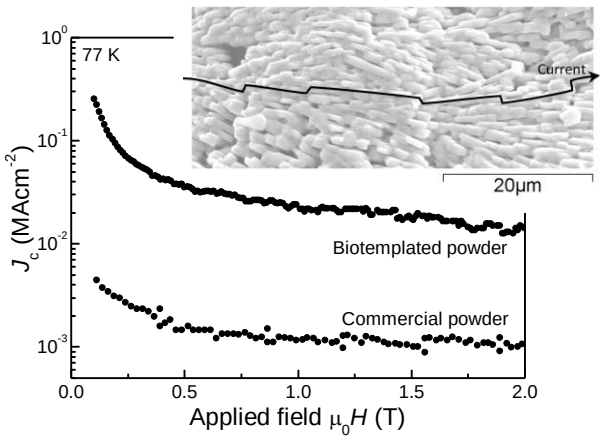

Fig. 7. Magnetisation J_c of biosynthesised platey YBCO powder compared to commercial material. (Inset) Cross-sectional SEM image showing platey crystals of YBCO and a schematic possible current path through the material. Sample and data from Z. Zhang, University of Cambridge.

V. Increasing Conductor Growth Rate

A relatively straightforward and less radical means than biotemplating for achieving the desired order of magnitude reduction in cost of coated conductors than some of the other approaches discussed here is simply to grow them faster. One way to do this without compromising on crystallinity, and while achieving excellent epitaxy is to grow films in the presence of a controlled thin liquid layer. One process of this type which we have studied is hybrid liquid phase epitaxy (HLPE). We have worked on optimisation of the HLPE process on short coated conductor samples achieving critical currents of 300 A on chemical solution deposited $La_2Zr_2O_7$-buffered Ni-W RABiTS substrates [36]. Of note is that the HLPE YBCO layer was formed in a matter of minutes. The liquid assisted growth process is compatible with many different deposition routes including MOCVD and e-beam evaporation, and so we believe it is a very opportune time for industry to explore and develop this process on longer length samples.

VI. Conclusion

There are several possible novel approaches to improving the properties of cuprate superconductors for conductor applications that have yet to be explored. In this paper, we have discussed several directions which we believe are worth pursuing intensively in the future. We have presented our own preliminary results on the generation of mixed pinning landscapes engineered through the combination of pinning additions of different lattice mismatch phases such as niobates and tantalates, and have outlined how this work might be taken to the next level. We have also described our attempts to date to achieve effective magnetic pinning in cuprate materials, in the context of similar work in low-T_c materials showing that drastic enhancements in J_c are possible if compatible materials and processes can be found. We have described in some detail a feasible materials-engineering approach to improving the T_c of existing cuprate families, highlighting the fact that even an apparently minor T_c enhancement to, say, 100 K would have a significant effect on the superconducting properties at an operating temperature of 77 K. Finally, we have suggested alternative synthetic approaches to the ubiquitous standard thin-film technologies for achieving textured high-T_c cuprate materials with dramatic potential for cost reduction.

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