INTERGRANULAR Jc DETERMINATION IN TOROIDAL HIGH TEMPERATURE SUPERCONDUCTORS USING A SOFT-CORE CLOSED MAGNETIC CIRCUIT

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Abstract

An examination of weak link coupling between grains has been made using a noncontact magnetic technique. We report intergranular Jc(T) measurements for unaligned and grain aligned sintered $YBa_2Cu_3O_{7-x}$ (YBC) toroids. Supercurrent is deduced from measurement of the net current encircling the core and Jc is found from supercurrent saturation in contrast to onset pickup voltage criteria. Agreement with recently reported direct transport measurements on the aligned sample suggests that the conduction mechanism is the same as for direct transport. A **universal** normalized Jc(T) behavior is found which follows the same junction critical current temperature dependence previously reported in bicrystal transport measurements. It is proposed that this technique may be extended for the measurement of critical currents in thin film samples and that present bulk intergranular current densities appear to be high enough to produce a soft-material limited superconducting electromagnet at 77K.

Introduction

A fundamental obstacle limiting applications of polycrystalline high-Tc superconductors is the low intergranular critical current densities which are 3-4 orders of magnitude smaller than intragranular Jc values. Several investigators have been able to attribute the low intergranular Jc's to a classic weak link nature of coupling between grains.[1-4]

One way to examine intergranular currents is to fabricate a superconducting toroid and induce supercurrents inductively. The measurement described here is similar in many respects to A.C. techniques such as those used to measure superconductive A.C. loss.[5] Although various experimental arrangements have been used to make such measurements,[6-13] a particularly simple arrangement results when toroidal superconductors are threaded on a soft magnetic core as shown in Fig. 1. Using this configuration, one may apply a current to the primary winding and observe the induced supercurrents which screen the applied field via measurements of flux changes in the secondary winding.

Since flux changes in the secondary are given by the net primary and induced currents, we can monitor the growth and saturation of the induced supercurrent by measuring the flux changes in the secondary when a known primary current is applied. Previous researchers[6-8] report a sudden onset of secondary pickup voltage which is consistent with a hard clip in the induced supercurrent density and is used to obtain Jc. However, due to difficulties inherent with extrapolating to zero pickup voltage, we felt it of interest to investigate the dynamics of the supercurrent buildup.

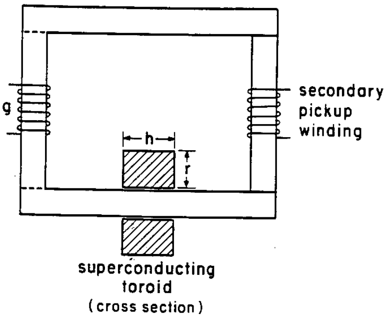

Figure 1. Schematic of transformer core sample measurement geometry.

In this way we have studied the temperature dependence of Jc for both unaligned and grain-aligned YBC toroids to try to find the nature and degree of grain to grain coupling in bulk samples. The geometry was chosen to give induced supercurrents in the a-b plane for the aligned sample.

Experimental Techniques

Typically, a 5.5 Hz triangular waveform was applied to the primary monitored by a 1 ohm series resistor to obtain the primary current. The primary current and secondary voltage were measured using a HP7090A data acquisition plotter which had a sensitivity of 2.5 microvolts. The system was controlled using custom written software on a 80286 based machine. After digitization of the signals, the secondary voltage was integrated to obtain the flux swing through the core. Eddy currents were found to be negligible for our core at these low frequencies. The temperature dependence of the permeability was measured without a sample and this was also found to be negligible. Nonlinearities in the permeability were of course taken into account as described in the analysis below. Fig. 2 shows typical waveforms for the primary current and secondary pickup voltage for applied current levels which induce saturated supercurrent in the toroid. In Fig. 2(b), we observe the surge voltage "blip" noted by others[6-8] when the supercurrent saturates. On an expanded scale in Fig. 2(c) we see an additional feature: a small but significant pickup which lags the primary by 90 degrees.

The unaligned sample was prepared from commercially available[14] YBC powder following standard techniques and was approximately the same size and shape as a Wint-O-Green lifesaver[15]. A detailed description of the sample preparation technique for the aligned

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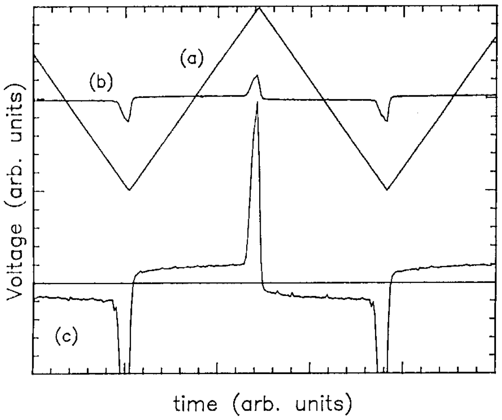

Figure 2. (a) Primary current waveform, (b) Secondary voltage surge associated with hard clipping of supercurrent density at Jc and (c) Same secondary voltage shown on an expanded scale to illustrate pickup before supercurrent saturation. All at a frequency of 5.5 Hz and temperature of 5K.

sample has recently appeared.[16] Holes were cut using an ultrasonic disk cutter. For the aligned sample, the c-axis was aligned coaxially with the hole. The degree of alignment reported by Tkaczyk and Lay[14] for X-ray rocking curves was 7-9 degrees full width at half maximum.

A schematic of the cryostat that has been designed and fabricated appears in Fig. 3. A thin walled stainless steel cryostat has been made to fit conveniently into a standard (Linde CMSH) LHe storage dewar. The sample space is thermally isolated from the bath by a vacuum space which contains a controlled pressure of He transfer gas and a bifilar heater thermally anchored to the sample space wall. The transfer gas in the sample space itself is controlled through the same valve plumbing as for the vacuum jacket.

A description of the sample holder and magnetic circuit may be given following Fig. 1. The magnetic circuit consists of two "C" shaped silicon steel transformer core sheets (relative permeability of 1000) wound with 80 turn (36AWG) primary and secondary windings. The magnetic core circuit formed when the "C" pieces are put together is a square with an average magnetic path length of 5.1 cm and a cross section of 0.035 cm^2 at the windings. A toroid is shown in cross section threaded onto the arm of the "C" pieces to illustrate the measurement geometry. The clamp which holds the soft magnetic pieces in intimate contact is made from solid Cu blocks to help eliminate thermal gradients and provide rapid thermal equilibration. A Cu/Au-0.07% Fe thermocouple is used to measure temperature and is thermally contacted to the Cu clamp via a mica sheet and GE 7031 varnish. The thermocouple has been calibrated at 4.2, 77.3 and 273.15K. It is estimated that temperature measurement error with the calibrated thermocouple remains less than 0.2K from 4.2 to 300K.

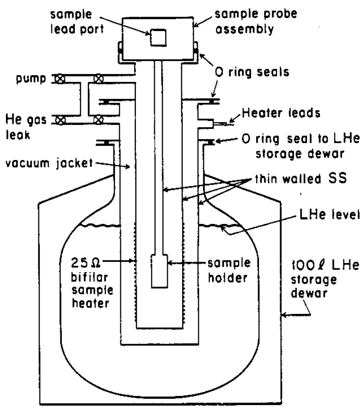

Figure 3. Schematic of Dewar and probe.

Results and Discussion

In Fig 4, the flux swing in the secondary is plotted against the amplitude of the primary current at various temperatures. The signature features are a linear regime at low primary currents followed by a rapid flux swing above a critical applied primary current. This is the magnetic circuit analogue of an I-V characteristic. Since the secondary flux swing is determined by the net current encircling the core, the supercurrent is determined by subtracting the known primary current from the measured net current. This is equivalent to subtracting the curve without supercurrent taken above Tc from the one obtained when supercurrent is opposing the applied primary current. This curve subtraction includes the nonlinear response of the core and allows us to determine Jc(T) from the supercurrent at which core screening breaks down.

The presence of the linear regime in Fig. 4 is a little surprising. In this regime, the supercurrent is 180 degrees out of phase with the primary current as expected, but with an amplitude of only 80% of that of the primary. Such a regime can be

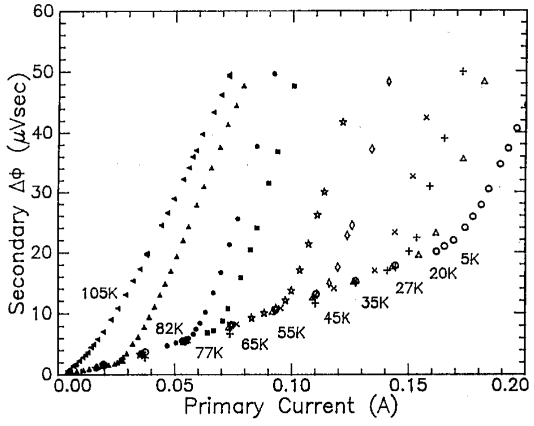

Figure 4. Flux swing measurements showing excess primary current required to magnetize the core as temperature is lowered and Jc increases.

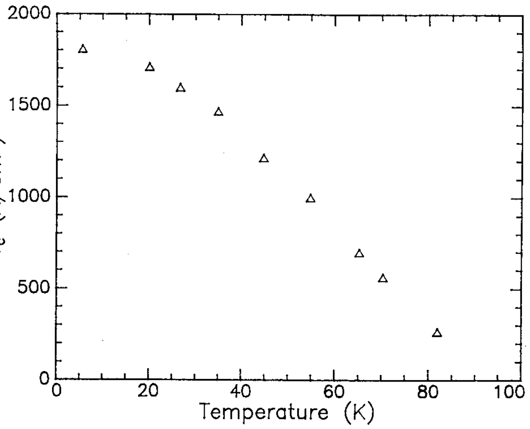

Figure 5. Jc(T) for grain-aligned YBC toroid

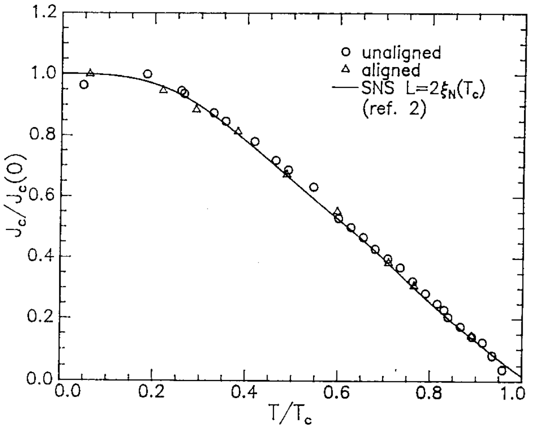

Figure 6. Normalized Jc(T) behavior for both aligned and unaligned samples. Tc=92K for both the aligned and unaligned samples. The solid curve is taken from ref. 2.

attributed to three possible sources namely, leakage in the magnetic circuit, resistive dissipation, or carrier inertia. Lack of an appropriate phase shift or any significant frequency dependence between 0.5 and 50 Hz appears to rule out resistive dissipation. Also, if the linear regime is attributed to carrier inertia alone, London electrodynamics gives a ratio of carrier density to effective mass of $n/m^* = 7\times10^{15}$ m^{-3} where m^* is given in m_e and a carrier charge of 2e is assumed. This leads to an unrealistic carrier density and effective mass as well as a London penetration depth on the order of centimeters. A flux leakage explanation appears to be qualitatively correct although about a factor of 5 larger than what would be expected for our circuit. Further work is indicated to determine if the linear regime can be accounted for quantitatively by flux leakage alone.

The linear regime of Fig 4 aside, each measurement clearly exhibits a breakdown current above which flux rapidly pours through the core when the supercurrent in the toroid is saturated giving an unambiguous discontinuity of the slope at Jc.

Fig. 5 shows the Jc(T) measured in this way for the grain aligned material which carried the highest supercurrent density at all temperatures. The inductively measured Jcs at 5K and 77K are found to be 1800 and 400 A/cm^2 respectively in good agreement with the low field measurements of Ekin et al.[3] confirming the consistency of the inductive method with direct transport. Although the technique does not discriminate surface versus bulk currents this quantitative agreement suggests that the conduction mechanisms and paths are essentially the same in the transport and inductive techniques.

The normalized Jc temperature dependence for aligned and unaligned samples is given in Fig. 6. Although the unaligned sample had a Jc(T=5K) value 2 orders of magnitude smaller than the aligned sample, the normalized temperature dependence is identical. It is remarkable that the observed Jc(T) behavior corresponds exactly with that observed in single grain to grain pairs[2]. That is an Ambegaokar-Baratoff[17] type behavior:

$$J_c(T) = \frac{\pi\Delta(T)}{2eR_{NN}}\tanh\left[\frac{\Delta(T)}{2kT_c}\right]$$

where e is the electron charge, R_{NN} is the specific tunneling resistance, and $\Delta(T)$ is the temperature dependent gap energy.

It was pointed out in the bicrystal work[2] that a similar temperature dependence is obtained by Likharev[18] for Jc(T) in SNS junctions for a ratio of

$$L/\xi_N(T_c) = 2$$

where L is the weak link length and $\xi_N(T_c)$ is the coherence length of the normal layer. The universality of the temperature dependence would suggest that the physical mechanism controlling the weak link limited current is essentially the same even for large 3D arrays of junctions, a wide distribution of grain misorientations, and for samples having slightly differing Tc.

Applications

An extension of this technique to multiply connected thin film samples is straightforward. However, using a C-shaped yoke with a narrow opening, this technique may be even adapted to the measurement of critical current densities in thin films for rapid screening without physical damage to the film. Because of their low H_{c1}, the contribution to the reluctance of the gap from the Meissner effect of the film should be small in high-Tc superconductors. If necessary it may be suppressed by applying a DC bias field. The screening current may be deduced from flux swing measurements as previously discussed. We note that if the sample is simply connected rather than toroidal, it can no longer be assumed that the whole flux path is encircled by screening currents and intragranular currents will contribute to shielding. This effect might be minimized by focusing the flux with a proper yoke design.

Another tantalizing application is in the fabrication of a superconducting electromagnet. We consider a soft core with a gap. In this case, a magnetizing current is applied to the primary coil to obtain the saturated critical current of the toroid. When the field is decreased one would reverse the toroid screening current resulting in a remanent state as depicted in Fig. 7 for the closed circuit. The presence of a usable gap would of course make a much larger current necessary for saturation of the soft material but this should not be an insurmountable problem. Since some bulk high-Tc materials possess transport critical current densities on the order of 10^4 A/cm^2 in zero field at 77K, a simple calculation with a gap length of 1 cm would indicate that a toroid of only 1 cm^2 cross section could produce enough magnetomotive force at 77K to obtain a 1T field in the gap. In fact, for the aligned sample studied here with Jc(H=0,T=77K) of 400 A/cm^2, a 25 cm^2 cross section toroid should suffice. Such a magnet might find widespread applications as a replacement for many larger, power consuming electromagnets.

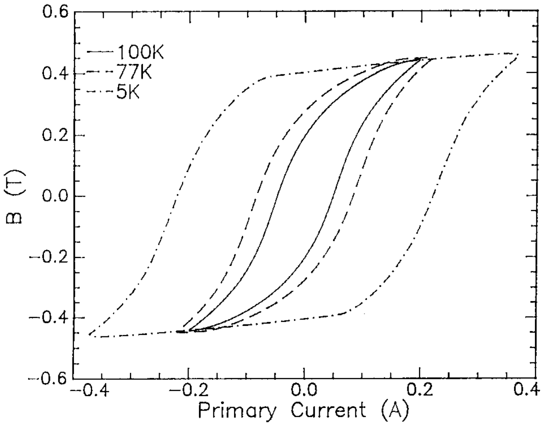

Figure 7. Loops showing induction versus primary current obtained for magnetizing the soft core above and below Tc.

Conclusions

A simple inductive method has been devised and used to obtain temperature dependent intergranular critical current densities in bulk polycrystalline and grain aligned ceramic superconductors. The Jc values are found to agree with previously reported direct transport measurements. It is remarkable that such bulk specimens show a universal normalized Jc temperature dependence which is the same as reported in transport studies of single grain boundary junctions. Noncontact intergranular screening and a 77K superconducting electromagnet are proposed as potential applications arising from variations of the measurement technique.

Acknowledgements

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References

1. J. W. Ekin, *Adv. Ceram. Mater.*, **2**, 586 (1987)
2. J. Mannhart, P. Chaudhari, D. Dimos, C. C. Tsuei, and T. R. McGuire, *Phys. Rev. Lett.*, **61**, 2476, (1988)
3. J. W. Ekin, H. R. Hart, Jr. and A. R. Gaddipati, *J. Appl. Phys.* **68**, 2285 (1990)
4. J. R. Clem, *Proc. Intl. Conf. on High-Temperature Superconductors*, Interlaken, Switzerland (1988)
5. J. D. Thompson, M. P. Maley, and J. R. Clem, *J. Appl. Phys.* **50**, 3531 (1979) (and references therein)
6. E. A. Harris, J. E. L. Bishop, R. L. Havill and P. J. Ward, *J. Phys. C.*, **21**, L673, (1988)
7. E. A. Harris, J. E. L. Bishop, R. L. Havill and P. J. Ward, *Cryogenics*, **28**, 685 (1988)
8. G. P. Meisner and C. A. Taylor, *J. Appl. Phys.* **66**, 5518 (1989)
9. K. Kanbara, T. Takizawa, H. Matsuura and R. Shimadate, *Physica C*, **156**, 727 (1988)
10. M. A.-K. Mohamed, J. Jung and J. P. Franck, *Phys. Rev. B* **41**, 6466 (1990)
11. E. M. Gyorgy, G. S. Grader, D. W. Johnson, Jr., L. C. Feldman, D. W. Murphy, W. W. Rhodes, R. E. Howard, P. M. Mankiewich, and W. J. Skocpol, *Appl. Phys. Lett.* **52**, 328 (1988)
12. H. Kupfer, I. Apfelstedt, R. Flukiger, R. Meier-Hirmer, W. Schauer, T. Wolf, and H. Wuhl, *Physica C*, **153-155**, 367 (1988)
13. C. C. Welch and B. R. Barnard, *Cryogenics* **29**, 411 (1989)
14. W. R. Grace Co., Columbia, Md.
15. We are indebted to D. P. Clougherty for pointing this out to us.
16. J. E. Tkaczyk and K. W. Lay, *J. Mater. Res.* **5**, 1368 (1990)
17. V. Ambegaokar and A. Baratoff, *Phys. Rev. Lett.*, **10**, 486 (1963) and **11**, 104(E) (1963)
18. K. K. Likharev, *Rev. Mod. Phys.*, **51**, 101, (1979)