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Magnetization of imperfect superconducting grains

R. L. Peterson
Electromagnetic Technology Division, National Institute of Standards and Technology, Boulder, Colorado 80303
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A critical-state theory of the magnetization of superconducting grains containing nonsuperconducting regions is presented which shows that the thickness of the sheath of supercurrents around these regions can be more important than the grain dimension in determining the magnetization. This may explain some apparently conflicting results on the magnetization of high-T_c powders of different sizes.

The scale on which supercurrents flow in pellets, films, and powders of $YBa_2Cu_3O_x$ and other high-T_c superconductors has been studied by many investigators. The dependence of the magnetization hysteresis ΔM on sample or particle size is commonly used in the analysis. Critical-state theories show that ΔM should vary as $J_c d$, where J_c is the critical current density and d is the linear dimension of the region containing the supercurrent and is the length in question.

When pellets of sintered granular $YBa_2Cu_3O_x$ are cut to smaller dimensions, or ground to a powder size approximating the grain size, ΔM remains approximately the same or decreases only slightly.[1-4] This shows that d in the expression for ΔM is not the sample size of these granular sintered bulk materials, and was one of the early reasons for believing that the grain boundaries define the regions within which most of the supercurrent flows at modest to high magnetic fields.

However, when the pellet is powdered to a yet finer size and the magnetic behavior of powders of different sizes is measured, samples prepared in different laboratories or by different procedures have shown differing results. In some cases the hysteresis varies linearly with powder size[1] or the size of the portions of a thin film defined by scribing,[5] showing that the entire "particle" out to its boundaries forms the region of supercurrent flow. In other cases,[2,6,7] ΔM has shown little variation with particle size; in these cases features within the particles are apparently restricting the dimensions of the current flow.

Thus it is commonly agreed that for some of the materials there are subgrain features which control ΔM and therefore the regions in which the supercurrents flow. What these features are is uncertain. The assumption of perhaps most investigators is that there may be barriers, perhaps twinning planes, through which the supercurrent cannot flow and which effectively confine the currents to small regions.

In this paper I propose an alternative model, which can be analyzed by means of critical-state theory, and which explains the observations. In a sense it is opposite to the currently popular model—it assumes that the currents are excluded from certain regions of the grain rather than confined within certain regions.

The motivation for considering an alternative model is severalfold. The confinement model is difficult to quantify. The twinning planes characteristic of $YBa_2Cu_3O_x$, perhaps forming cages or boundaries confining the supercurrent, would not seem to be involved since samples of $YBa_2Cu_3O_x$ have been prepared showing the opposing results (compare Refs. 1, 2, 5, and 6). Also, it is difficult to see how planar dislocations or other features can substantially confine the supercurrent, as they must when ΔM is observed to be independent of particle size; leakage of current around them would cause ΔM to vary with particle size. It is well established that at low magnetic fields internal features do not block the supercurrent.[6,8]

The alternative model is based on the assumption that superconducting grains may contain islands of nonsuperconducting material. Insufficient oxygenation would be a primary cause although others are possible. An experiment on single crystals of $YBa_2Cu_3O_x$ (≈ 200 μm on a side) showed that the supercurrent in that case apparently did not flow in the interior of the crystal;[9] the same could be true for smaller grains, although there the oxygenation is much easier to accomplish. For the subsequent analysis, it is immaterial whether the nonsuperconducting islands are normal, semiconducting, or insulating inclusions, or even cavities.

A critical-state analysis of a superconducting particle having nonsuperconducting islands near its center shows that ΔM will vary principally as the thickness of the current sheath rather than as the size of the particle. Consider a cylindrical tube of superconductor having outer radius r_o and inner radius r_i, whose wall thickness is thus $w = r_o - r_i$. Each of these dimensions is assumed to be large compared with the magnetic (London) penetration depth. Kim and co-workers[10,11] first studied this situation both theoretically and experimentally. The experimental results show that when a field H is applied parallel to the axis of the cylinder, the field in the center of the tube follows the applied field smoothly after the critical state is reached, that is, after the supercurrents have extended completely throughout the wall of the tube. Sometimes flux jumps are seen initially, but these vanish as the field increases. The field in the center is about equal to the field within the superconducting wall at its inner surface.

The tube is now in the critical state, with critical current density $J_c(H_i)$ throughout the wall. I present here results based on the Bean model,[12] which takes J_c to be independent of the internal field $H_i(r) = B(r)/\mu_0$, because it gives a formula that is grasped quickly. Other assumptions about $J_c(H_i)$, such as that of Kim and coworkers[10,11] can be made with no essential difference in the final result. Magnetization expressions based on the Kim model can be obtained in closed form, but are sufficiently complicated that their implications are not easily seen by inspection; they are given in the Appendix.

The magnetization is computed as

$$M = -H + \frac{1}{A}\int_0^{r_i} 2\pi r H_i(r_i)dr + \frac{1}{A}\int_{r_i}^{r_o} 2\pi r H_i(r)dr , \quad (1)$$

where the area A used in averaging the internal field is πr_o^2. The internal field in the critical state when H is increasing is, in the Bean model,

$$H_i(r) = H - J_c(r_o - r) . \quad (2)$$

Evaluating Eq. (1) with this form of $H_i(r)$, and the equivalent formula for decreasing H, gives the magnetization difference at the same value of H as

$$\Delta M = 2J_c w(1 - x + x^2/3) , \quad (3)$$

where $x = w/r_o$.

Thus, if the wall thickness is small compared to the radius of the cylinder, *the characteristic length for the magnetization hysteresis is the wall thickness, not the cylinder radius*. Further, the hysteresis does not change rapidly with decreasing r_o for w significantly smaller than r_o. Figure 1 illustrates this for both the Bean and Kim models.

A critical-state calculation gives the same type of result for a slab whose center is filled with nonsuperconducting material:

$$\Delta M = 2J_c w(1 - w/D) , \quad (4)$$

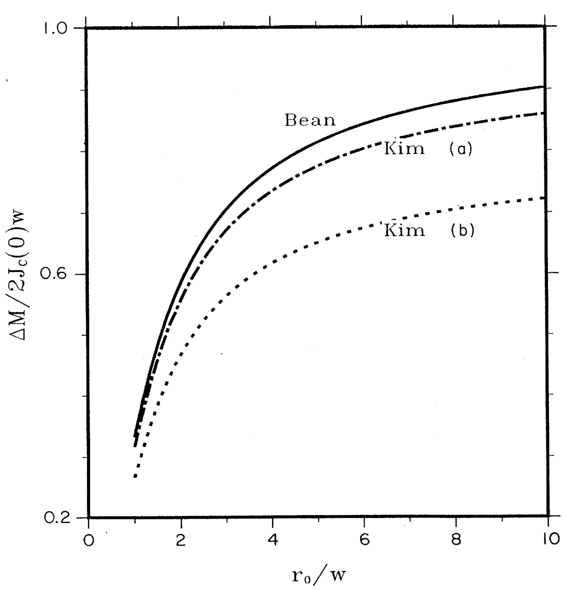

FIG. 1. Dependence of the magnetization hysteresis on size of the grain relative to the thickness of the supercurrent sheath. The solid line is from the Bean model. The other lines are from the Kim model (see Appendix) for an applied field $H = 5 \times 2J_c(0)w$, with $H_o = 20H$ (curve a) and $H_o = 4H$ (curve b).

where w is the width of the superconducting region on each side of the slab, and D is the width of the slab. The same type of expression probably also holds for a more general geometry in which the current flows primarily to the outside of a central area containing several nonsuperconducting regions, although a calculation to prove this would be very difficult. Figure 2 illustrates this schematically. The currents between the nonsuperconducting regions within the grain largely cancel and the net current is confined mostly to the outer regions. If the grain is broken, the magnetization would not change significantly since the average width of the region containing the supercurrents changes only a little. However, in a high-quality sample for which the size of the nonsuperconducting island is small relative to the grain size, the magnetization changes more rapidly with grain size.

These results may be the explanation for the differing dependencies on powder size mentioned earlier, in which the hysteretic magnetization of the superconducting materials was measured at fields on the order of 1 or a few teslas, and the materials were presumably in the critical state. In the theory given here, samples having differing amounts of nonsuperconducting region will have different hysteretic character upon powdering to the same particle size. ΔM should show behavior approximating that of Fig. 1 upon powdering. From this curve the effective width of the supercurrent sheath could be estimated.

To this point I have emphasized the situation where the supercurrent is mostly excluded from the central portions of the grain. A critical-state approach can also address the case in which many small nonsuperconducting islands are distributed rather uniformly throughout the interior of a grain. The nonsuperconducting regions can be idealized as unconnected beads on strings, the strings being arranged in concentric circles of different radii. Currents between beads on a given string will be negligible, so that the net current can be considered to flow in concentric circles or cylinders about the center of the grain.

The critical-state magnetization of two or more concen-

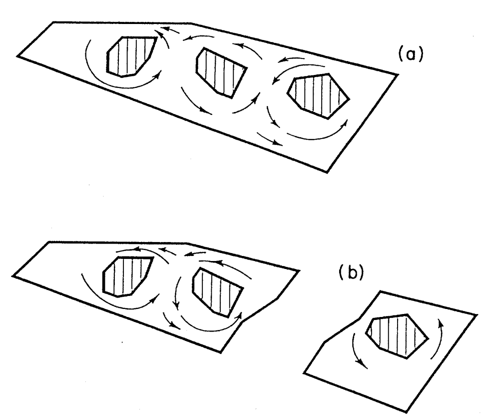

FIG. 2. (a) A hypothetical grain with inclusions of nonsuperconducting materials (shaded). The curved arrows indicate the current patterns. (b) The grain split apart, showing approximately the same average width for the region of the supercurrent.

tric hollow cylinders is easy to obtain in the Bean model and is calculated as above. If the nth cylinder has outer and inner radii of R_n and r_n, and the outer cylinder is the Nth, the resultant hysteresis is

$$\Delta M = \frac{2J_c}{3R_N^2} \sum_{n=1}^{N} (R_n^3 - r_n^3) . \quad (5)$$

In terms of the wall thickness $w_n = R_n - r_n$, this takes the form

$$\Delta M = 2J_c \sum_{n=1}^{N} w_n(\delta_n^2 - x_n\delta_n + x_n^2/3) , \quad (6)$$

where $x_n \equiv w_n/R_N$ and $\delta_n \equiv R_n/R_N$.

A number of special cases can be examined from this result. Just two will be presented here. If the supercurrent wall widths w_n are small compared to the wall radii R_n, that is, if the supercurrents flow in narrow sheaths about the center of the particle,

$$\Delta M \approx 2J_c(w_N + w_{N-1}\delta_{N-1}^2 + \cdots + w_1\delta_1^2) . \quad (7)$$

The hysteresis then varies as the sum of the weighted wall thicknesses (sheath thicknesses) and not as the outer cylinder radius (particle size). In this case ΔM varies sublinearly with particle dimension.

In an opposite limit, if all supercurrent wall thicknesses and all spacings between the walls are equal to w, that is, if the nonsuperconducting islands are all about equal in size and distributed uniformly, we find

$$\Delta M = \frac{2J_c w N}{3} \left[1 + \frac{3}{4N} \right] . \quad (8)$$

If N is very large, that is, if there are very large number of small superconducting islands, this result varies as wN, which in this case is one-half the radius of the particle. Hence the critical-state approach shows that for a very large number of rather uniformly distributed nonsuperconducting regions, ΔM will vary linearly with the outer cylinder radius, or grain size.

Thus critical-state theory shows a range of behavior of ΔM, depending on the configuration of the nonsuperconducting regions.

The other model—confinement—claims that the supercurrents may in some cases be contained within small regions inside the grain. It is opposite to the proposal of this paper—the currents are *included in* rather than *excluded from* certain regions. No specific picture for a containment mechanism has yet been forthcoming, possibly because it is difficult to understand how it would occur. I have already mentioned the problems with twinning planes. The networking of planar dislocations or other defects would have to be such that current cannot flow around them, for then ΔM would vary with particle size. The containment picture requires that current does not exist outside the containment regions; in other words, there cannot be a mixture of contained currents and current flowing around these regions. If there were, ΔM would vary with grain size. Thus the containment regions must completely fill the grain and be separated only by surfaces, or be superconducting islands surrounded by nonconducting material, all within a single grain.

To compare the two ideas, consider the two extreme cases. (i) ΔM varies linearly with powder size. In this case, the grain is either homogeneously superconducting or contains nonsuperconducting regions which are small relative to the grain size and rather uniformly distributed. There can be no current containment regions unless they are large, of the order of the grain itself. (ii) ΔM is practically independent of powder size. In this case, the grain could contain either large nonsuperconducting islands or small containment regions. Intermediate variation of ΔM could be explained on either picture.

The distinction between the two ideas should be testable by modifying the properties of the grains in known ways, for example, by forming grains under different oxidation conditions or by creating different densities of defects. Measuring ΔM as a function of powder size in nontwinned materials would be useful in assessing the role of twinning planes in $YBa_2Cu_3O_x$.

In summary, I have applied the critical-state theory of the magnetization of hard superconductors to the situation of imperfect grains. The theory shows that when the supercurrent is confined to the outer portions of the grain because the inner regions are nonsuperconducting, the magnetic hysteresis will vary principally as the width of the region in which the current flows rather than as the width of the grain. I suggest that this may explain why some materials show a variation of ΔM with particle size while others do not.

Discussions with J. W. Ekin have been helpful and I extend my thanks to him.

APPENDIX

I give here the expressions for the negative magnetization (M_-), occurring for an increasing field H, and the positive magnetization (M_+), occurring for decreasing field, resulting from use of the Kim formula applied to a hollow right circular cylinder. The Kim assumption for the field dependence for the critical current density is

$$J_c(H_i) = \alpha/(H_i + H_o) , \quad (A1)$$

where H_o is a characteristic field and $\alpha = J_c(0)H_o$. The internal field is, from the Maxwell equation $\nabla \times \mathbf{H}_i = \mathbf{J}$ and the assumption that $\mathbf{J}$ equals the critical current density $\mathbf{J}_c$,

$$H_i = H_o + [H'^2 \pm 2\alpha(r - r_o)]^{1/2} , \quad (A2)$$

where $H' \equiv H + H_o$. The plus sign is used when H is increasing, and the minus sign when H is decreasing. Application of Eq. (1) then gives

$$M_{+,-} = -H' + \left[\frac{r_i}{r_o} \right]^2 H_{+,-} + \frac{1}{\alpha^2 r_o^2} \left[\frac{(H'^5 - H_{+,-}^5)}{5} - \frac{h_{+,-}^2 - (H'^3 - H_{+,-}^3)}{3} \right] , \quad (A3)$$

where

$$H_{+,-}=(H'^2 \pm 2aw)^{1/2}, \quad \text{(A4)}$$

and

$$h_{+,-}=(H'^2 \pm 2ar_o)^{1/2}. \quad \text{(A5)}$$

(These expressions for $M_{+,-}$ are not those of Kim, Hempstead, and Strnad[10] because their M is defined as the difference between the field in the center and the applied field.) ΔM is found from $M_+ - M_-$. The result, normalized to $2J_c(0)w$ for easy comparison with the result based on the Bean model, is plotted in Fig. 1 for two values of H_o.

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