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On the negative values of the geometric factors in the intragranular flux-trapping model and the hysteresis in the $J_c(B_a)$ dependence of polycrystalline superconductors

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Abstract

A mathematical reformulation of the problem of the hysteretic dependence of $J_c(B_a)$ in polycrystalline superconductors using the mean-value theorem was considered to obtain information on the geometric factors and percolative paths of the current through the study of different samples. In this paper the polycrystalline superconductor is regarded as a single Josephson junction whose parameters G (geometric factor) and H_0 (critical field of the junction) depend on the applied magnetic field H_a, in a way connected to the flux-trapping characteristics of the superconducting grains.

1. Introduction

The transport critical current density J_c, of the polycrystalline high-temperature superconductors has been found to be strongly hysteretic in applied magnetic field, H_a [1–4]. This behavior is commonly attributed to the presence of trapped flux in the grains [2].

As is well known in the case of (RE)–Ba–Cu–O polycrystalline superconductors, the maximum of the $J_c(B_a)$ dependence in decreasing applied magnetic fields is obtained when $B_a = B_p$ where $B_p > 0$ for every value of $B_{am} > \mu_0 H_{c1g}$ (here B_{am} is the flux density of the maximum applied magnetic field and H_{c1g} the first critical field of the superconducting grains); and B_p increases with B_{am}. The (Bi–Pb)–Sr–Ca–Cu–O system shows a behavior quite different from the one usually observed for (RE)–Ba–Cu–O superconductors, i.e., $B_p = 0$ for every value of B_{am}. To explain the differences between these behaviors Muné et al. suggested the introduction of negative values of the geometric factors in the intragranular flux-trapping model [5]. In this paper we regard the polycrystalline superconductor as one Josephson junction whose parameters G (geometric factor determining the local magnetic field at the junction) and H_0 (critical field of the junction) depend on the external magnetic field as demonstrated through the application of the mean-value theorem, and based on this new mathematical formulation we prove the necessity of the introduction of negative values of G not only in the (Bi–Pb)–Sr–Ca–Cu–O system, but in certain cases of the (ER)–Ba–Cu–O system as well.

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2. Experimental

Samples with the nominal compositions of $Bi_{1.64}Pb_{0.36}Sr_2Ca_2Cu_3O_y$, $Bi_{1.4}Pb_{0.6}Sr_2CaCu_2O_y$ and $GdBa_2Cu_3O_{7-\delta}$ were prepared by a conventional solid-state reaction using the basic oxides and carbonates. The calcination of the Bi powders was performed at 800–820°C for 24 h. The powders were ground, uniaxially pressed and sintered at 845–850°C. The first composition was slowly cooled in the furnace and the second one rapidly in air. The critical temperature of the samples was determined by the four-probe technique and $T_{c1} = 110$ K and $T_{c2} = 81$ K were obtained, respectively. In the Gd ceramic preparation four calcinations were performed at 880°C, 900°C, 920°C and 940°C for 16 h. The powders were ground after each calcination. Finally they were pressed, sintered for 500 h and slowly cooled at a rate of 0.2°C/min down to room temperature [6].

Thin slabs were cut from the samples prepared as described above with typical dimensions of $d = 0.2$ mm (thickness), $w = 2$ mm (width) and $l = 10$ mm (length). The four-probe technique was used to measure the $J_c(B_a)$ dependence in zero-field cooling (ZFC) conditions in an increasing applied magnetic field (virgin curve) and in a decreasing applied field (returning curve), the latter for different values of B_{am}. As is well known, when B_{am} surpasses a certain value, the shape of the returning curve does not change even when B_{am} increases. This case is usually denominated the saturation case [7,8] which was measured for all the samples.

3. The mathematical formulation of the problem

The most general formulation of the $J_c(B_a)$ dependence for the granular superconductors takes into account the statistical distribution of the lengths of the junctions, their angles of orientation relative to the intergranular magnetic field, and the geometric factors of the intergranular regions whose absolute values equal the effective demagnetization factors of the grains configuration around the junctions, and their signs are related to the type of percolative path inside the ceramic [5,6]. This can be written in the following manner:

$$J_c(B_a) = \int_{\theta_{min}}^{\theta_{max}} \int_{L_{min}}^{L_{max}} \int_{G_{min}}^{G_{max}} \left| \frac{\sin\left(\pi \frac{\Phi}{\Phi_0}\right)}{\pi \frac{\Phi}{\Phi_0}} \right| \times \eta(G) \beta(L) \Omega(\theta) \, dG \, dL \, d\theta, \quad (1)$$

where $\Phi = 2\lambda L \mu_0 H_i \sin\theta$, $\eta(G)$, $\beta(L)$, and $\Omega(\theta)$ are the statistical distributions of G, L and θ, respectively; L is the length of the junction and θ is the angle between the normal to the plane of the junction and the intergranular magnetic field H_i. H_i depends on the applied magnetic field H_a, the geometric factor G, and the granular magnetization M in the following way:

$$H_i = H_a - GM. \quad (2)$$

Eq. (1) can be simplified if we introduce the parameter $H_0 = \Phi_0 / 2\lambda\mu_0 L \sin\theta$ and can be written as follows:

$$J_c(B_a) = \int_{H_{0min}}^{H_{0max}} \int_{G_{min}}^{G_{max}} \left| \frac{\sin\left(\pi \frac{H_i}{H_0}\right)}{\pi \frac{H_i}{H_0}} \right| \times \eta(G) \xi(H_0) \, dG \, dH_0; \quad (3)$$

here $\xi(H_0)$ is the statistical distribution of H_0. If we apply now the mean-value theorem we obtain

$$J_c(B_a) = \left| \frac{\sin\left(\pi \frac{H_a - G'M}{H_0'}\right)}{\pi \frac{H_a - G'M}{H_0'}} \right|, \quad (4)$$

where $G_{min} < G' < G_{max}$ and $H_{0min} < H_0' < H_{0max}$.

In this way the polycrystalline superconductor can be regarded as a single Josephson junction whose parameters G' and H_0' depend in general on the external magnetic field. We can simplify Eq. (4) by defining $f(B_a) = (H_a - G'M)/H_0'$ which gives

$$J_c(B_a) = \left| \frac{\sin(\pi f(B_a))}{\pi f(B_a)} \right|. \quad (5)$$

Two conditions of extreme points can be obtained from expression (5):

$$\tan(\pi f(B_a)) = \pi f(B_a) \tag{6}$$

and

$$\frac{df(B_a)}{dB_a} = 0. \tag{7}$$

The first condition will not be considered here because it describes the situation in which the maximum value of the $J_c(B_a)$ curve equals one, i.e., the case of an "ideal" ceramic with uniform H_0 and G. The second, however, is related to the maximum of the returning curve where $f(B_a) = H_i'/H_0'$. It is necessary to point out that H_i' is the internal magnetic field of the "equivalent" Josephson junction whose $J_c(B_a)$ characteristic equals the current density of the polycrystalline superconductor because its parameters G' and H_0' depend on the external magnetic field.

In all the cases the virgin and returning curves satisfy Eq. (4) which can be written as follows:

$$J_c(B_a) = \left| \frac{\sin\left(\pi \frac{H_i'}{H_0'}\right)}{\pi \frac{H_i'}{H_0'}} \right|. \tag{8}$$

It is very important to note that Eq. (8) can be applied to both the virgin and returning curves, just taking into account that the $G'(H_a)$ and $H_0'(H_a)$ dependences are different in each case. Figs. 1 and 2 show the positive branch of the $f(B_a)$ dependence for the virgin and returning curves in Gd–Ba–Cu–O and (Bi–Pb)–Sr–Ca–Cu–O samples, respectively. $f(B_a)$ was obtained by solving Eq. (5) after substituting the $J_c(B_a)$ experimental data. We suppose that H_i' changes sign when $f'(B_a) = 0$ and $J_c(B_a)$ have a maximum. In this way H_i' has a discontinuity at B_p because the experimental fact that the maximum of the returning $J_c(B_a)$ curves never reaches the intensity of $J(0)$ in the virgin ones indicates that H_i' never vanishes in the first case.

4. Results and discussion

In Figs. 3, 4, and 5 the experimental and calculated $J_c(B_a)$ dependences are shown for the different samples in the cases of the virgin and returning saturated curves. The calculated $J_c(B_a)$ dependences were generated following the method described in Ref. [8], based on the application of Eq. (3). In order not to introduce too many unknown parameters, a single value of H_0 was used, i.e, $\xi(H_0) = \delta(H_0 - H_{0m})$, where δ is the Dirac delta function and H_{0m}

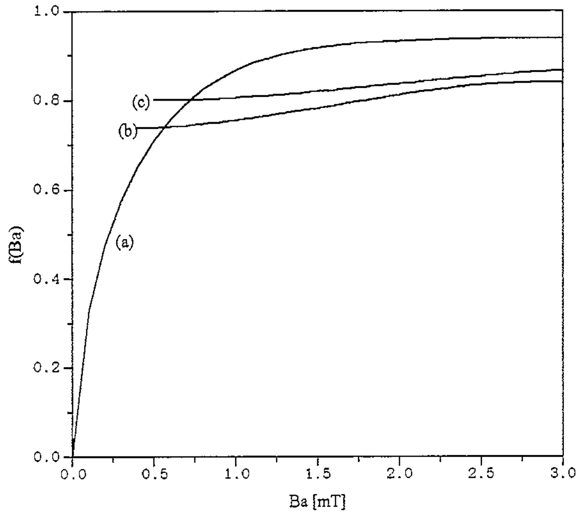

Fig. 1. $f(B_a)$ vs. B_a characteristics for the sample with nominal composition $GdBa_2Cu_3O_{7-\delta}$: (a) virgin curve; (b) and (c) returning curves from 10 mT and 26 mT, respectively.

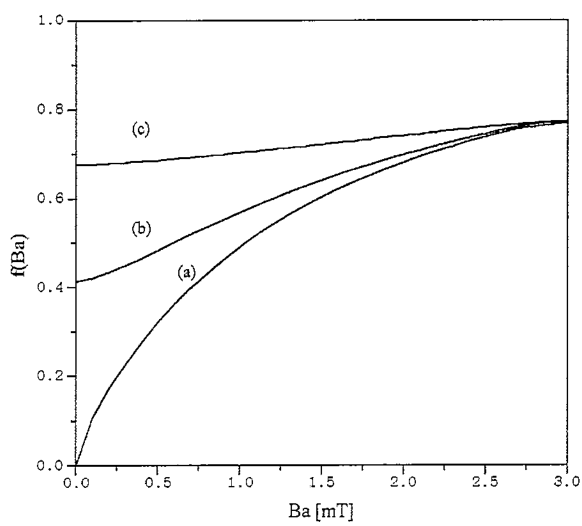

Fig. 2. $f(B_a)$ vs. B_a characteristics for the sample with nominal composition $Bi_{1.64}Pb_{0.36}Sr_2Ca_2Cu_3O_y$: (a) virgin curve; (b) and (c) returning curves form 16 mT and 26 mT, respectively.

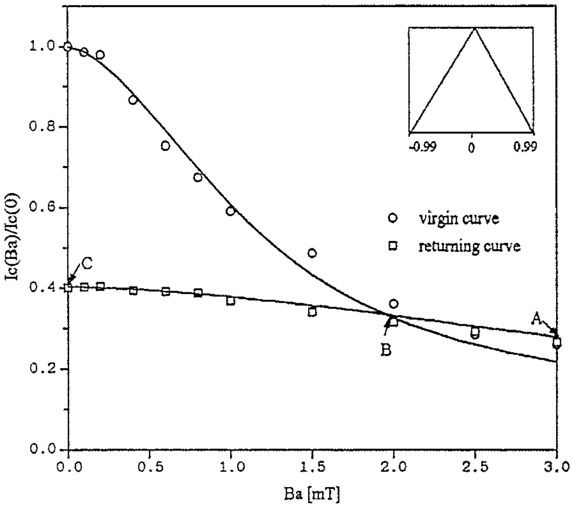

Fig. 3. Calculated and experimental critical current densities as a function of the flux density of the applied magnetic field, B_a, for the virgin and returning curves corresponding to the saturation case for the sample with nominal composition $Bi_{1.64}Pb_{0.36}Sr_2Ca_2Cu_3O_y$. For this sample, $H^* = 16$ mT and the mean value of the critical field of the junctions was $H_{0m} = 2.1$ mT. The inset displays the distribution chosen for the geometric factors G.

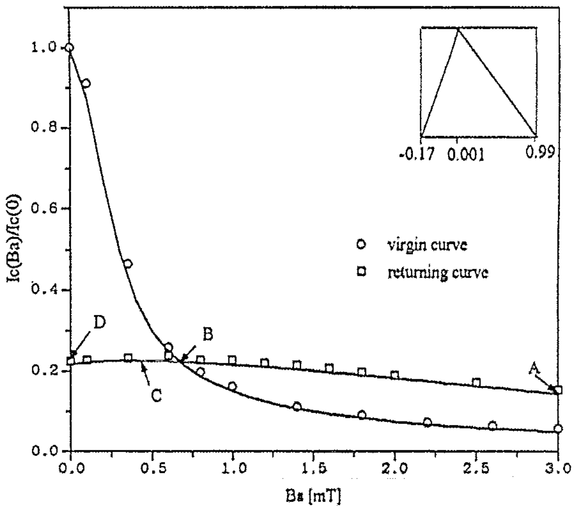

Fig. 5. Calculated and experimental critical current densities as a function of flux density of the applied magnetic field, B_a, for the virgin and returning curve corresponding to the saturation case for the sample with nominal composition $GdBa_2Cu_3O_{7-\delta}$. For this sample, $H^* = 15$ mT and the mean value of the critical field of the junctions was $H_{0m} = 0.58$ mT. The inset displays the distribution chosen for the geometric factors G.

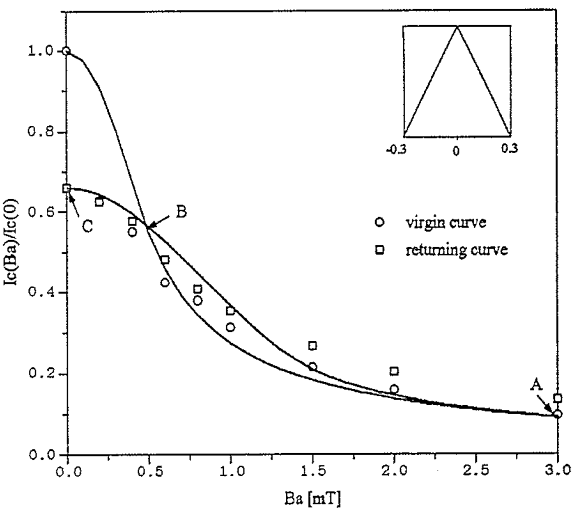

Fig. 4. Calculated and experimental critical current densities as a function of flux density of the applied magnetic field, B_a, for the virgin and returning curve corresponding to the saturation case for the sample with nominal composition $Bi_{1.4}Pb_{0.6}Sr_2CaCu_2O_y$. For this sample, $H^* = 9$ mT and the mean value of the critical field of the junctions was $H_{0m} = 0.87$ mT. The inset displays the distribution chosen for the geometric factors G.

is a constant obtained from the fitting of the virgin curve.

The new mathematical formulation of the problem gives us the possibility to analyze some features which cannot be evaluated directly from Eq. (1). Let us consider the points both in the returning and virgin curves corresponding to the same value of $J_c(B_a)$. When $J_c^r(B_a) = J_c^v(B_a)$, where J_c^r and J_c^v are the current densities for the returning and virgin curves, respectively, we can suppose that

$$|H'_{iv}| = |H'_{ir}|, \tag{9}$$

and we have

$$\left|\frac{H_a^v}{1-G'}\right| = \left|\frac{H_a^r - G''\dfrac{H^*}{2}}{1-G''}\right|, \tag{10}$$

where H_a^v and H_a^r are the intensities of the applied magnetic field for the virgin and returning curves, respectively; H^* is the full penetration field of the Bean's model [8], and $G_{min} < G'' < G_{max}$. Observe that, in general, G'' can be different from G', since we have applied the mean-value theorem to the integrals of two different functions corresponding to the virgin and returning saturated curves. Two expressions for G'' can be obtained from Eq. (10):

$$G''_1 = \frac{\frac{H_a^r}{H_a^v}(1-G') - 1}{\frac{H^*}{2H_a^v}(1-G') - 1} \tag{11}$$

for the region where $H'_i > 0$ in the returning curve (See Fig. 5, returning curve, AC region), and

$$G''_2 = \frac{\frac{H_a^r}{H_a^v}(G'-1) - 1}{\frac{H^*}{2H_a^v}(G'-1) - 1} \tag{12}$$

in the case $H'_i < 0$ (see Fig. 5, returning curve, CD region). $G''_2 > 0$ for all the values of G' $(-1 < G' < 1)$ but in the case of G''_1 the same does not occur. The denominator of the expression (11) is positive because in most of the cases for good sintered samples $H^*/2H_a^v > 5$ and $(1-G') > 0.5$.

However, the numerator can be positive or negative. When $H_a^r < H_a^v$ (see Figs. 3, 4, and 5, returning curves, regions BC) and we take $G' > 0$, then $H_a^r/H_a^v(1-G') < 1$ and $G''_1 < 0$, which indicates that we must consider negative values in the statistical distribution of G.

Figs. 3 and 4 show the behavior of two different samples of the (Bi–Pb)–Sn–Ca–Cu–O system. They are similar because both have a symmetrical statistical distribution of geometric factors with the difference that $\eta(G)$ is narrower for the sample shown in Fig. 4. In both samples we can note that $G'' > 0$ in the regions AB and $G'' < 0$ in the BC. We must expect a similar granular structure in these materials [9] even when they have different critical temperatures than corresponding to the (2223) and (2212) phases in the (Bi–Pb)–Sn–Ca–Cu–O system [10,11]. In the sample whose virgin and returning curves are shown in Fig. 5, $G'' < 0$ only in the region BC. This result indicates that the statistical weight of the negative values is less in this case than in the former ones.

Fig. 6 shows the virgin and returning curves corresponding to a polycrystalline superconductor whose statistical distribution of the parameters G comprises only positive values, as extracted from

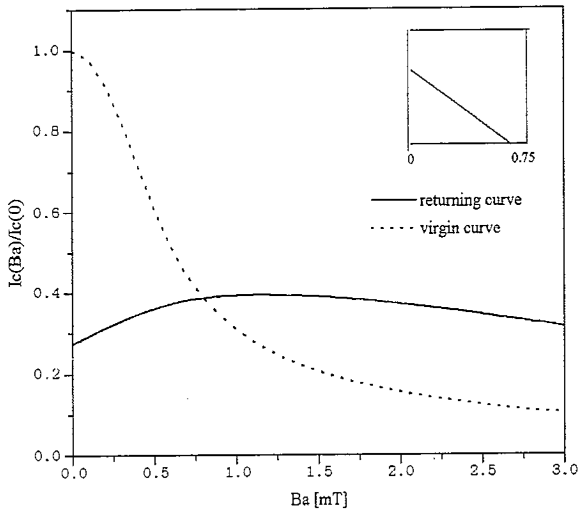

Fig. 6. Virgin and returning curves generated for a polycrystalline superconductor the statistical distribution of parameters G of which have only positive values. We took $H^* = 20$ mT and the mean value of the critical field of the junctions as $H_{0m} = 1.29$ mT. The inset displays the distribution chosen for the parameter G.

Ref. [8]. In these cases the position of the maximum of the returning curve is always at the right of the virgin curve in the zone of positive values of the external field. If some region of the returning curve had a behavior qualitatively different regarding the virgin one than the one presented in Fig. 6, we would be able to assure the presence of negative values of G in its statistical distribution. Then, we can obtain information about the statistical distribution of the geometric factors directly from experimental data before a precise fit of the virgin and returning curves with the intragranular flux-trapping model [8]. The facts discussed above demonstrate that the presence of negative values of G can be justified mathematically in the frame of the intragranular flux-trapping model. Even more, it can be assessed that, if the use of negative values of G is not enough for the fitting of the experimental curves, the model itself must be abandoned.

On the other hand, it is reasonable to think that the long time of synthesis in the samples of the (RE)–Ba–Ca–O system introduces certain variations in the granular structure and because of this reason its hysteretic behavior in the $J_c(B_a)$ dependence can change. Thus, like in the (Bi–Pb)–Sr–Ca–Ca–O system, percolative paths with $G < 0$ can exist, as demonstrated in Ref. [6] for long sintering Gd–Ba–Cu–O ceramic samples. This fact tells us the possibility of some alignment of the grains, though in a lesser degree than in the case of the Bi ceramics [5].

5. Conclusions

The behavior of the $J_c(B_a)$ dependence in polycrystalline superconductors can be simulated, using the mean-value theorem, by a single Josephson junction whose parameters G' and H_0' depend on the magnetic field applied to the material assuming an intragranular flux-trapping model. Based on this formulation we proved that the shape of the returning curve relative to the virgin one is determined by the distribution of the local geometrical factors at the intergrain regions. The necessity of the introduction of a negative value in the statistical distribution of G in an intragranular flux-trapping model was mathematically proved not only in the (Bi–Pb)–Sr–Ca–Cu–O system but for certain samples of the Gd–Ba–Cu–O as well.

In the same way, based on the new mathematical formulation of the problem, it was possible to obtain information about the statistical distribution of geometric factors directly from experimental data before the precise fitting of the virgin and returning curves. On the other hand, the long time of synthesis in the samples of the (RE)–Ba–Cu–O provokes the appearance of percolative paths with $G < 0$ which suggests the possibility of alignment in the grains but in a lesser degree that in the Bi ceramics which is not easily observed in other experiments. The (Bi–Pb)–Sr–Ca–Cu–O samples seem to have a quite similar granular structure (even when they have a different phase composition) because of their qualitatively similar $J_c(B_a)$ hysteretic behavior.

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