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THE EFFECT OF GRAINS SIZE AND TEMPERATURE ON SUPERCONDUCTING PARAMETERS*

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Abstract

A survey of published data on the effect of the grains size and temperature on magnetic parameters of $YBa_2Cu_3O_7$ is given. The magnetization values are divided into two major parts. 1. Intrinsic parameters such as: Hc_1, Hc_2, λ etc which are not affected by the grains size and 2. values such as: the shielded and Meissner fractions , the intragrain critical current density (J_c) and the demagnetization factor which reflect specific features of the real size of the granular ceramic superconductor. The close relation between the grains size and the second group of parameters is discussed.

Keywords: grains, intrinsic properties, superconductivity

Introduction

High T_c superconductors (HTSC) with the critical temperature (T_c) above 77 K have provided an unprecedented stimulus for solid-state scientists over the past ten years. All these compounds have a layered structure with CuO_2 planes. In general the crystal structures involve large unit cells in which point defects like oxygen vacancies and substitution of elements strongly affect the physical properties. The Cu ions carry a magnetic moment which may play a role in the appearance of superconductivity (SC), therefore, magnetic properties of these compounds are of particular interest. In this short paper we deal only with magnetic properties of the most famous and investigated $YBa_2Cu_3O_7$ (YBCO) compound with $T_c = 92$ K. Much of our information regarding (HTSC) has been obtained through magnetization measurements of granular ceramics. The magnetization values and the parameters extracted from them could be extremely difficult to handle, since the detailed magnetic structure of inhomogeneous samples is not perfectly clear. As a rule, these values are divided into two major parts:

1. The SC parameters such as T_c, Hc_1, Hc_2, λ etc., where Hc_1 and Hc_2 are the lower and upper critical fields and λ is the penetration depth, which are intrinsic properties of the material and independent of the grains size.

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2. Values such as: the shielded fraction (*f*), Meissner fraction, the intragrain critical current density (J_c) and the demagnetization factor *N* which reflect specific features of the real size of a superconductor. We survey some studies of the interrelation between the grains size and the magnetic properties of granular superconductors. Most of the studies described here were performed in our laboratory and we also review briefly several papers published by others which pertain to the same topic. The experimental details are found in the relevant papers.

Results and discussion

The effect of applied pressure on the shielded fraction (f)

The magnetization of sintered samples is governed by the effective demagnetization field seen by the sample. The calculation of this field could be extremely difficult, as the shape of the whole sample is generally and usually not ellipsoid, and there is a competition between the demagnetizing factors of the macroscopic sample on the one hand and that of the grains on the other. For cylinders, the calculation of the demagnetizing factors is not straightforward, since the magnetization is not homogeneous and in general, varies throughout the cylinder in both the radial and axial direction.

A further complication of the description of the magnetization of inhomogeneous granular ceramic superconductors is the field dependence of the shielded volume (*v*), which contributes to the diamagnetic signal. An increase of the applied field H_0 causes an increase of the amount of flux penetrating the weak links and grains and thus leads to a monatomic decrease of the volume that is shielded from the magnetic induction, *B*. This decrease is expressed in $M(H_0)$ curves by an increasing deviation from linearity, even below H_{c1} of the grains.

We have carried out [1] a detailed study of polycrystalline YBCO cylinders (radius *r* and thickness *c* of 0.25 cm) in a large range of packing factors, between 10 and 80%. The YBCO material was prepared by conventional solid–state reaction. In order to obtain the highly packed samples (77% samples 1, 2), powder of well defined YBCO was pressed at 9200 kg cm^{-2}. The samples with packing factor 65% (3 and 4) were obtained by pressing the powder at 1500 kg cm^{-2}. With the intention to control the size of the grains and the strength and nature of the weak links, all samples (1–4) were treated by different heat treatments which followed a 2 h sintering at 950°C. Samples 1 and 3 were cooled to 450°C at a slow rate of 10°C h^{-1}, while samples 2 and 4 were furnace cooled in this interval of temperatures (~100°C h^{-1} during the first hour). All the samples were further annealed at 450°C and furnace cooled to room temperature under flow of oxygen.

All samples (1–4) were zero–field cooled (zfc) down to 4.2 K and the magnetic moments were measured as a function of applied field H_0 up to 1000 Oe, in two perpendicular orientations H_0 parallel to $c(H_0 \| c)$, and parallel to $r\ (H_0 \| r)$]. Generally speaking, the field dependence of the magnetic moment of the entire volume (*M*) for all samples in both directions are quite similar.

Applying the Clausius–Mossotti approach to the problem of granular ceramic superconductors, a phenomenological model, described in details in [1], was suggested. This model was used to evaluate magnetization measurements of granular YBCO samples of different packing factors and to calculate the field dependence of the shielded fraction $f = v/V$ where V is the cylinder volume. If N and n are the demagnetization factors of the macroscopic cylinder and the grains respectively, then, the final equation describing f is:

$$f = M(1-n) / [M(N-n) - H_0/4\pi] \tag{1}$$

Figures 1(a) and 1(b) show the shielded fraction f of samples 1–4 for $H_0 \| c$ as a function of H_0, calculated using Eq. (1), with $n = 1/3$ and $N = 0.53$. One can immediately recognize the four different regimes in the curves.

(a) $H_0 < H_{c1}^{wl}$ (*wl* means weak links) The shielded fraction f of samples 1 and 3 (the samples that were cooled slowly after sintering) is constant $H_{c1}^{wl} \approx 10$ Oe Fig. 1 (b). For samples 2 and 4, which were obtained by cooling at a much faster rate, which does not allow the formation of weak links with homogeneous characteristics, the behavior is different and we do not find an interval of constant f and *a* well defined H_{c1}^{wl}. Note also, that f of samples 1 and 3 is smaller than that of samples 2 and 4. We interpret this result on the basis of difference in the concentration of shielded pores in the samples. Long heat treatments lead to coalescence of closed shielded pores to a larger, open and unshielded pore, and thus cause a lowering of the shielded fraction in this regime.

(b) $H_{c1}^{wl} \le H_0 \le H_{c2}^{wl}$ – This interval is characterized by a sharp decrease of f. The location of $H_{c2}^{wl} \approx 50$ Oe is constant and quite well defined for all samples Fig 1(b).

(c) $H_{c2}^{wl} \le H_0 \le H_{c1}^{grains}$ – The shielded fraction is nearly constant. The location of H_{c1}^{grains} is smeared due to in homogeneous grains' characteristics. In this field regime f hierarchy is such that:

$$f_{1500,\mathrm{fast}} < f_{1500,\mathrm{slow}} < f_{9200,\mathrm{fast}} < f_{9200,\mathrm{slow}}$$

where the subscripts are related to the pressure applied to the powder during pressing to the cooling rate. This reasonable result means that large pressing force and slow cooling rates increase the fraction of the grains in the total volume of the sample and their typical size [1]. SEM studies of the samples show similar hierarchy in the typical sizes of the grains (between 2 μm and 10 μm). The grains size studied here are much smaller than the London-penetration depth λ.

(d) $Hg \ge H_{c1}^{grains}$ – chacterized by a continuing decrease of f keeping the same hierarchy Fig 1 (a). (Hg is the effective field inside the grain.)

The obtained H_{c1}^{wl} and H_{c2}^{wl} values are consistent with values published by Senoussi *et al.*[2]. Note that all the critical fields are intrinsic properties which are independent of the grains volume as mentioned above.

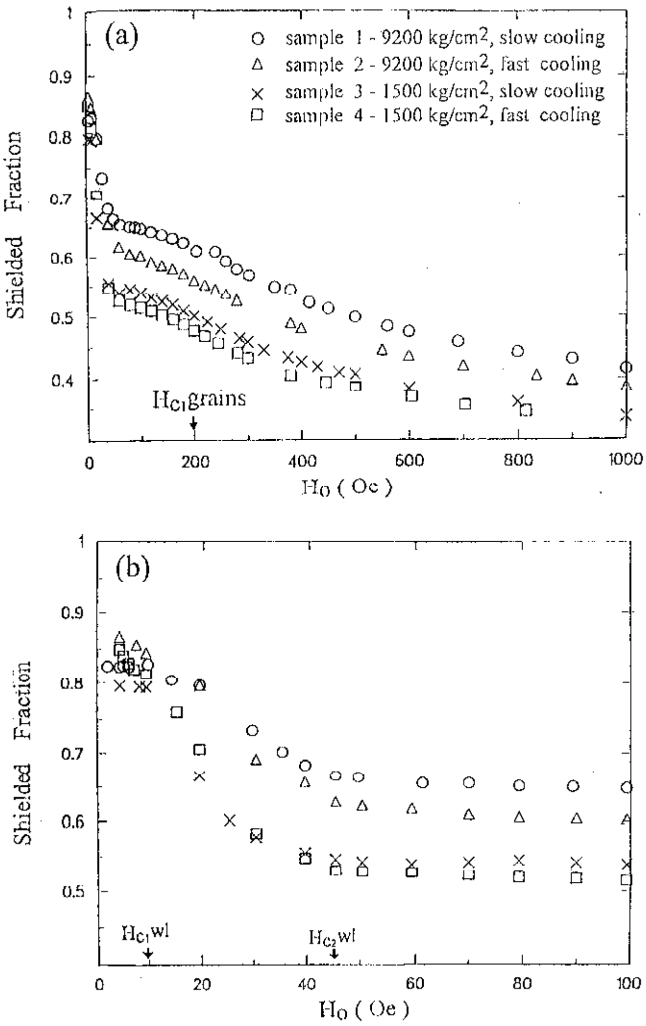

Fig. 1 The applied field dependence of the shielded fraction f for YBCO cylinder ($H_0 \parallel c$) calculated using Eq. 1 for (a) H_0<1000 Oe and (b) H_0<100 Oe

The effect of the grains size on the shielding curve

The influence of the intergranular connectivity on the field dependence of the shielding curve (*zfc* branch) in sintered YBCO pellet [3], which has been successively powdered to 200, 30, 1 and <0.5 μm size particle is shown in Fig. 2. The Gaussian-size distribution has been controlled to lie close to the peak, and the magnetic measurements were carried out maintaining the pellet and the powders in the same geometry. For the sake of comparison the data measured on a pellet

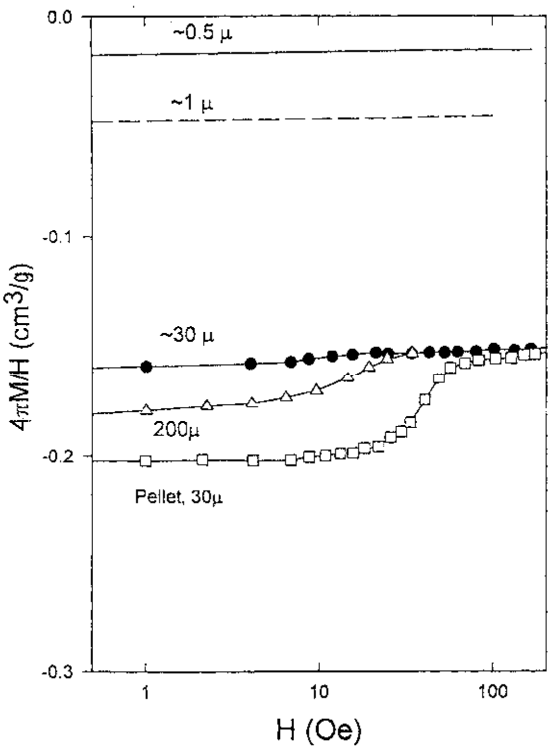

Fig. 2 *Zfc* curves for different grains size of YBCO powder

with an average grains size of 30 μm is also shown. The main interesting feature, is the sharp drop in the ZFC moment as the average particle size, a, increases and becomes compatible with the London-penetration depth, i. e. $\lambda/a \rightarrow 1$. When the particle size approaches λ the ZFC magnetization follows the well known behavior given by: $M_{\max}/M = 1-3\lambda_0/a^{*} \coth a/\lambda_0 + 3(\lambda_0/a)^2$ where λ_0 is 1600 Å [3].

The effect of grain size on intragrain critical current density J_c

Intragrain critical current density (J_c) is an important parameter which reflects specific features of the real structure of a superconductor. The J_c value at various temperatures and its field dependence are extremely important for practical applications, but for polycrystalline materials it is rather difficult to obtain the exact value. The most informative experiments for obtaining J_c for polycrystals involve measurements of $M(H_o)$ hysteresis loops at various temperatures. According to the well known Bean model, J_c in a given applied magnetic field is proportional to $\Delta M/D$ where D is the average grains' size. The hysteresis loops measured at 78 K for samples with an average grain sizes of I and 50 μm [4] are shown in Fig. 3.

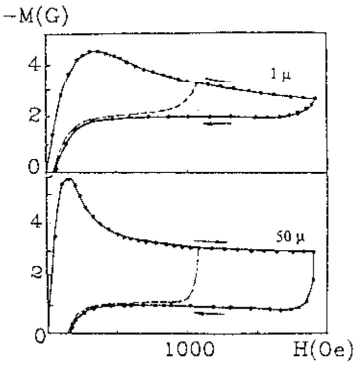

Fig. 3 Magnetization M *vs.* magnetic field H_0 for YBCO powder with different grains size measured at 78 K

Two features are readily observed. 1. The hystersis width ΔM is lower for the sample with small grain size. 2. The field dependence of width does not change at fields of about 500 Oe or higher for the coarse–grain sample, while in the case of fine–grained one it changes through all fields measured. Thus, Fig. 3 shows that the change in the grain size exerts influence over the behavior of the magnetization loops. The field dependence of J_c values calculated from these curves are higher by an order of magnitude for samples with 1 μm [4] than these calculated for the samples with 50 μm. It is thus assumed that the increase in J_c is caused by the effect of increase in the density of twinning boundaries. The main pinning centers at 78 K are twinning boundaries and their density increases by an order of magnitude when the average grain size decreases from 50 to 1 μm.

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