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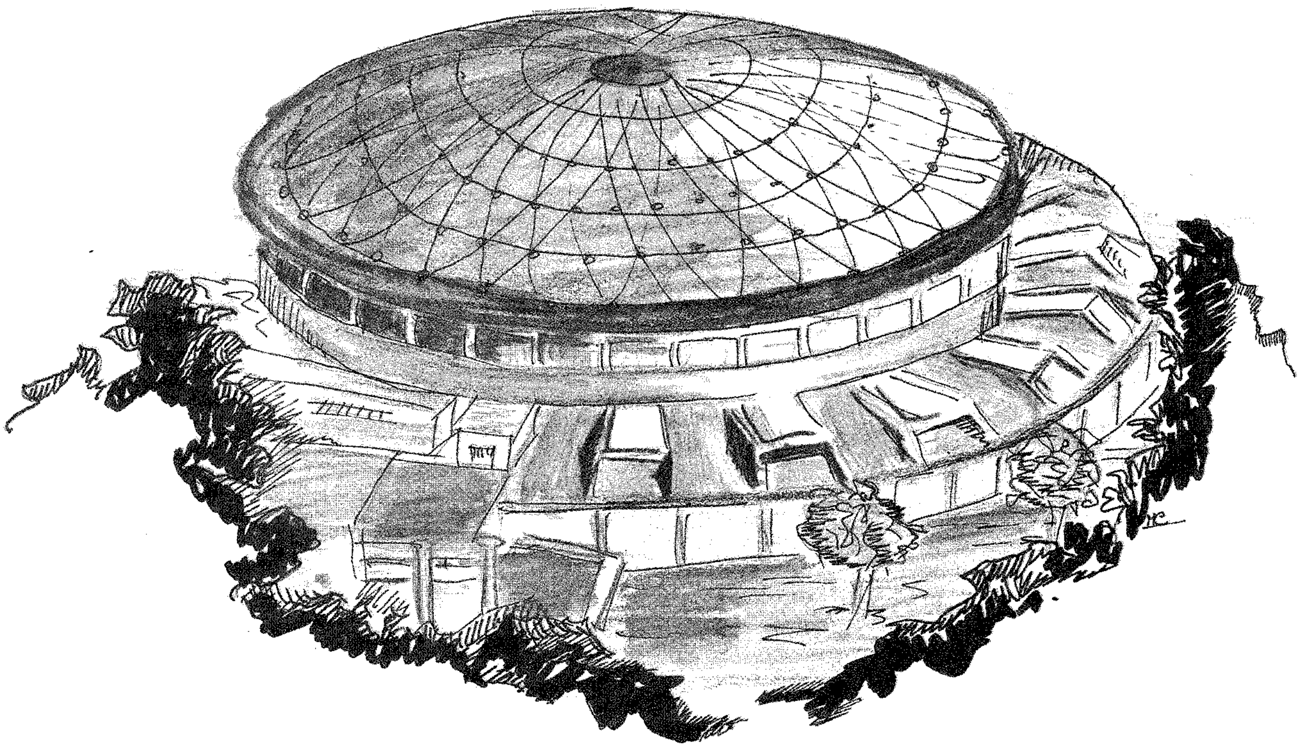

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TORQUE MEASUREMENTS OF TEXTURED $Y_1 Ba_2 Cu_3 O_{7-x}$ SINTERED PELLETS

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Abstract

We present a 4.2 K magnetic torque balance study of a textured sintered YBCO sample produced by pyrolysis using in the thermal treatments an ozone enriched oxygen atmosphere. The shape of the torque signal as a function of the angle and of the measuring field shows features that are intermediate between those of a standard sintered pellet and those of a single crystal. The anisotropic experimental values of the lower critical field H_{c1} rather well compare with those previously found on YBCO single crystals; the intensity of the remaining magnetization, measured after a field cycling procedure, suggests the existence of a strong pinning for $H \geq H_{c1}^{(c)}$ along the c axis.

Introduction

Sintered high T_c superconducting oxides have generally a very low critical current density, J_c that is limited by the weak coupling existing between the superconducting clusters of the granular material [1]. In thin crystalline films, on the other hand, the measured J_c is rather high but it has been recently shown to be very sensitive to the misalignment of the axis a, b or, even more, of the c axis between two next-neighbor microcrystals [2]. Thus bulk samples with oriented grains are a reasonable way to obtain useful sintered superconducting oxides. Indeed, in such a "new class" of sintered ceramics critical current as high as $1.7*10^4$ A/cm^2 have been achieved at 77 K.[3] These textured samples are generally highly dense (95%) and contain a large amount of tightly packed crystallite. As a consequence of the texture, their properties are strongly anisotropic; in particular, the magnetic ones can be studied by means of magnetic torque. Indeed this technique has been already successfully applied to the study of the anisotropy of the lower critical field H_{c1} and of the pinning forces [4] in a YBCO single crystal.

In this paper using this technique we have analyzed magnetic properties of sintered pellets, fabricated by a modified pyrolysis method [5]. In particular in our fabrication process during all thermal treatments an ozone enriched oxygen atmosphere has been used. In Sec.2 the main steps of the fabrication process are described. In Sec.3 the experimental results are reported which quite clearly show the presence of anisotropic properties of the magnetic behavior.

Sample fabrication

Samples are fabricated by a modified pyrolytic method. The main step of our standard procedure are :

1) drying separately of the industrial-grade (99% purity) powders of $BaCO_3$, CuO , Y_2O_3 on an oven for 12 h at 80 C,

2) weight check after drying for a correct stoichiometry,

3) grinding each powder by a magnetic agitator at 300 RPM for 5 min, with PTFE coated magnetic balls,

4) add slowy 65% concentrated HNO_3 , avoiding clots, to obtain nitrate solutions with an excess of nitric acid (for a total starting weight of 150 g we use 120 ml for $BaCO_3$ and 84 ml for both CuO and Y_2O_3),

5) put together all the three liquids and add 252 g of previously dried (as in step 1) citric acid. As always, we use a magnetic agitator for about 20 min at 700 RPM,

6) add slowly, 25% concentrated, NH_4OH until the PH reaches a critical value of about 6.8 ,

7) take each time 50 ml of reacted liquid and put into a 2 liters glass beaker. Warm-up the liquid until spontaneous pyrolysis begins,

8) fill with the ultra-fine reacted powder (50-100 nm) a high-quality alumina crucible and heat it into a furnace at 950 C for 12 h in an ozone enriched oxygen atmosphere. This gas has to flow until the cool down to room temperature (cooling rate 50 C/h),

9) grinding of the black material obtained just almost completely superconductor; filter at 150 MESH and press at about 10 KBar to obtain cylindrical pellets,

10) heat again like step 8.

As final result we obtain a very dense (5.5-5.9 gr/cm^3) homogeneous ceramic material that can withstand water or others typical previously forbidden stresses. The single phase material shows a good neutron diffraction pattern and very good diamagnetic and mechanical properties. Further details on the preparation procedure and physical characterization are reported elsewhere [6,7] .

We believe that the key points of this process are:

a) an optimum mixing obtained by liquid nitrate solutions,

b) the combustion that starts from a viscous state,

c) the ozone atmosphere.

In particular the use of O_2 enriched with few percent of O_3 gives large improvements to the physical properties of the material and in particular the magnetic ones: the presence of very reactive ozone easily allows the right oxygen stoichiometry. Moreover, we could hope to obtain some other, even unstable, compounds with higher transition temperature.

Details of oxygen exchange in our fabrication procedure and thermogravimetric analysis are in progress.

Experimental results

The torque measurements have been performed on a cylindrical sample (diamagnetization factor 0.1) at 4.2 K using as detector a capacitive cell [4] . The following experimental procedures have been used:

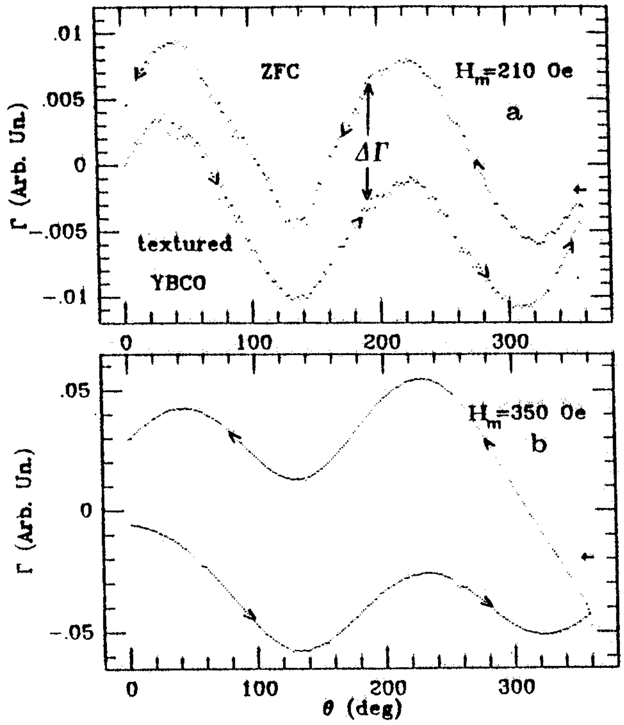
1) after zero field cooling (ZFC), the field is raised to a value H_m in the initial direction $\vartheta = 0$ and is then rotated at a constant speed from 0 to 360° and back (about 40°/min). The torque signal Γ is monitored permanently: $\Gamma = H_m \wedge M$, where M is the magnetization,

2) after ZFC the field is ramped up to the cycling value H_{cy} (maximum value 8.7 kOe) and then down to zero before raising the field to a low testing value (usually 50 Oe) H_m and rotating as in (1).

In Fig. 1 we show two typical recordings (a,b) of Γ as a function of ϑ obtained following protocol 1 on the textured sample and, for comparison, the qualitative behavior for respectively a completely disordered sample (c) and a single crystal (d).

In a disordered sample the only contribution to the torque Γ arises from the relaxating magnetization determined by the vortexes pinned into the sample by the external field H_m (Fig.1c). On the contrary in a single crystal a magnetization in a fixed direction, proportional to the component of the external field, generates a 180° periodic signal (Fig.1d) .

The observation of a well defined 180° periodic torque (Fig.1a,b) is the strongest evidence for the presence of a texture. This signal, in fact, is typical of the diamagnetic shielding of the plate crystallite contained in the sample [4] .



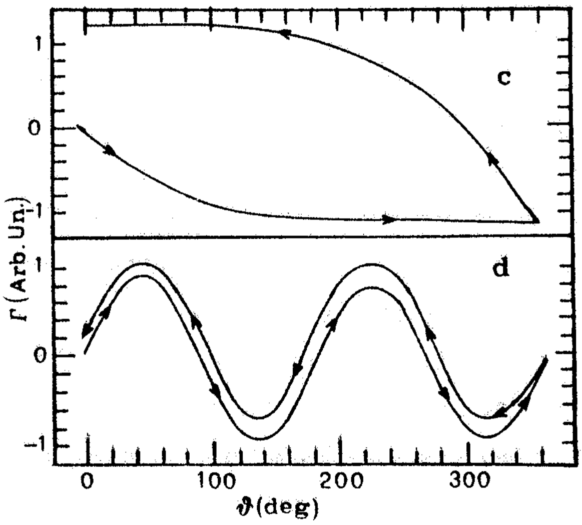


Fig. 1. Torque recordings after protocol 1 from a textured sintered sample (a) for H_m = 210 Oe and (b) H_m = 350 Oe; (c) and (d) are the qualitative behavior for respectively a completely disordered sample and a single crystal.

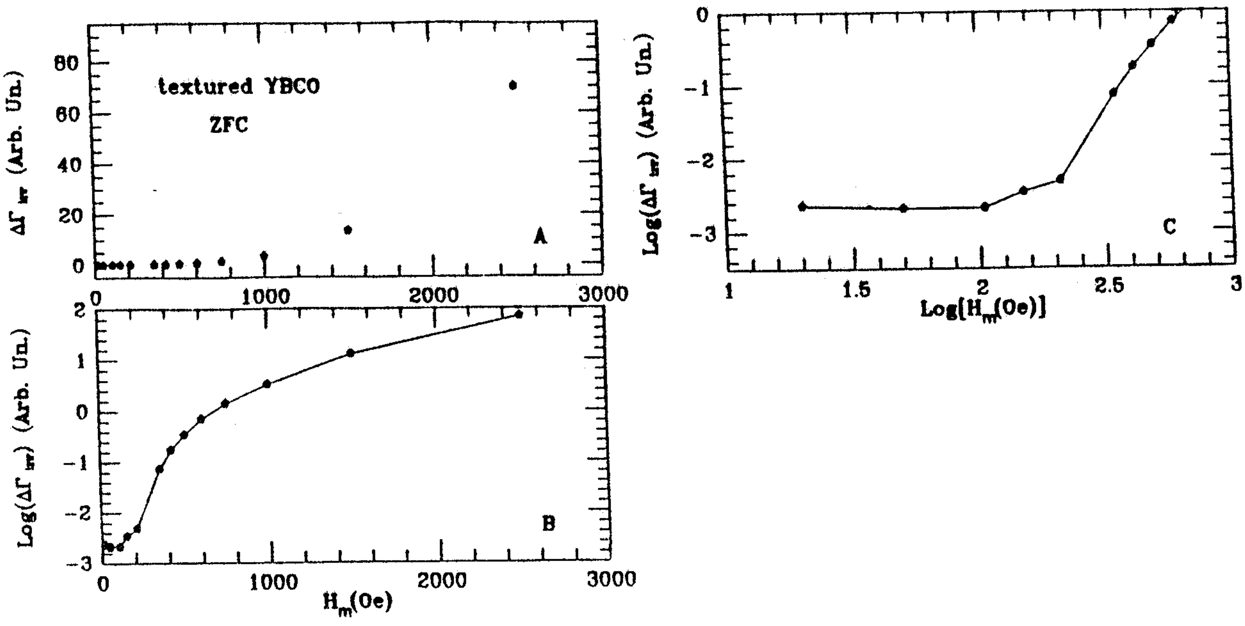


Fig. 2. Peak-to-peak hysteresis vs. H_m in lin-lin (A), lin-log (B) and log-log (C) scales.

The fact that forward and back torque curves are not superimposed is due to a small rotational hysteresis associated with the angular movement of the vortices that have already penetrated the sample. The maximum of the rotational hysteresis $\Delta\Gamma$ vs. H_m (the field values are not corrected for the demagnetization factor) measured following protocol 1 was plotted in Fig. 2, where the value of $\Delta\Gamma_{irr}$ has been taken as the peak to peak difference between the forward and the back torque curves. The logarithmic plots (Fig.2b,c) shows the existence of three different regimes: a) a weak rotational hysteresis regime between 20-100 Oe; b) a second regime defined by a first increase of such hysteresis between 100 and 300-350 Oe and finally c) a strong hysteretic regime associated with a massive penetration of the magnetic flux into the grains. It is worthwhile to point out that the plot of the integrated rotational hysteresis, instead of the peak-to-peak hysteresis would have made the crossover between the different regimes more evident. The low and constant hysteretic level observed for H_m<100 Oe is due to the saturation of the flux penetration in the intergranular regions, and it is absent only for H_m<25 Oe, which is the critical field for penetration in the weaklink regions [6,8] of our sample, under the condition of a true zero field cooling [1]. This hysteretic plateau is absent in single crystals [4] .

As soon as $H_m \geq 100$ Oe, we enter in the second regime. The increase of the rotational hysteresis is to be associated with the flux penetration along those a-b planes that are aligned with the direction of the applied field. Indeed, the values of the $H_{c1}^{(a\text{-}b)}$ lower critical field value observed in a single crystal range between 100 and 250 Oe. In this field range, however, especially for the lower fields, flux penetration in the defects of the grains along the c axis may contributes also to the torque signal so that the onset for flux penetration along the a-b planes can be partially smeared. Note, that in this regime, the torque behaviour is still dominated by the diamagnetic contribution due to the screening current looping on the a-b planes.

For $H_m \geq$ 300-350 Oe we detect the penetration of the magnetic flux along the c axis. In ordinary sintered samples the onset of this third regime is completely smeared out by the large distribution of critical fields. This rather low field value, 300-350 Oe, for flux penetration along c has to be associated only with the extreme tail of the $H_{c1}^{(c)}$ distribution values. Fig.3 shows a set of forward torque curves recorded following protocol 1 and allows us to follow the development of the flux penetration in the sample and that of the flux orientation along the c axis. The diamagnetic screening signal continuously shifts in angle, up to 750 Oe, indicating a redefinition of the flux pinning direction and of the associated screening current planes. From the torque measurements on single crystals we know that for field higher than $H_{c1}^{(c)}$ the torque signal arise from flux pinned along the c axis [4] ; as a consequence we deduce that for H≥750 Oe all the grains have been penetrated by the magnetic flux that is now pinned along the c axis; now we are in the condition to identify the direction of the preferential texture which can be

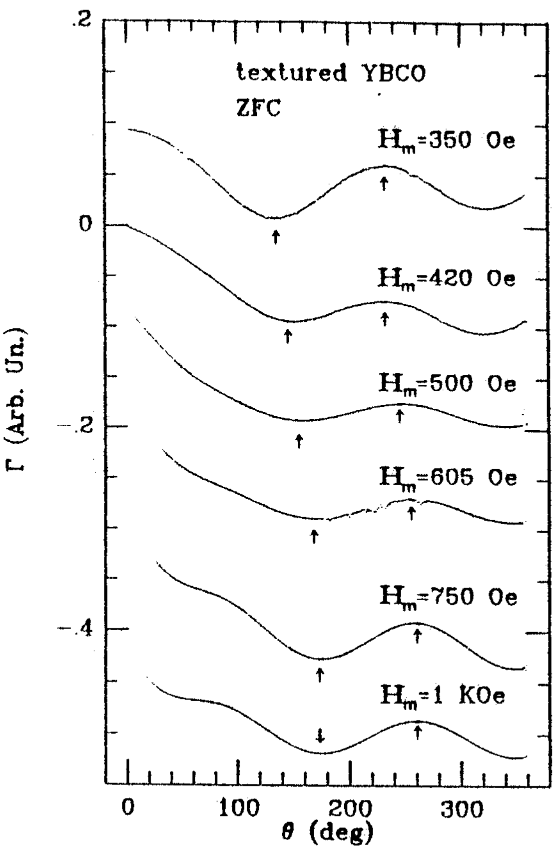


Fig. 3. Torque forward recordings for different H_m; note the shift of the peaks of the diamagnetic signal.

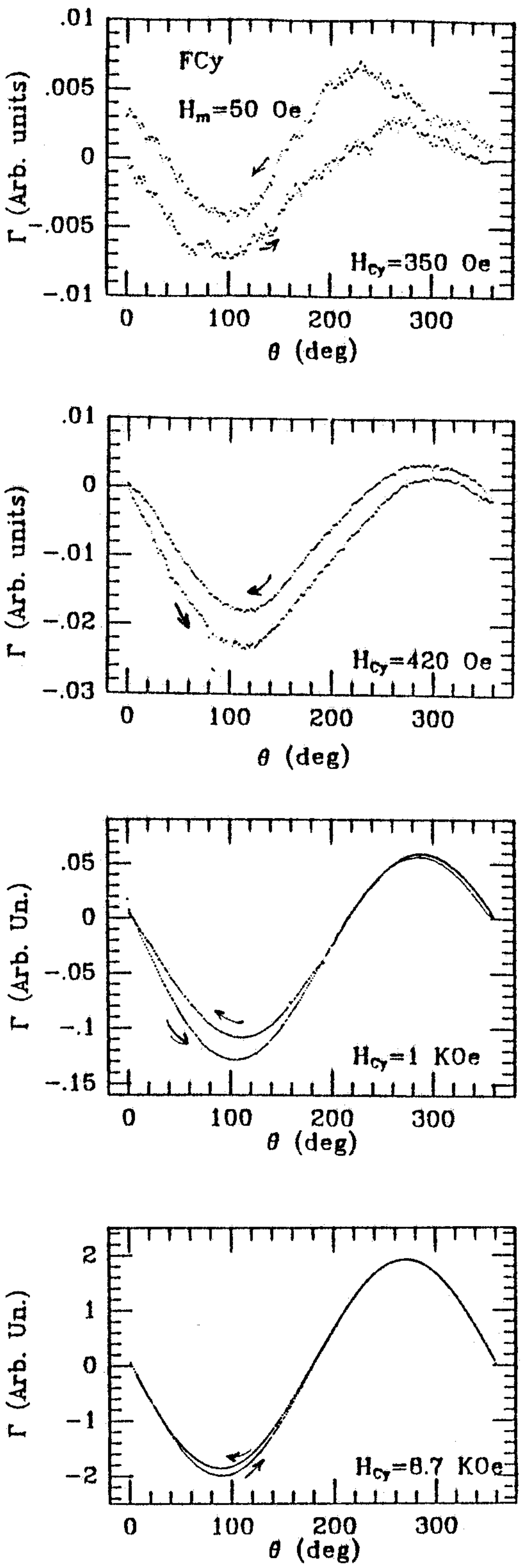


Fig. 4. Torque recordings following protocol 2 for different cycling fields; testing field 50 Oe.

In Fig. 4 we show a set of recordings following protocol 2 . The 360° periodic signal is characteristic of the pinning of the vortices in the initial direction not expelled by the sample during the ramping down of the field H_{cy} [9]. During the rotation of H_m the flux lines stay in their stable direction which can change only if the rotating field H_m is of the same order of magnitude or larger than H_{cy}.

In Fig. 5 we plotted the intensity of the sinusoidal signal vs H_{cy}. It is quite evident that pinning occurs for fields higher than 300-350 Oe, in agreement with the indication, obtained from the study of a single crystal, of a strong flux pinning along the c direction [4].

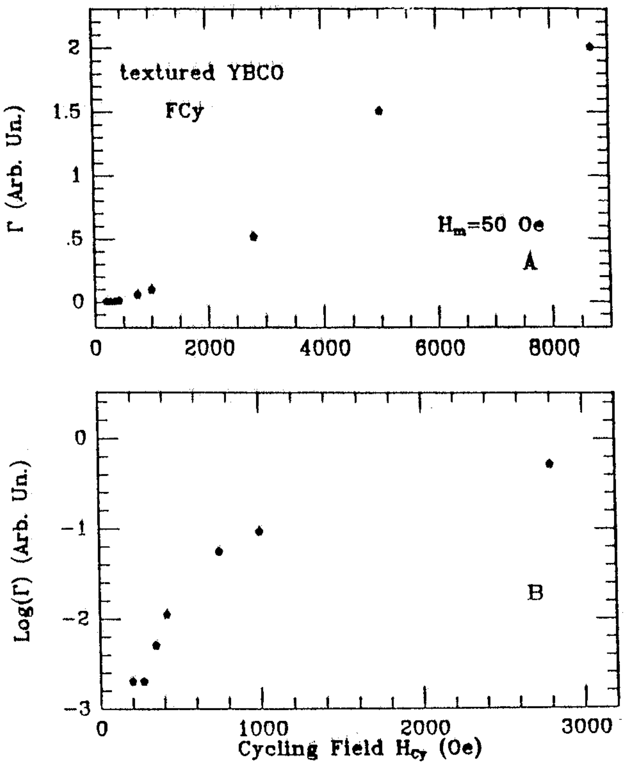


Fig. 5. Plot of the peak-to-peak torque signal (following protocol 2) vs. the cycling field intensity in lin-lin (A) and lin-log (B) scale.

Conclusion

The measurements performed by a magnetic torque balance clearly show an anisotropic behavior of sintered YBCO samples obtained by citrate pyrolysis fabrication methods using in any thermal treatment an ozone enriched oxygen atmosphere. Indeed experimental data are more similar to the data obtained with single crystal than to the results of disordered superconducting materials. This similarity shows the existence of microcrystals with some preferential orientation or texture.

Further analysis and experiments are still in progress in order to find the fabrication process conditions that generate or support the texture. Anyway we have a clear evidence that the use of ozone, in our fabrication process, improves in a large way the mechanical and electrical properties of samples.

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