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## Magnetic properties and thermal expansion of $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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### Abstract

Linear thermal expansion measurements have been performed up to 70 K with a capacitive dilatometer on a  $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$  polycrystalline sample which has been characterized by SEM micrography and magnetic Meissner effect. We report first some of these results which show the absence of any magnetically ordered impurity phase at low temperature in our sample. The thermal expansion also does not show the anomaly at 11 K previously reported, so we suggest that this anomaly was rather due to the magnetic ordering of an impurity phase than resulting from crystal field effects. © 1998 Elsevier Science S.A.

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### 1. Introduction

For many years, studies on the magnetic, thermal, transport and elastic properties of high temperature superconductors have been performed on polycrystalline and granular ceramic samples, mainly those of the family  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , because of their interesting properties due to the interplay between magnetism and superconductivity. The 4f electrons contribution to the low temperature thermal expansion of the  $\text{R}_2\text{CuO}_4$  series (R=Pr, Nd, Sm, Gd, Eu) have been investigated earlier [1]. The same approach has been made for the  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$  series showing the possible existence of a similar contribution to the thermal expansion coefficient [2]. Former measurements of this coefficient [3], made on  $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , have shown an anomaly around 11 K which seemed to confirm the proposed theoretical model. However, the authors of Ref. [3] mention that this anomaly may also be related to the magnetic ordering of an impurity phase also observed in specific heat measurements [4]. In order to clarify this point, new magnetic and linear thermal expansion measurements have been performed on a well characterised  $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$  polycrystalline sample.

### 2. Experimental

The sample, obtained by the standard ceramic method preparation, had a cylindrical geometry (length: 8.92 mm, diameter: 4.47 mm). To meet the dilatometry cell dimension requirements, a 5.0 mm length was carefully cut with a wire technique and the two opposite faces rectified to be parallel by polishing. The other part of a measured mass, 0.29775 g, was characterised by means of magnetic measurements. Using an extraction method in a dc superconducting magnetometer, we performed field cooled (FC) measurements under a magnetic field down to 5.0 Oe, where the sample was cooled under a field through  $T_c$  over a temperature range 100–4.2 K, and then, magnetisation measurements were made at 5.0 K with fields up to 1 kOe. The calibration of such low fields has been obtained precisely by measuring, in place of the sample, the signal given by a very pure paramagnetic sample made from gadolinium sulphate powder  $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$  (5N). SEM analysis on the sample has been made in order to discuss the low field dc Meissner effect data in connection with the grain size and also to check that the compactness is suitable for dilatometry. The linear thermal expansion measurements of the cylinder were carried out from 4 to 70 K using a very sensitive capacitive dilatometer as reported earlier [5], with a capacitance cell similar to one

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described previously [6]. For comparison with the present work, we also measured the corresponding yttrium compound  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  prepared exactly in the same conditions and our results agree well with those reported in Ref. [3].

### 3. Results and discussion

The data of the dc Meissner effect corrected for demagnetisation effect are shown in Fig. 1, where  $4\pi(M/H_{\text{eff}})$  is plotted as a function of  $T$  as the sample is cooled through  $T_c$  at  $H=5.0$  Oe. The values of  $M$  are obtained from the measurement of the total magnetic moment through division by the volume calculated from the sample mass and X-ray density. It shows an onset transition at 89.5 K with a transition width of  $\Delta T_c=3.1$  K and an 85% of flux expulsion at this field. This large amount of flux expulsion can be understood considering the small applied field and the large average size of the grains ( $10\text{--}20\ \mu\text{m}$ ) seen by the SEM analysis (Fig. 2a and b). In the ZFC magnetisation measurements (Fig. 3), the sample was cooled from above  $T_c$  to 4.2 K at zero field and  $M$  was recorded at  $T=5.0$  K as  $H$  increased up to 1 kOe (special care was taken to keep a small sweep-rate of 5 Oe below 500 Oe). Two slopes,  $\chi_1$  and  $\chi_2$ , can be observed in the diamagnetic state of the sample (Fig. 3) together with two different onsets of the deviation of the magnetisation curve from these slopes, giving two values of  $H_{c1}$ ; 40 Oe and 250 Oe respectively, as can be seen in the insert of Fig. 3. This result reflects the anisotropy of the lower critical field even well below  $T_c$ . In fact, the first value may be associated to the crystallites for which the field is oriented close to the  $a$ - $b$  plane (values of  $H_{c1}\leq 50$  Oe in the Cu-O planes have been reported in Ref. [7]), and the second one has the same order of magnitude than the value found in Ref. [8].

The measured thermal expansion coefficient  $\alpha=1/L\times$

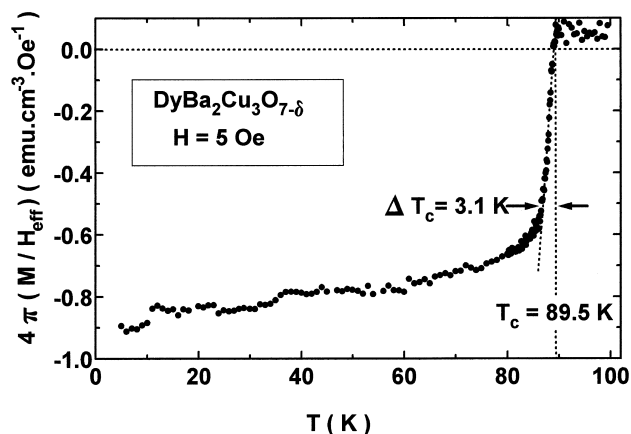
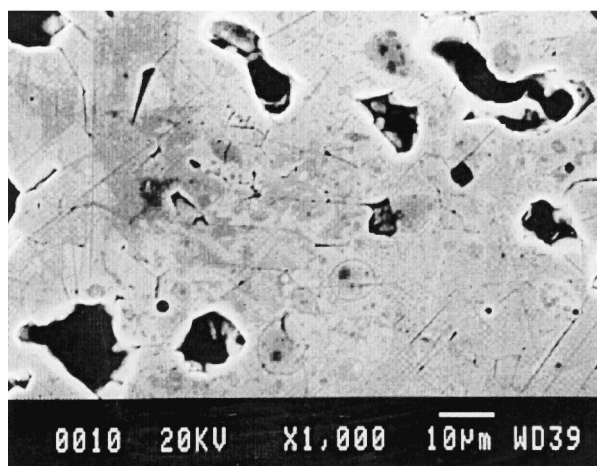


Fig. 1. FC susceptibility  $4\pi(M/H_{\text{eff}})$  as a function of decreasing temperature, for an applied dc field of 5.0 Oe (these data are corrected for the demagnetising factor).



(a)



(b)

Fig. 2. SEM micrographs of the  $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$  sample: (a) roughly polished, showing grain size in the  $10\text{--}20\ \mu\text{m}$  range; (b) well polished ( $1\ \mu\text{m}$ ), showing a low level of porosity (holes in dark black).

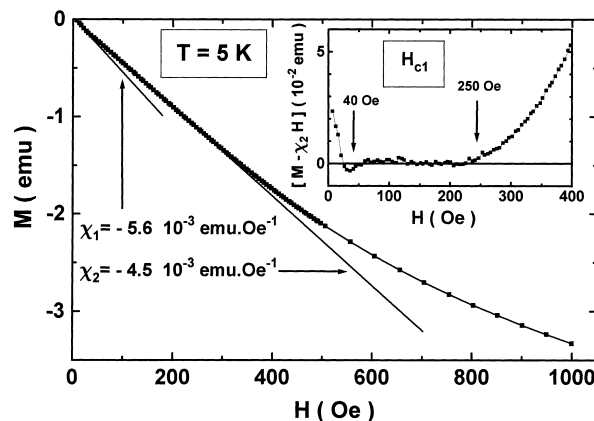


Fig. 3. ZFC magnetisation curve at  $T=5.0$  K (without correction of demagnetising factor). The lower critical fields,  $H_{c1}=40$  Oe and 250 Oe, are shown in the insert.

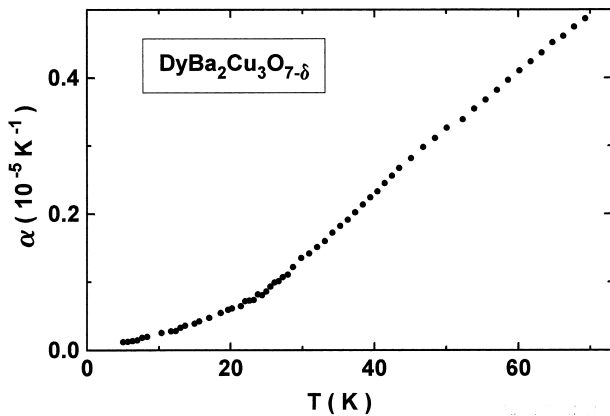


Fig. 4. Linear thermal expansion coefficient  $\alpha(T)$ .

$dL/dT$  is plotted versus  $T$  in Fig. 4. Within the accuracy of these measurements, no significant anomaly can be detected in the curve  $\alpha(T)$ , in contrast to the results reported in Ref. [3] which show a pronounced positive anomaly reaching  $10^{-6} \text{ K}^{-1}$  near 11 K. Moreover, from our data, a plot of  $\alpha$  vs  $T^3$  is almost linear up to 30 K for  $\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , as well as for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , indicating that in both cases the phonon contribution is almost the only one present. After all these results, one can deduce that our sample does not contain any magnetic ordered impurity phase that may be formed during the elaboration of such ceramic samples, mainly  $\text{Dy}_2\text{BaCuO}_5$  (green phase) studied earlier in Ref. [9]. So, one can suggest that the anomaly reported in Ref. [3] originates mainly from the presence of such a phase. In order to confirm this point of view, it would be interesting to perform some specific heat measurements on our sample, and thermal expansion measurements on the magnetic compound  $\text{Dy}_2\text{BaCuO}_5$ .

In conclusion, the existence of a 4f contribution to the thermal expansion coefficient of the compound

$\text{DyBa}_2\text{Cu}_3\text{O}_{7-\delta}$  cannot be completely excluded, but, from the results reported herein, it might be very small and hard to detect within the sensitivity of our experimental setup or other similar dilatometers [3,6] and it would require performing these measurements on a single crystal.

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