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# Anisotropy of the magnetization of single crystals and bulk high-$T_c$ superconductors

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Measurements of the angular dependence of the zero-field-cooled (ZFC), field-cooled (FC) and remanent magnetization of both high-$T_c$ single crystals and polycrystalline samples show significant anisotropy. Comparison of the results obtained on both, prove that most anisotropy is governed by the shape of the sample via the demagnetizing factors. For ceramic samples, there is competition between the demagnetizing factors of the sample as a whole, N, and that of the grains, n. According to a suggested phenomenological model, the magnetic moment is related to the external field by a relation involving an effective demagnetizing factor, $N_{eff}$, which depends on the fraction of superconducting volume, which is itself a field dependent quantity. At low fields, $N_{eff}$ approaches N and n for highly compact and dilute samples, respectively Between these extremes, $N_{eff}$ is a weighted average of N and n.

The crystallographic asymmetry of the Cu-O layered superconducting compounds is clearly manifested in strong anisotropies of their physical properties In this paper we shall present results of ZFC, FC and remanent measurements and address the question of the origin of the observed anisotropy

## 1. EXPERIMENTAL DETAILS

Measurements have been performed on several Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O single crystals and Y-Ba-Cu-O polycrystalline cylinders of different aspect ratios, using a PAR 155 Vibrating Sample Magnetometer, which enables a rotation of the sample relative to the direction of the external field, $H_0$. The details regarding the measured samples were described in previous papers[1-3]. The experimental procedures are as follows. The sample was cooled to 4 2 K either in a field (FC) or in a nominal zero field (less than 3 Oe) The "cooling angle", $\phi$, is the angle between **H** and the **c** axis during the cooling process. In the ZFC measurements a field was subsequently applied at 4 2 K and $M_{ZFC}$ was measured as a function of the angle $\theta$ between **H** and **c**. The axis of rotation was parallel to the **ab** planes. In the FC process the sample was rotated in the cooling field and $M_{FC}$ was obtained as a function of $\theta$. Similar rotation of a field-cooled sample in the absence of external field yields $M_{rem}(\theta)$.

## 2. RESULTS AND DISCUSSION

Fig. 1 exhibits the angular dependence of $M_{ZFC}$, $M_{FC}$ and $M_{rem}$ of an Y-Ba-Cu-O single crystal as a function of the rotation angle, $\theta$, for $\phi$=0° and $\phi$ =60°. The dotted line through the ZFC results is a fit to

$$(1) \quad M_{ZFC}(\theta) = -\frac{H_0}{4\pi}\left(\frac{\cos^2\theta}{1-N_c} + \frac{\sin^2\theta}{1-N_b}\right),$$

where $H_0$ is the external field and $N_{b,c}$ are the demagnetizing factors of the $\theta$=0° and $\theta$=90° directions, respectively. $N_{b,c}$ were extracted from conventional tables[4], approximating the crystal to be of an ellipsoidal shape. The independence of $M_{ZFC}$ on $\phi$ suggested by Eq. 1 was experimentally confirmed. This result implies that this measurement probes only the angular dependence of the demagnetizing factor and not any intrinsic property. For the crystal measured in Fig. 1 we obtain $M_{ZFC}{}^{c}/M_{ZFC}{}^{ab}=(1-N_b)/(1-N_c)\approx 5$.

The dotted line through the $M_{rem}$ results is a fit to

$$(2)\quad M_{rem}(\theta,\phi) = M^{c}_{rem}\cos\theta\cos\phi + M^{ab}_{rem}\sin\theta\sin\phi,$$

where $M_{rem}{}^{c}$ and $M_{rem}{}^{ab}$ are the initial remanent values obtained for $\phi=0°$ (**H**||**c**) and $\phi=90°$ (**H**||**ab**), respectively. For the crystal measured in Fig 1 we obtain an anisotropy ratio $M_{rem}{}^{c}/M_{rem}{}^{ab} \approx 5$ Other Y-Ba-Cu-O single crystals show similar anisotropy The Bi-Sr-Ca-Cu-O single crystal showed larger anisotropy, $\approx 50$. This anisotropy demonstrate the tendency of the irreversible magnetization, resulted by pinning of vortices, to be oriented essentially parallel to the c axis.

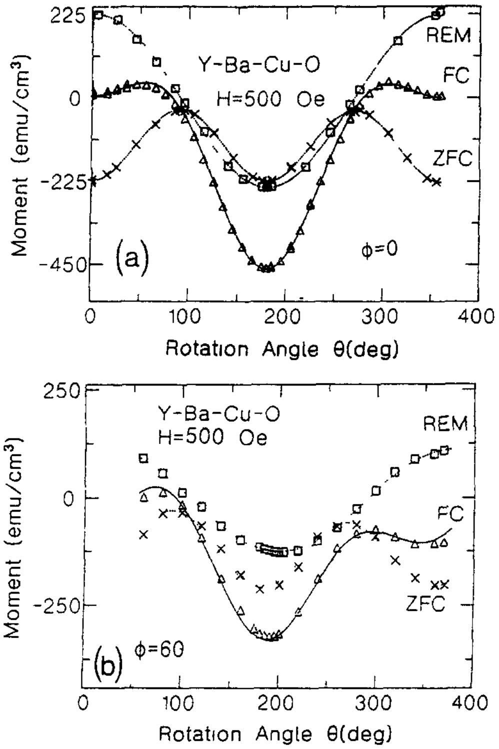

Fig. 1. Angular dependence of the remanent, zero-field-cooled and field-cooled magnetization of the Y-Ba-Cu-O single crystal at 4.2 K and $H_0$=500 Oe, for cooling angles (a) $\phi=0°$ and (b) $\phi=60°$. The solid line is the sum $M_{ZFC}+M_{rem}$.

The solid line is the sum of the $M_{ZFC}$ and $M_{rem}$ curves. The $M_{FC}$ results are marked by triangles. The fit is good as long as the field and temperature are low enough, resulting a rigid rotation of the trapped flux. This relation between $M_{FC}$, $M_{ZFC}$ and $M_{rem}$ holds for all $\phi$.

Proving that the ZFC magnetization is governed only by the anisotropic shape of the crystal and that the FC moment is just the sum of $M_{ZFC}$ and $M_{rem}$, we are left with the question of the origin of the anisotropy of $M_{rem}$. Remanent measurements probe the anisotropy of flux trapping and pinning If we are observing an intrinsic property, then our measurements support the phenomenological effective mass tensor approach[5], which predicts an "easy" direction for vortices parallel to the c axis. The opposing intrinsic pinning approach[6] predicts an "easy" direction parallel to the ab planes, due to strong pinning of the layered structure.

It might be argued that demagnetizing corrections are negligible in remanent measurements, since flux trapping occurs in the vicinity of the irreversibility line, where M is small, and the measurement is performed in the absence of external field. However, the following simple mathematics, suggests otherwise. During the cooling process

$$\begin{aligned}(3)\quad B_{FC} &= H_i + 4\pi M_{FC} = \\ &= H_0 - 4\pi N M_{FC} + 4\pi M_{FC} = \\ &= H_0 + 4\pi M_{FC}(1-N),\end{aligned}$$

where $H_i$ is the internal field After turning the field off, $H_0$=0, assuming all the flux remains pinned B remains $B_{FC}$ and the remanent moment will generate its own demagnetizing field, $-4\pi N M_{rem}$. Inserting into Eq. 3, we obtain

$$(4)\quad B_{FC} = 4\pi(1-N)M_{rem},$$

and thus

$$(5)\quad M_{rem} = B_{FC}/(4\pi(1-N)).$$

It seems, therefore, that $M_{rem}$ might depend on the external shape through its dependence on the demagnetizing factors.

Following the above mentioned argument and the Comment of Kolesnik et al.[7], we have performed similar measurements on polycrystalline cylinders of different aspect ratios[3]. These samples were characterized by SEM and XRD, and were found to be granular with no preferred orientation. We found for this samples that

$$\frac{M^c_{rem}}{M^r_{rem}} = \frac{M^c_{ZFC}}{M^r_{ZFC}} < \frac{1-N_r}{1-N_c} \tag{6}$$,

where $N_c$ and $N_r$ are the demagnetizing components in the directions parallel to the axis of the cylinder and to its radius, respectively, extracted from tables[4], assuming the cylinders to be of an ellipsoidal shape. The first equality of Eq. 6 was previously observed to hold for single crystals. The identical results observed on both single crystals and polycrystalline samples, prove that the observed anisotropy is caused by the anisotropic external shape of the samples and is not an intrinsic property. Similar results have been recently reported by Hellman et al.[8] Their measurements on polycrystalline isotropic type II superconductors show that the irreversible magnetization is oriented essentially parallel to the smallest dimension and is almost independent of the applied field direction. They explain their results using the critical state model, noting that the external shape puts constraints on the possible direction of flow of the current loops.

The influence of the external shape was excluded in the reports of Kolesnik et al.[9] and Liu et al.[10]. Kolesnik et al. have measured $Pb_2Sr_2Y_{0.3}Ca_{0.7}Cu_3O_8$ single crystals, that tend to grow relatively well in the c direction. An anisotropic 1.06x0.95x0.28 mm crystal showed a behavior similar to that observed by us on Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O In a 1.06x0.25x0.27 mm single crystal the influence of external shape was excluded This crystal show higher initial remanent for **H||ab**, supporting the intrinsic pinning approach Liu et al. have measured grain oriented Y-Ba-Cu-O samples. Taking into account demagnetization corrections, they find a c-axis preference. It is possible that the results of the two works could be highly affected by the presence of imperfections in the measured samples, and do not probe the crystal structure anisotropy. This might explain the discrepancy in their conclusions

The experimental anisotropy ratio of the remanent and ZFC measurements of single crystals, was found to be equal to $(1-N_b)/(1-N_c)$ For polycrystalline cylinders, the experimental ratio was found to be smaller than $(1-N_r)/(1-N_c)$ (Eq. 6). This inconsistency is quite obvious, since polycrystalline samples are not homogeneous and the macroscopic dimensions of the sample are not the only relevant dimensions. Calculations by Taylor[11] for homogeneous cylinders predict that its anisotropy should be smaller than that of an ellipsoid of the same aspect ratio. However, Kunchur and Poon[12] extracted the demagnetizing factors of homogeneous lead and Nb-Ti from magnetic measurements, concluding that applying the ellipsoidal approximation to cylinders of small aspect ratios is justified. It seems that the inequality in Eq. 6 results from the granular structure of polycrystalline samples, in which there is a competition between the demagnetizing factors of the external shape on the one hand and that of the grains on the other. Applying the Clausius-Mossotti approximation, we have suggested a phenomenological model, which accounts for the granular nature of polycrystalline samples. The details of the model have been recently published[3]. The model relates the measured magnetic moment, M, to the external field, $H_0$, using an effective demagnetizing factor, $N_{eff}$.

$$M = -\frac{H_0 - B_s}{4\pi}\,\frac{f}{1 - N_{eff}} \tag{7}$$,

where

$$N_{eff} = fN + (1-f)n \tag{8}$$,

where N and n are the demagnetizing factors of the external shape and grains, respectively, $B_s$ is the induction in the superconducting elements ( it is negligible, as long as the field inside the grains is smaller than $H_{c1}{}^{grains}$) and f is the superconducting fraction ( the ratio of the volume of the superconducting elements to the total volume). In a case of f=1 one gets that $N_{eff}$=N, while for f<<1 one obtains, $N_{eff}\approx$ n. This result implies that for highly compact samples, $N_{eff}$ is imposed by the macroscopic shape, and for dilute samples it is governed by the shape of the individual grains. This result was observed experimentally by Senoussi et al.[13]. It should be noted that f is a field dependent quantity and therefore, $N_{eff}$ is, as well This model explains the field dependence of the ratio of the moments measured in the direction parallel and perpendicular to the axis of the cylinder, presented in Fig. 2. At very low fields, f is largest, $N_{eff}$ approach N, and thus the measured anisotropy approaches $(1-N_r)/(1-N_c)$. The drastic drop of f between $H_{c1}{}^{wl}$ (≈10 Oe, wl - intergranular weak links ) and $H_{c2}{}^{wl}$ (≈50 Oe) causes a drastic drop in the anisotropy ratio. At fields exceeding $H_{c2}{}^{wl}$ this ratio is essentially constant and stays so even at fields exceeding $H_{c1}{}^{grains}$ (≈ 200 Oe)

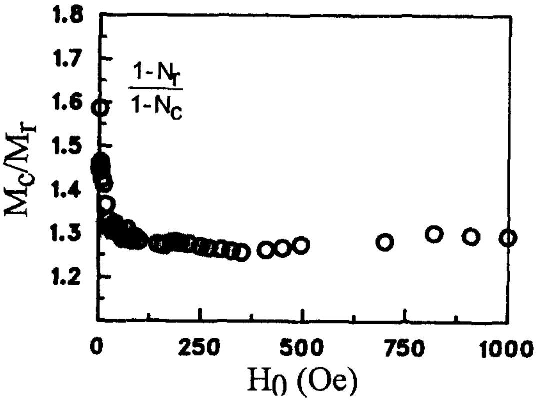

Fig 2. The applied field dependence of $M_c/M_r$ of a polycrystalline cylinder Note that at low fields the ratio approaches $(1-N_r)/(1-N_c)\approx 1.6$ The drastic drop occurs between $H_{c1}{}^{wl}$ and $H_{c2}{}^{wl}$ At fields exceeding $H_{c2}{}^{wl}$ the anisotropy ratio is essentially constant.

## 3. CONCLUSIONS

### *a. Regarding single crystals*

1) The angular dependence of the ZFC magnetization is governed only by the shape of the crystal.

2) The angular dependence of the remanent magnetization is substantially affected by the shape of the crystal. The remanent moment is preferably oriented parallel to the smallest dimension.

3) At sufficiently low fields and low temperatures the FC moment rotates with the crystal as a rigid entity, and the relation $M_{FC}=M_{ZFC}+M_{rem}$ holds for all orientations.

4) No decisive conclusion regarding the preferred flux trapping direction of an utopian defect-free single crystal

### *b. Regarding granular samples*

1) Magnetization results of granular samples should be treated using an effective demagnetizing factor, $N_{eff} = fN + (1-f)n$.

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## REFERENCES

1. I. Felner et *al.*, Phys. Rev. B, 40 (1989) 5239.
2 U Yaron et *al*, Phys. Rev B, 44 (1991) 12531 .
3 U. Yaron et *al*, Phys. Rev B, 46 (1992) 14823.
4. J. Osborn, Phys. Rev., 67 (1945) 351 .
5 J.R. Clem, Physica C, 162-164 (1989) 1137; V.G. Kogan, *ibid* 1689; V.G. Kogan Phys. Rev. B, 38 (1988) 7049
6. M. Tachiki and S. Takahashi, Solid State Commun., 70 (1989) 291
7 S Kolesnik et *al.*, Phys. Rev. B, 43 (1991) 13679
8 F. Hellman et *al.*, Phys. Rev. Lett., 68 (1992) 867.
9 S Kolesnik et *al.*, Phys. Rev B, 45 (1992) 10158
10 L Liu et *al*, Phys Rev. B, 43 (1991) 11418; L. Liu et *al.*, Phys. Rev. B, 45 (1992) 3054.
11. T.T Taylor, J. Res. Nat. Bur. Stand., 64B (1960) 199
12 M.N. Kunchur and S J. Poon, Phys. Rev. B.,43 (1991) 2916
13. S. Senoussi et *al*, Physica C, 165 (1990) 364 .