Field-induced anisotropy in high-T_c superconductors

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We have studied the angular dependence of the magnetization of $YBa_2Cu_3O_7$ by rotating the sample relative to the applied magnetic field. At low temperatures and low fields the irreversible part of the field-cooled magnetization M_{irr} rotates with the sample as a rigid entity. The rigidity is broken and part of M_{irr} disappears above an angle ϕ^* which decreases with increasing either temperature or field. This behavior resembles that of spin glasses and differs qualitatively from the behavior found for Nb, a type-II superconductor.

Since Bednorz and Müller[1] reported on the possibility of a high-T_c superconductor we have witnessed frequent discoveries of new materials with high transition temperatures.[2] In the effort towards understanding and characterizing the features of the new superconductors an enormous amount of work has been devoted to their magnetic properties. One of the intriguing features of these materials found in magnetic measurements[3–5] is the striking resemblance to the magnetic properties of spin glasses.[6] For example, the zero-field-cooled (ZFC) branch of the magnetization is irreversible, metastable, and is lower than the field-cooled (FC) branch (i.e., it is more diamagnetic in the case of superconductors). These spin-glass features motivated the present experiment. In spin glasses it is well known that the remanent magnetization is coupled to the macroscopic anisotropy and can be rotated as a rigid body,[7,8] but nonrigid rotations are observed for relatively high fields and temperatures.[9] We report here on similar features in a high-T_c superconductor. We have studied the angular dependence of the FC magnetization of $YBa_2Cu_3O_7$ by rotating the sample relative to the applied magnetic field. We find that at low temperatures and low fields the irreversible part of the magnetization M_{irr} rotates with the sample as a rigid entity. At higher fields and temperatures, the rigidity is broken and part of the M_{irr} disappears above an angle ϕ^* which decreases with increasing either temperature or field. These results resemble the behavior in spin glasses and are qualitatively different from those observed in the low-temperature type-II superconductor, Nb.

The sample was prepared from a mixture of $BaCo_3$, Y_2O_3, and CuO powders (at least 99.9% pure) in stoichiometric proportion according to the formula $YBa_2Cu_3O_7$. Finely ground powders were pressed into a pellet approximately 1.5 cm in diameter, and heated to 900 °C for 16 h in flowing oxygen. The product was then quenched to room temperature, reground, and heated again to 900 °C for 48 h, then cooled to ambient temperature. Powder x-ray diffraction shows that most of the observed lines could be indexed with the orthorhombic cell with lattice constants $a = 3.822$ Å, $b = 3.891$ Å, and $c = 11.67$ Å in fair agreement with published data.[10]

The angular dependence of the magnetization was investigated via measurements of the magnetization on a vibrating sample magnetometer (VSM) with a 2π-rotating sample holder. In the FC measurements the sample was cooled in an applied field H from well above the transition temperature T_c to the measuring temperature, which is stabilized to better than 0.1 K. With the same field on, the sample was rotated by an angle ϕ relative to the applied magnetic field. We then measured the magnetization M as a function of ϕ. In the ZFC measurement the sample was cooled in zero-applied field and then H was switched on, remaining constant during the measurement of $M(\phi)$.

Figure 1 exhibits typical $M(\phi)$ data for FC and ZFC runs in a field $H = 117$ Oe. The most obvious feature in this figure is the strong angular dependence for the FC magnetization. To understand this feature we recall that in VSM measurements the measured magnetization M is the projection of the total sample magnetization **m** on the

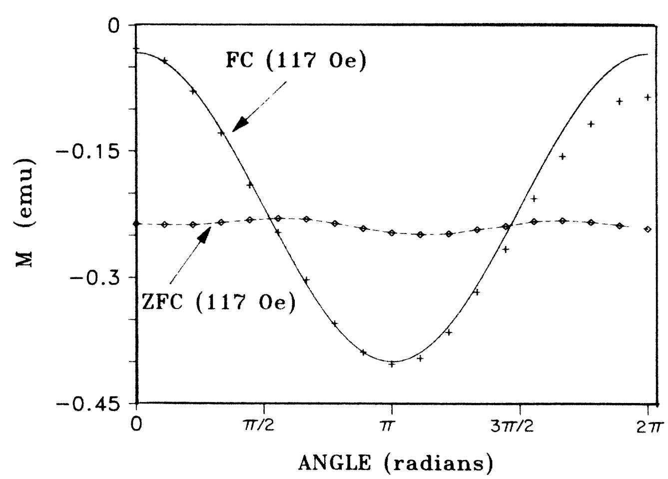

FIG. 1. Angular dependence of the field-cooled (FC) and zero-field-cooled (ZFC) magnetization in $YBa_2Cu_3O_7$ at 4.2 K for $H = 117$ Oe. The solid line among the FC data points is a result of a least-squares fit according to Eq. (2) (see text).

direction of the field **H**. The magnetization **m** is composed of reversible and irreversible components:

$$\mathbf{m} = \chi\mathbf{H} + \mathbf{M}_{irr} . \tag{1}$$

In the polycrystalline $YBa_2Cu_3O_7$ the bulk susceptibility is of course isotropic and thus the reversible contribution χH is always in the direction of H. The irreversible part, M_{irr}, on the other hand, might be coupled to the sample and rotate with it. It should also be taken into account that as a result of the field M_{irr} might lag by an angle θ relative to the sample for a rotation ϕ of the sample. Thus, the measured contribution of the irreversible magnetization is expected to be $M_{irr}\cos(\phi-\theta)$ where $\theta \leq \phi$ and the measured magnetization is

$$M = \chi H + M_{irr}\cos(\phi-\theta) . \tag{2}$$

The solid line in Fig. 1 is the result of a least-squares analysis of the FC data according to Eq. (2) which yields $\phi = 0$ implying a strong anisotropy. Note the perfect fit except for slight deviations at a high angle of rotations. It is important to note that $M(\phi)$ is reversible, i.e., the sample is rotated from 0 to 2π and back to 0 and $M(\phi)$ is only slightly altered.

Another important feature displayed in Fig. 1 is that the variations in $M(\phi)$ for the ZFC data are much less significant. This means that M_{irr}, associated with the ZFC magnetization, is small compared to χH. The fact that M_{irr} is not zero is evident from the remanent magnetization, measured directly by switching off the field. Moreover, the value of χ obtained from the fit for the FC data is within 10% of the average value of the ZFC data. The measured variations in $M(\phi)$ for the ZFC data exhibit a $\cos^2\phi$ contribution. This effect is hardly seen in Fig. 1 due to scale reduction, but it is more obvious in the following figures. Most of this contribution is due to demagnetization effects which mask the actual angular dependence of the small irreversible contribution.

Figure 2 displays the effect of the applied magnetic field on the angular dependence of the field-cooled magnetization. For the convenience of representation we present M/H data. Generally speaking, all curves exhibit the same shape. However, note that $M(2\pi) < M(0)$ and this difference is more pronounced for the higher fields. More important, $M(\phi)$ is no longer reversible. This is demonstrated in the lower part of Fig. 3 where we exhibit the angular dependence of the magnetization at 4 K for $H = 1500$ Oe. At an angle ϕ^* (which decreases with increasing field and temperature) there is a total breaking of the rigid moment and the FC curve coincides with the ZFC one. Moreover, the FC signal is not recovered when the sample is rotated back from 2π to 0, demonstrating the complete disappearance of the original irreversibility.

In the low-field regime, the asymmetry in $M(\phi)$ is important only at a high angle of rotations (see Fig. 1). Thus, using Eq. (2), we can deduce M_{irr} directly from the raw data by taking $M_{irr} = [M(0) - M(\pi)]/2$. The inset of Fig. 2 exhibits the field dependence of M_{irr}. Note the resemblance of $M_{irr}(H)$ to the thermoremanent magnetization (TRM) in spin glasses.[6]

The effect of the field on the rigidity of M_{irr} is more dramatic at high temperatures. This is already obvious from Fig. 3 where we demonstrate that the effect of 45 Oe at 67 K is qualitatively similar to that of 1.5 kOe at 4.2 K. Further evidence of the temperature effect is displayed in Fig. 4 where we exhibit the angular dependence of magnetization [normalized to $M(0)$] at 67 K for various fields. The initial $\cos\phi$ dependence is abruptly interrupted at a relatively small angle ϕ^* which decreases with the increase of H. Similar to the behavior at 4.2 K (Fig. 3) the backward angular dependence is reversible and is similar to the ZFC data.

To complete the experimental description, we compare the results with FC data for Nb (Fig. 5). The figure exhibits $M(\phi)$ dependencies for Nb which resemble those of $YBa_2Cu_3O_7$. There are, however, two important differ-

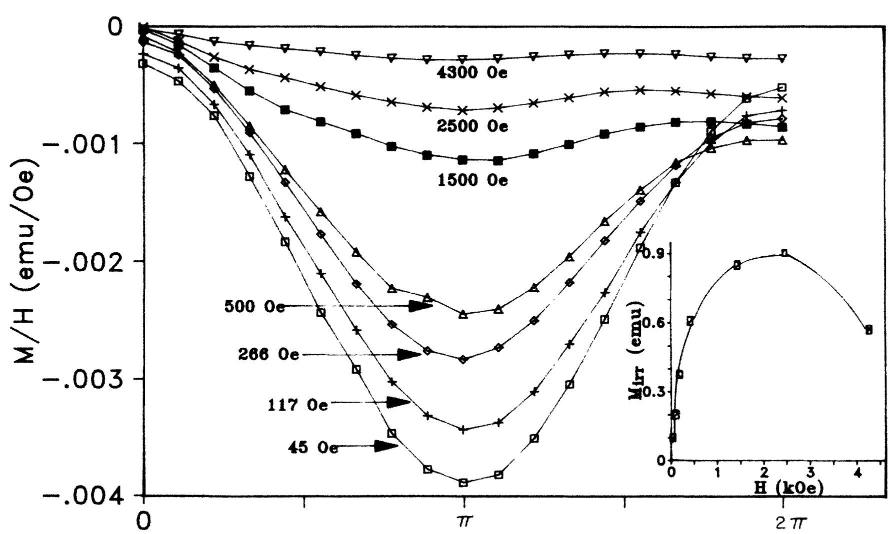

FIG. 2. Angular dependence of the field-cooled magnetization for $YBa_2Cu_3O_7$ at 4.2 K for various fields. The solid lines are a guide for the eye. Inset shows the field dependence of M_{irr} deduced from the angular dependence data (see text).

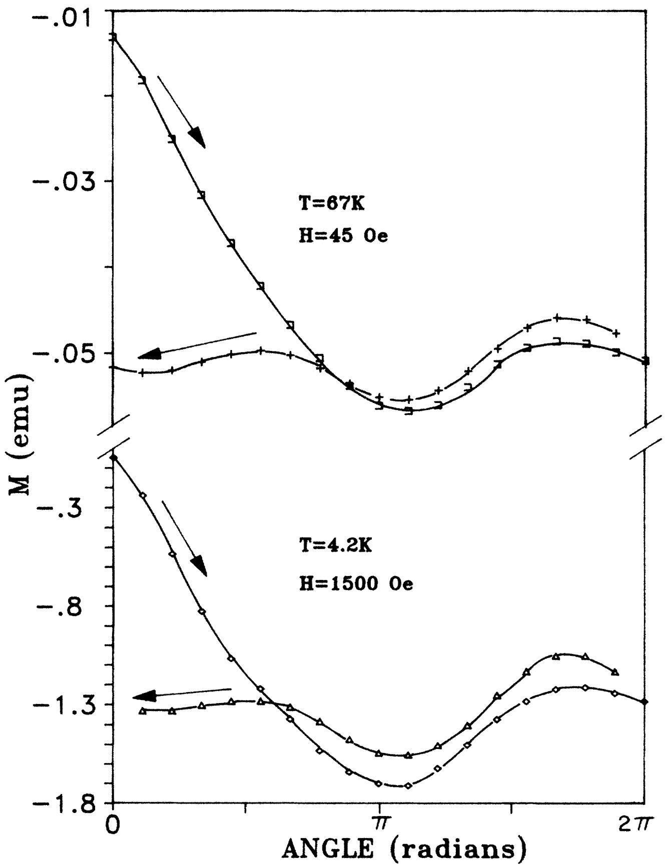

FIG. 3. Angular dependence of the field-cooled magnetization for $YBa_2Cu_3O_7$ at 4 K for 1.5 kOe and at 67 K for 45 Oe. Arrows indicate the direction of the rotation. The measurement starts at $\phi = 0$, the sample is rotated to 2π and back to zero. Note the break of rigidity and the irreversibility. The data taken from 2π to zero coincide with the (reversible) angular dependence of the zero-field-cooled magnetization. The lines are a guide to the eye.

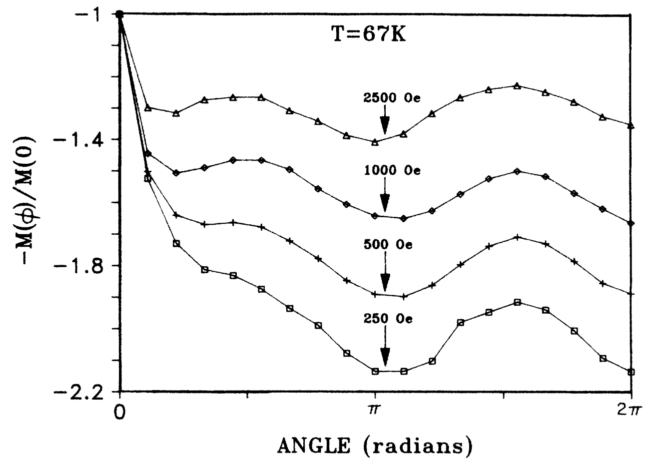

FIG. 4. Angular dependence of field-cooled magnetization normalized to $M(0)$ for $YBa_2Cu_3O_7$ at 67 K for various fields.

ences: (i) $M(\phi)/H$ curves for Nb coincide for the various fields for most of the angular span, as expected for type-II superconductors below H_{c1}. For $YBa_2Cu_3O_7$, on the other hand, the situation is completely different (see Fig. 2), implying that there is no true Meissner regime for this material. (ii) In the high-field regime we observe a plateau above an angle $\theta_c(H)$, implying that the magnetic moment is not capable of following the sample but the lag ϕ-θ is a constant (see inset, Fig. 5). Very similar results, though on a limited angular span, were obtained by Heise[11] in his torque experiments. The plateau in high fields is limited to a small angular span and the overall shape of $M(\phi)$ is symmetric around π and is reversible, implying that M_{irr} is still a rigid body in spite of the fact that in this case $T/T_c \simeq 0.5$. This is to be contrasted with the asymmetry and the irreversibility for $YBa_2Cu_3O_7$ where the rigidity is broken and M_{irr} already vanishes for

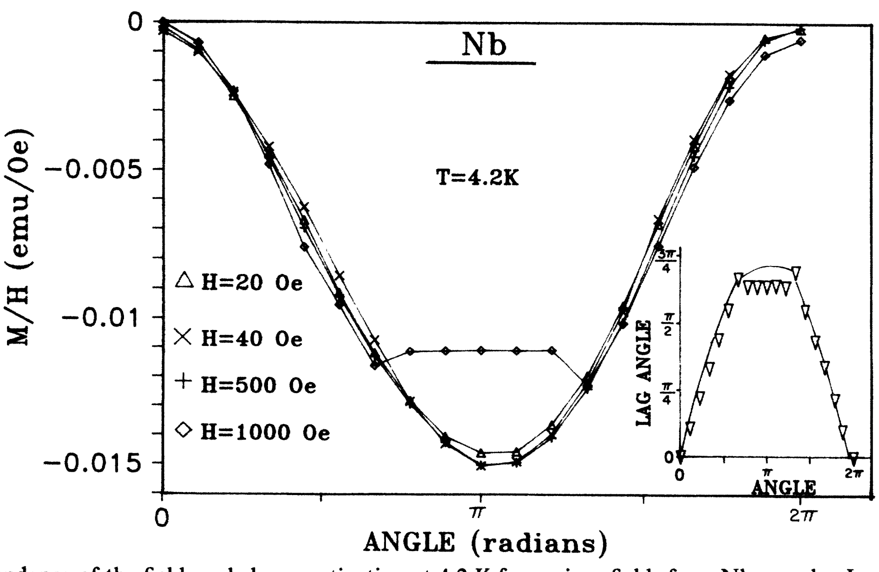

FIG. 5. Angular dependence of the field-cooled magnetization at 4.2 K for various fields for a Nb sample. Inset shows the lag $\phi - \theta$ as a function of the rotation angle ϕ for $H = 1$ kOe.

$T/T_c < 0.05$.

Both Nb and $YBa_2Cu_3O_7$ are type-II superconductors characterized by a mixed phase above H_{c1} in which flux might be trapped and pinned to imperfections or dislocations. Pinned flux might explain the glassy features found in experiments[3–5] and it yields, in particular, a natural explanation for the present experiment: The trapped flux, which contributes a positive magnetization at $\phi = 0$, is rotated with the sample and generates a $\cos\phi$ shape. The fact that a $\cos\phi$ shape is found in the FC experiments for fields well below H_{c1} implies that flux is trapped during the cooling process, while crossing the mixed phase. This implication, however, gives rise to some difficulty. At low temperature and low field the "pure" superconducting phase is energetically more favorable and thus the ZFC magnetization, and not the FC, is the stable state, in complete disagreement with experimental observation of magnetic relaxation in the ZFC branch.[12]

A different approach for the explanation of our results is based on recent superconducting glass models which have been suggested mainly for granular superconductors[13–15] but are quite appealing for the oxide superconductors[3,16] because of their porous nature. In this picture the magnetic field induces frustration[17] by favoring nonuniform phase differences between neighboring grains which are weakly connected via a Josephson coupling. The concrete analogy between this frustrated phase and the magnetic spin-glass system yields a natural explanation for irreversible phenomena in the oxide superconductors. In this sense, the striking similarity between the results presented here and the experimental findings in spin glasses[8,9] lends much support to this approach. We feel, however, that this is not a complete explanation of our results mainly because of the phenomena found in Nb, which does not have the grainy features. It is plausible that both mechanisms, fluxons and "weak links," should be invoked for a complete understanding of the experimental results.

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