New glassy features in high-T_c superconductors

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Magnetic measurements on a high-T_c superconductor $YBa_2Cu_3O_7$ reveal new spin-glass-like features. 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 temperature or field. This behavior resembles that of spin glasses and differs qualitatively from the behavior found for Nb.

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In the effort to understand and characterize the features of the new superconductors, an enormous amount of work has been devoted to their magnetic properties. One of the intriguing features of their materials found in magnetic measurements[1–3] is the striking resemblance to the magnetic properties of spin glasses.[4] We have recently reported[2] on glassy features in $YBa_2Cu_3O_7$. Here we report on new features[5] which resemble those of spin glasses. We have studied the angular dependence of the field-cooled 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 temperature or field. These results resemble the behavior in spin glasses[6–8] and are qualitatively different from those we observe in the ordinary 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 an orthorhombic cell with lattice constants $a = 3.822$ Å, $b = 3.891$ Å, and $c = 11.67$ Å, in fair agreement with published data.[9]

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. 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.

Figure 1 exhibits typical $M(\phi)$ data for various fields. The most obvious feature in this figure is the strong angular dependence of the magnetization. To understand this feature we recall that in VSM measurements the measured magnetization M is the projection of the total sample magnetization $\mathbf{m}$ in the direction of the field $\mathbf{H}$. The magnetization $\mathbf{m}$ is composed of reversible and irreversible components:

$$\mathbf{m} = \chi \mathbf{H} + \mathbf{M}_{irr}. \quad (1)$$

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 as a rigid entity. 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 $\theta \leqslant \phi$ and the measured magnetization is

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

Several features characterize the data of Fig. 1. 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. Moreover, irreversibility in $M(\phi)$ sets in at high angles. This is demonstrated in the lower part of Fig. 2 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 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. 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. 1 exhibits the field dependence of M_{irr}. Note the resemblance of $M_{irr}(H)$ to the thermoremanent magnetization (TRM) in spin glasses.[4]

The effect of the field on the rigidity of M_{irr} is more dramatic at high temperatures. This is obvious from Fig. 2 where we demonstrate that the effect of 45 Oe at 67 K is

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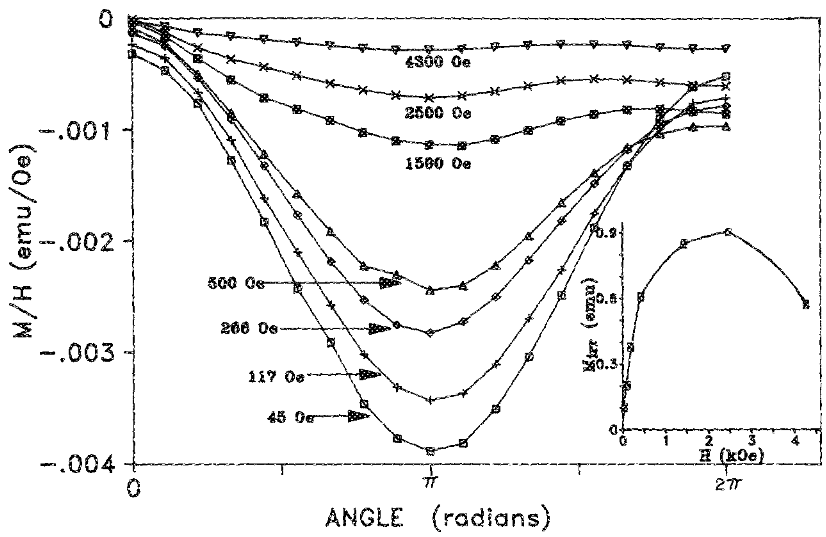

FIG. 1. Angular dependence of the field-cooled magnetization for $YBa_2Cu_3O_7$ at 4.2 K for various fields. The solid lines are a guide for the eye. Inset shows the field dependence of M_{irr} deduced from the angular dependence data (see text).

qualitatively similar to that of 1.5 kOe at 4.2 K. For higher fields the initial cos ϕ dependence is abruptly interrupted at a relatively small angle ϕ^* which decreases with the increase of H.

To complete the experimental description, we compare the results with FC data for Nb (Fig. 3). The figure exhibits $M(\phi)$ dependencies for Nb which resemble those of $YBa_2Cu_3O_7$. There are, however, two important differences: (i) $M(\phi)/H$ curves for Nb coincide for the various fields for most of the angular span, as expected from type-II superconductors below H_{c1}. For $YBa_2Cu_3O_7$, on the other hand, the situation is completely different (see Fig. 1), 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 $\phi - \theta$ is a constant (see inset, Fig. 3). Very similar results, though on a limited angular span, were obtained by Heise[10] 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 $T/T_c < 0.05$.

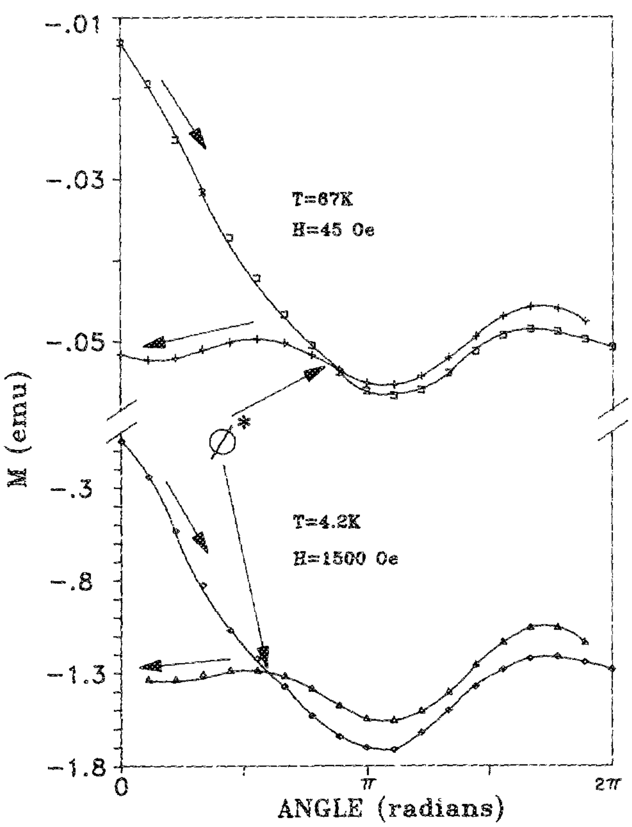

FIG. 2. Angular dependence of the field-cooled magnetization for $YBa_2Cu_3O_7$ at 4 K for 1.5 kOe and at 67 K for 45 Oe. Arrows indicate the direction of the rotation. The measurement starts at $\phi = 0$, the sample is rotated to 2π and back to zero. Note the break of rigidity at the critical angle ϕ^* denoted by the arrows 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.

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[1-3] 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 ϕ shape. The fact that a cos ϕ 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.

A different approach for the explanation of our results is based on recent superconducting glass models which have been suggested mainly for granular superconductors[11-13] but are quite appealing for the oxide superconductors because of their porous nature. In this picture the magnetic field induces frustration[14] 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[6,7] lends much support to this approach.

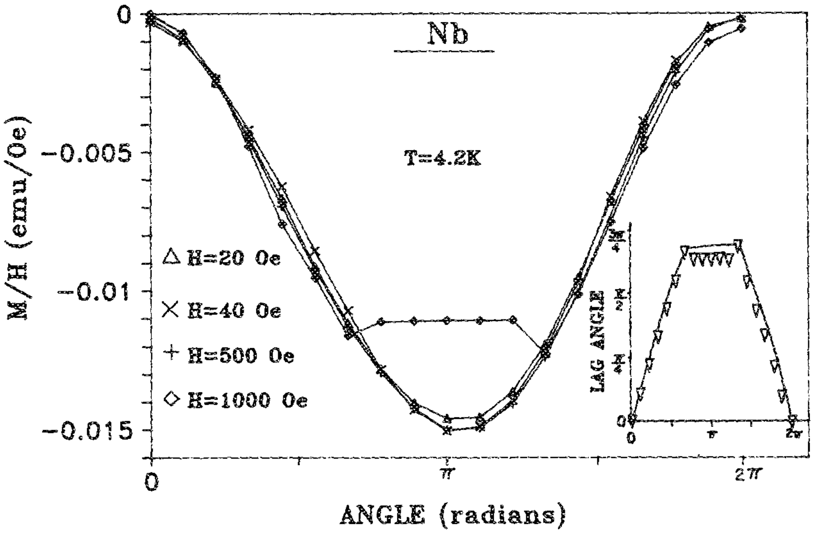

FIG. 3. Angular dependence of the field-cooled magnetization at 4.2 K for various fields for a Nb sample. Inset shows the lag $(\phi - \theta)$ as a function of the rotation angle ϕ for $H = 1$ kOe.

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