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Magnetic properties of a high-T_c superconductor $YBa_2Cu_3O_7$: Reentrylike features, paramagnetism, and glassy behavior

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Magnetic measurements on a high-T_c superconductor $YBa_2Cu_3O_7$ are reported. Diamagnetism sets in, at low fields, below a temperature $T \simeq 90$ K. At low T the field-cooled magnetization becomes positive again. We show that the low-T phase is still superconducting and the apparent reentry behavior is due to strong paramagnetic contributions. Glassy features appear below a temperature whose field dependence differs substantially from that observed in spin glasses.

Last year Bednorz and Müller[1] reported on the possibility of a breakthrough in producing superconductors with high transition temperature T_c. Since then the highest T_c has advanced substantially, and very recently Wu *et al.*[2] reported on a Y-Ba-Cu-O system with $T_c \gtrsim 90$ K. In this paper we present magnetic measurements on a superconductor of the latter family, a $YBa_2Cu_3O_7$ sample. Both the zero-field-cooled (ZFC) and the field-cooled (FC) magnetizations of this sample exhibit a diamagnetic behavior which sets in, at low fields, below 90 K. The FC diamagnetic susceptibility at low temperatures and fields is of the order of $\sim 20\%$ of the complete Meissner effect $(1/4\pi)$. The ZFC magnetization decreases monotonically with decreasing T. The FC branch, on the other hand, exhibits a minimum and starts increasing upon further cooling, reaching a positive value at low temperature. Glassy features such as a large difference between the ZFC and the FC susceptibilities and time-dependent remanent magnetization appear below a "glass" temperature T_g. The field dependence of this temperature is qualitatively different than the analogous de Almeida–Thouless line[3] in spin glasses.[4-6] The glassy features as well as the apparent reentry behavior are discussed below.

The sample was prepared from a mixture of $BaCO_3$, Y_2O_3, and CuO powders (99.9% pure at least) in stoichiometric proportion according to the formula $YBa_2Cu_3O_7$. Finely ground powders were pressed into a pellet approximately 1.5 cm diam 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, and 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 which are in fair agreement with data given in Ref. 7.

The magnetic measurements have been carried out on a superconducting quantum-interference device (SQUID) magnetometer. The magnetization was measured following two procedures. (i) The sample is cooled in zero field. At low temperature, a field H (20 Oe $\leq H \leq$ 10 kOe) is applied and the ZFC branch of the susceptibility M/H is measured while temperature is increased. (ii) With the field still on, the sample is cooled to low temperature and the field-cooled FC branch of the susceptibility M/H is measured while temperature is increased. We emphasize that in this paper we analyze data that have been taken during heating in both the FC and the ZFC procedures. A susceptibility during cooling shows hysteretic effects and diamagnetization appears a few degrees above that measured during heating. For example, at the lowest field of the measurements, diamagnetism of the FC branch disappears at 85 K during heating and reappears at 90 K during the cooling cycle.

Figure 1 exhibits a typical ZFC-FC run with $H = 3$ kOe. The difference between the two branches reflects the glassy nature of the superconducting state which has also been observed in the La-Ba-Cu-O system.[8] We first focus attention on the low-temperature behavior of the FC branch. As seen in Fig. 1, below $T^* = 13$ K the measured

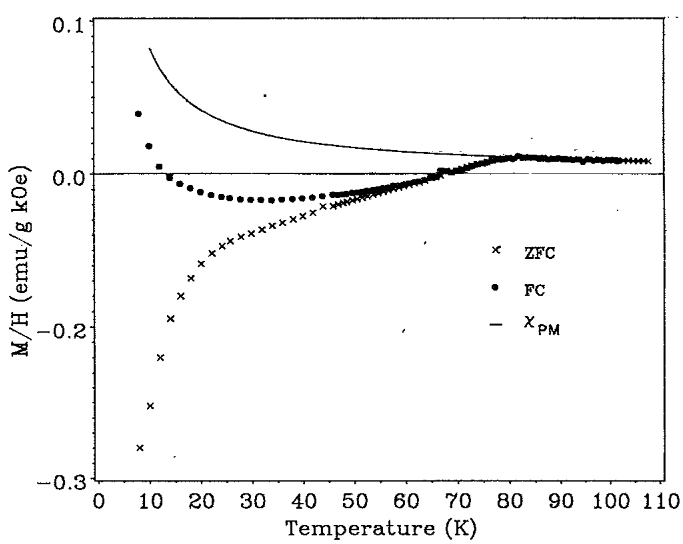

FIG. 1. Zero-field-cooled and field-cooled branches of the susceptibility M/H of $YBa_2Cu_3O_7$ measured in $H = 3$ kOe. The solid line is the paramagnetic contribution, Eq. (1), fitted above T_c and extrapolated to low temperatures.

FC susceptibility is *not* diamagnetic. The temperature T^* increases with the increase of the field H, as demonstrated in Fig. 2.

Does T^* signal a destruction of the superconducting phase? This is probably *not* the case. We demonstrate in the following way that the low-temperature phase is superconducting. After field cooling the sample to $T < T^*$, we switch off the field and observe a positive remanent magnetization M_r with a magnitude larger than that of a magnetization M in the presence of the field. The positive M_r is apparently the result of a flux trapping and the fact that $M_r > M$ implies that the induced magnetization still has a substantial diamagnetic component. We therefore conclude that the superconducting phase survives at low temperatures.[9]

To explain the positive susceptibility obtained at low temperature we assert that the susceptibility M/H is composed of diamagnetic contribution χ_d and a paramagnetic (PM) "background" χ_{PM}. In fact, a small temperature-dependent PM contribution is clearly observed above T_c. The temperature dependence of the susceptibility at $T > T_c$ is found to be well characterized by

$$\chi_{PM} = \chi_0 + \frac{C}{T} . \qquad (1)$$

We find that extrapolating χ_{PM} to lower temperatures accounts well for the observed reentrant features. This holds for all the fields of this experiment. The values of χ_0 and C are found by least-squares analysis of the susceptibility above T_c. The solid line in Fig. 1 is the result of such a fit for $H = 3$ kOe, where we find $\chi_0 = 5.4\times10^{-7}$ emu/g and $C = 8\times10^{-4}$ K emu/g for the high-temperature data and extrapolate the result to low temperatures. To obtain the "pure" diamagnetic contribution we subtract χ_{PM} from the raw data. The diamagnetic susceptibilities obtained by such a procedure are exhibited in Fig. 3 for the same fields as in Fig. 2. It is apparent that χ_d is indeed diamagnetic at all temperatures. It is tempting to interpret χ_0 as a Pauli susceptibility of the conduction electrons and C as a Curie constant of localized magnetic moments. However, we note that both parameters show a significant reduction as a function of magnetic field. This is true particularly for χ_0 which changes by more than an order of magnitude in the field range of the experiment. We do not yet understand this field dependence. Nevertheless, the fact that the field-cooled $M/H - \chi_{PM}$ saturates at low T with a negative value indicates that *most of the PM contribution at low temperature is of the same origin as that which is observed at high temperature.* The above-mentioned field dependence of χ_{PM} is related to a substantial nonlinearity in $M(H)$ observed even at the highest temperature of this experiment (110 K).

We now discuss the glassy features of the magnetization. The difference between the ZFC and the FC

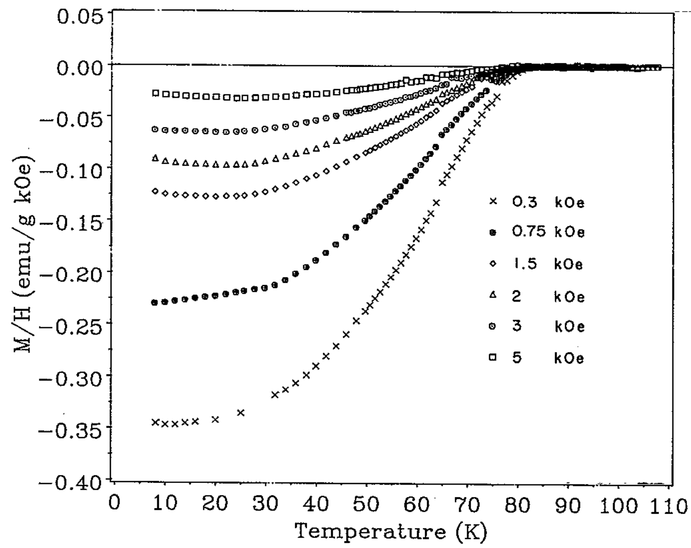

FIG. 3. Diamagnetic contribution $(M/H - \chi_{PM})$ for the field-cooled susceptibilities of Fig. 2.

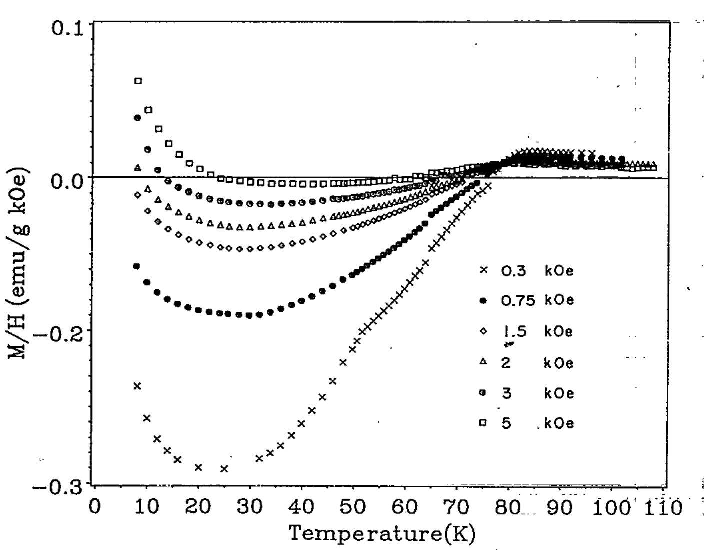

FIG. 2. Field-cooled branches of the susceptibility M/H of $YBa_2Cu_3O_7$ measured in the indicated fields.

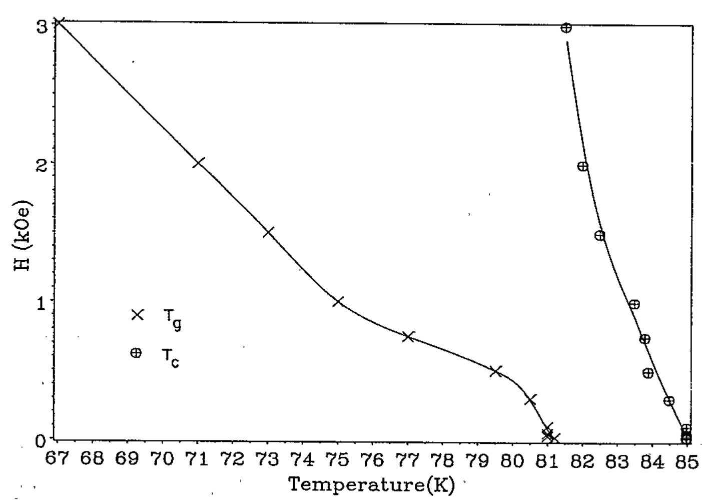

FIG. 4. The field dependence of the glass temperature and of the superconducting temperature for $YBa_2Cu_3O_7$. The solid lines are a guide for the eye.

branches resembles the magnetic behavior of spin glasses, as noted by Müller *et al.*[8] The ZFC branch is clearly metastable and shows a pronounced time effect, whereas the FC branch is stable. The difference between the FC and the ZFC branches is due to flux trapping and is expected in all "dirty" superconductors. As seen in Fig. 1, this difference vanishes at a temperature T_g which is below the temperature T_c at which diamagnetic effects vanish. The field dependence of T_g is shown in Fig. 4. For comparison we also show the field dependence of T_c. Note that T_c is defined by the vanishing of $M/H-\chi_{PM}$ (see Fig. 3). The results of Fig. 4 show a rather weak dependence of T_g on H at low fields. Furthermore, $T_g(H)$ follows roughly the field dependence of $T_c(H)$ in the low-field range. This is clearly different from the spin-glass case, where the irreversibility temperature shows[4-6] a strong field dependence of the form $T_g(0)-T_g(H) \propto H^{2/3}$. This latter form agrees with the spin-glass mean-field theory (the de Almeida–Thouless line).[3]

The existence of a glassy phase in granular superconductors has been predicted theoretically.[10-12] A concrete analogy between such a phase and the spin-glass phase is provided by the "weak-link" model of superconducting grains in a nonsuperconducting host interacting via a Josephson coupling.[10] An applied magnetic field induces frustration by favoring nonuniform phase differences between neighboring grains. Although this model may give rise to a "spin-glass" phase it should be stressed that the role of the applied magnetic field is very different from that in spin glasses. In real spin glasses the magnetic field suppresses the spin-glass phase by aligning the spins. This results in a marked decrease of T_g by H, i.e., the de Almeida–Thouless line. In contrast, in the case of granular superconductors, increasing the field *increases* the system's frustration, and therefore should enhance its glassy behavior. This holds particularly at small fields such that the superconducting properties of the individual grains are not affected by H. Indeed, phase diagrams based on mean-field approximation[11,12] predict only weak dependence of T_g on H. We thus conclude that the observed weak-field dependence of T_g at low fields is consistent with the "weak link" model of granular superconductors and differs from the characteristic de Almeida–Thouless line observed in spin glasses.

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[1] J. G. Bednorz and K. A. Müller, Z. Phys. B **64**, 189 (1986).
[2] M. K. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, A. J. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. **58**, 908 (1987).
[3] J. R. L. de Almeida and D. J. Thouless, J. Phys. A **11**, 983 (1978).
[4] P. Monod and H. Bouchiat, J. Phys. (Paris) Lett. **43**, L-45 (1982).
[5] R. V. Chamberlin, M. Hardiman, L. A. Turkevich, and R. Orbach, Phys. Rev. B **25**, 6720 (1982).
[6] Y. Yeshurun and H. Sompolinsky, Phys. Rev. B **26**, 1487 (1982).
[7] J. J. Capponi, in proceedings of the European Physical Society meeting, Pisa, April 1987 [Phys. Scr. (to be published)].
[8] K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
[9] We note that the existence of a real reentrant phase has been reported in a Lu-Ba-Cu-O superconducting system by J. A. Mydosh, in proceedings of the European Physical Society meeting, Pisa, April 1987 [Phys. Scr. (to be published)].
[10] C. Ebner and A. Stround, Phys. Rev. B **31**, 165 (1985).
[11] S. John and T. Lubensky, Phys. Rev. Lett. **55**, 1014 (1985); Phys. Rev. B **34**, 815 (1986).
[12] M. V. Feigelman, L. B. Ioffee, A. I. Larkin, and V. M. Vinokur (unpublished).