Susceptibility, crystal-structure, and specific-heat studies of the high-T_c superconductor and quenched $YBa_2(Cu_{0.91}Fe_{0.09})_3O_x$

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Magnetic susceptibility, powder x-ray diffraction, and specific-heat measurements were used to study the high-temperature superconductor $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{7.1}$ and the nonsuperconductor quenched $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{6.1}$ samples. The crystal structure for both samples is tetragonal with almost the same lattice parameters. No magnetic spin-glass state of Fe is observed in the two samples at low temperatures and the quenched sample exhibits antiferromagnetic behavior, whereas the magnetization is nonlinear as a function of the applied field. For the superconducting sample, magnetic irreversibilities appear below a temperature T_g whose field dependence differs substantially from that observed in spin glasses. T_g scales with the applied field as $H^{1/4}$. The linear term obtained from the specific-heat studies is extremely high, $\gamma = 73$ mJ/mol K^2.

I. INTRODUCTION

Recent susceptibility and magnetization measurements on the high-T_c materials, have shown certain similarities to well-known spin-glass materials.[1,2] For example, the shielding zero-field-cooled (ZFC) branch of the magnetization is irreversible, metastable, and lower than the reversible field-cooled (FC) Meissner branch, i.e., it is more strongly diamagnetic. The temperature T_g, which is defined as the temperature which separates the reversible and irreversible branches, is magnetic-field dependent and scales with the applied field as $H^{2/3}$.[3] It was assumed that the granular nature of these materials and the short coherence length are responsible for the existence of internal Josephson junctions at the twin boundaries, and are the origin of the so-called, superconducting glassy state.[4] Recently, the magnetic irreversibilities (in single crystals) were explained in the framework of thermally activated flux creep model, providing an alternative to the previous superconducting glass model.[5]

It has been reported by several groups[6,7] that partial substitution of Cu in the orthorhombic superconductor $YBa_2Cu_3O_7$ by Fe, progressively decreases the T_c of the system, and for Fe concentration higher than 2% the crystal structure transforms to tetragonal. Mössbauer[8] and neutron-diffraction[9] measurements definitely show that the Fe ions substitute preferentially at the Cu(1) sites located on the basal plane of the structure and only about 10%–20% of Fe substitutes for Cu in the Cu(2) sites on the CuO_2 planes. Moreover, low-temperature Mössbauer studies of samples doped with more than 5% at.% Fe show[8,10–12] magnetic broadening (around 10–20 K) at temperatures which depend on Fe concentration, and indication of magnetic ordering. At 4.1 K the spectra become more complicated showing a distribution of magnetic hyperfine fields which are reminiscent of spectra of spin-glass systems. It was assumed that spin-glass short-range magnetic order of Fe ions coexists with superconductivity in the materials at 4.1 K.[12,13] These two independent spin-glass features in the Fe-doped samples motivated the present work.

In order to clarify whether the two spin-glass phenomena are connected or not, extensive magnetic-susceptibility measurements at low temperatures and low magnetic fields have been carried out on both the fully oxygenated superconducting $YBa_2Cu_{2.73}Fe_{0.27}O_{7.13}$ and on the quenched nonsuperconducting $YBa_2Cu_{2.73}Fe_{0.27}O_{6.1}$; both samples were taken from the same batch. Since the Mössbauer spectra of the quenched sample at 4.1 K also shows the same complicated shape mentioned above,[8] it was assumed that the spin-glass features of Fe ions are not affected by the quenching process. Our bulk susceptibility measurements on both samples show no evidence for spin-glass or long-range magnetic ordering[14] of the Fe ions at low temperatures. The Mössbauer spectra at 4.1 K may therefore be attributed to slow paramagnetic relaxation effects. In addition, the glassy behavior of the superconducting state is studied. It is found that T_g scales with the applied magnetic field as $H^{1/4}$ in contrast with the $\frac{2}{3}$ power law found in other systems.[3] The specific-heat measurements show no evidence of any magnetic transitions and the γ value obtained from the linear region of $C_p(T)/T$ vs T^2 is 73 mJ/mol K^2, remarkably larger than in the pure $YBa_2Cu_3O_7$ sample.

II. EXPERIMENTAL DETAILS

The sample with a nominal composition $YBa_2Cu_{2.7}$-$Fe_{0.3}O_x$ was prepared by conventional methods.[2] X-ray diffraction measurements show that the sample is single phase (content of impurity phases are less than 2%). Part of this sample was heated to 800 °C for 2 h and then quenched in a liquid-air bath. The exact iron and copper concentrations were determined by chemical analysis using the atomic absorption method. The Fe concentration is slightly less than the nominal composition. According to neutron diffraction studies[9] the oxygen concentration for the fully oxygenated $YBa_2Cu_3O_x$ doped with 10 at.% Fe is 7.13. The chemical formula deduced for the superconducting sample is, therefore, $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{7.1}$. For the nonsuperconducting quenched sample the oxygen content was obtained by weight loss, and we assign $x = 6.1$ to this sample. Surprisingly, no measurable differences were observed between the lattice parameters of the two tetragonal samples which are $a = 3.874(2)$ Å and $c = 11.70(3)$ Å. These values are in fair agreement with data given in Ref. 15, although they report a small increase in c for the quenched sample. The dc susceptibility measurements on solid ceramic pieces were carried out in a commercial SHE superconducting quantum interference device (SQUID) magnetometer and in a 155 PAR vibrating sample magnetometer in various fields 10 Oe $< H <$ 40 kOe as a function of temperature in the range 4.2–300 K. The magnetization was measured by two different procedures: (a) The sample was zero field cooled (ZFC) to 5 K, a field H was applied, and the magnetization of the shielding branch was measured as a function of temperature. (b) The sample was FC from above T_c in a field H and the Meissner branch was measured. The heat capacity was measured over the temperature range 1.5–70 K in an automated adiabatic calorimeter employing the Nernst step heating method.

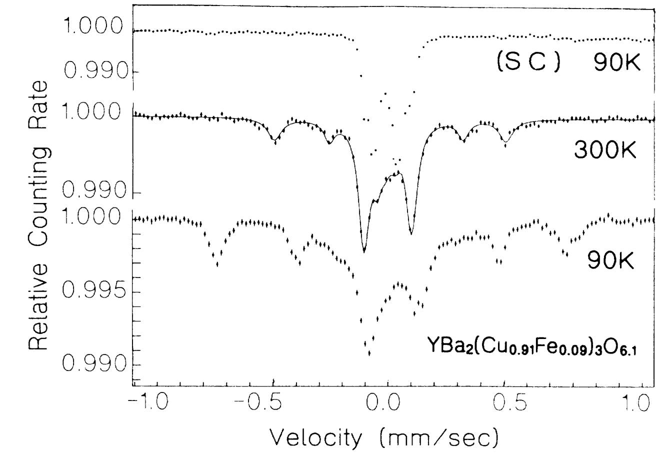

FIG. 1. Mössbauer spectra at 90 and 300 K for superconducting and quenched samples. The solid line is just for guidance.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{6.1}$ (quenched)

Recent neutron-diffraction measurements[16] on $YBa_2Cu_3O_{6.1}$ show the existence of long-range three-dimensional antiferromagnetic order of the Cu spins at the Cu(2) sites. The magnetic moments are within the CuO_2 plane and the planes are coupled antiferromagnetically along the c axis. The Néel temperature is about 410 K. Mössbauer studies[17] (performed on samples taken from the same batch which was measured here) show that for iron-doped samples, the fraction of iron ions entering the Cu(2) sites in the nonsuperconducting quenched sample order magnetically with the same Néel temperature as in the undoped material. This fact proves that the ordering temperature is not affected by the presence of iron impurities. Figure 1 (taken from Ref. 17) displays typical Mössbauer spectra of the superconducting and quenched 9 at.% Fe doped in $YBa_2Cu_3O_x$. The sextet in the quenched sample is due to Fe in the Cu(2) sites and as the temperature is raised the magnetic splitting decreases until at $T_N = 415$ K where the sextet disappears. The magnetic moments of Fe lie in the CuO_2 planes.[17] No magnetic splitting is observed in the superconducting sample at 90 K. In Fig. 2 the FC and ZFC-magnetic curves of the quenched sample are presented, and it seems clear that there is no sign of spin-glass magnetic behavior of Fe ions at low temperatures, although the Mössbauer spectra show broadening and magnetic hyperfine structure at low temperatures.[8] The coincidence of the ZFC and FC curves also occurs at low magnetic fields. Surprisingly, there is a nonzero moment at zero field and the magnetization curve is nonlinear as a function of the applied field, and a sharp upturn is observed. This upturn is displayed together with the hysteresis loop, both measured at 80 K, in Fig. 3. The coercive field is 600 Oe. We may assume that the Fe moments in the CuO_2 planes lead to a small deviation from antiparallel alignment of the antiferromagnetic nature of the Cu(2) sites and a nonzero moment is present at zero field. The small canting angle in this weak ferromagnetic structure undergoes a transition above 100 Oe and causes the hysteresis loop obtained.

In Fig. 4 the different susceptibility curves obtained below and above the upturn transition are shown. The difference between the susceptibility curves obtained at low (50–100 Oe) and high (500–15000 Oe) magnetic fields is obvious. In this figure the susceptibility curve for

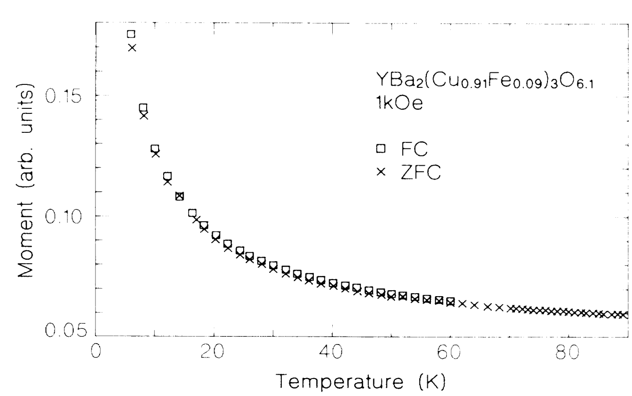

FIG. 2. ZFC and FC magnetic-susceptibility curves of the quenched sample $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{6.1}$.

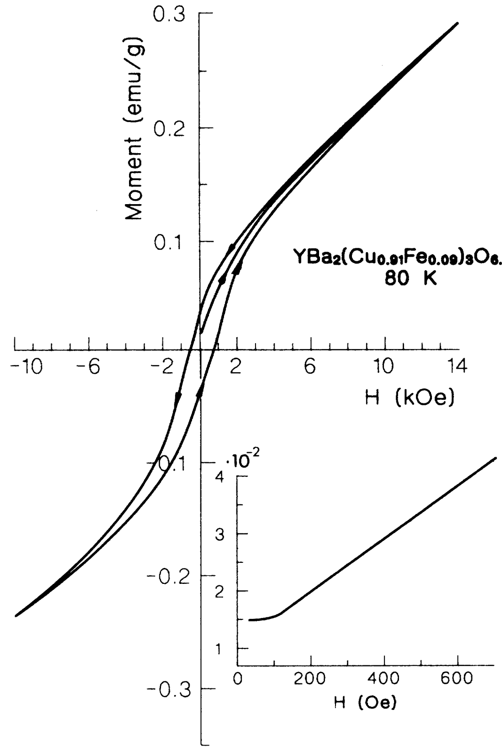

FIG. 3. Magnetic hysteresis loop for the quenched sample at 80 K. Inset: the magnetization at low magnetic field is shown.

the superconducting sample at 15 kOe is also shown, and the higher values of this sample relative to the quenched sample will be discussed later. The temperature dependence of the susceptibility for the quenched sample in the two regions is well characterized by the Curie-Weiss law

$$\chi = \chi_0 + C/(T - \Theta) , \quad (1)$$

where χ_0 is the temperature-independent susceptibility, C and Θ are the Curie-Weiss constant and temperature, respectively. The molar values obtained by least-squares fits for 50 Oe and 500 Oe are $\chi_0 = -6.2\times10^{-3}$ and -1.4×10^{-3} emu/mol Oe, $C = 0.16$ and 0.35 emu/mol Oe K, and $\Theta = -1.0$ and 1.3 K, respectively (the solid lines in Fig. 4). In this compound the Cu ions in the Cu(1) sites are predominantly monovalent and nonmagnetic. The contribution to the magnetic moment arises from two sources: (1) Fe^{3+} and Cu^{2+} ions which are antiferromagnetic coupled within the CuO_2 planes and (2) Fe^{3+} ions (about 80%) which occupy the Cu(1) sites. There are too many unknown parameters to allow us to offer any physically meaningful interpretation to the difference in these molar Curie-Weiss constants.

All the magnetic features mentioned above are attributed to the antiferromagnetic nature of Cu and Fe in the Cu(2) sites of the quenched sample, and not to other magnetic phases such as unincorporated Fe. Otherwise these features would be detectable also in the superconducting sample above T_c, but there is not any sign for magnetic phases in this sample (see below).

B. $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{7.1}$ (superconducting)

T_c for this compound (obtained at 5 Oe) is 69(1) K and the reduction of T_c when Fe is doped in $YBa_2Cu_3O_7$ ($T_c = 92$ K) appears to be not a magnetic effect [since Ni and/or Zn substitution has a much larger effect upon the suppression of T_c (Ref. 18)] but due to introduction of structural disorder in the crystal structure. The hysteresis loop (only half of it) at 4.1 K for applied fields up to 40 kOe is shown in Fig. 5. The sample was cooled in zero field to obtain the virgin curve. From the inset in Fig. 5, which shows the virgin curve on an extended scale, it can be seen that M vs H begins to curve (flux begins to penetrate the sample) at $H_{c1} = 200$ Oe. The magnetization curve is reversible as long as one never exceeds H_{c1} and the value of the volume susceptibility is 26% of $-1/4\pi$ at 4.1 K. From the width of the hysteresis loop, it is possible to obtain a crude estimate of the critical current density J_c using $J_c = 10\Delta m/d$ where Δm denotes the difference between the negative and positive parts of the magnetization and d is the thickness of the sample. J_c

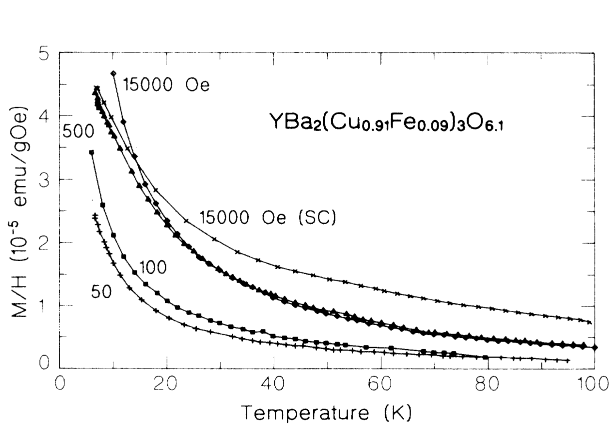

FIG. 4. Susceptibility curves vs temperature for the quenched sample at 50, 100, 500, 15 000 Oe. Note the difference between the low- and high-field curves. The susceptibility of the superconducting at 15 000 Oe is also shown.

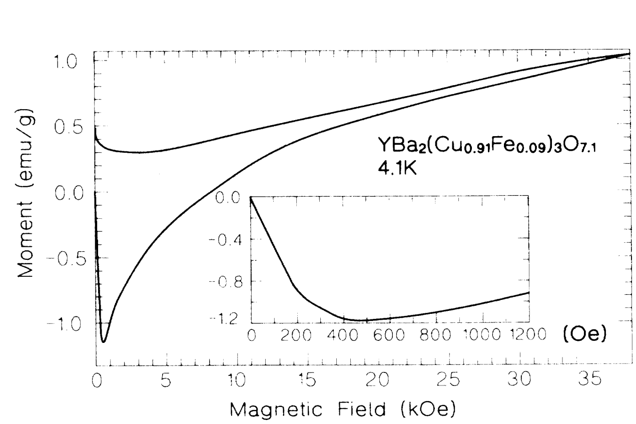

FIG. 5. The magnetization M vs the applied field H for the superconducting $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{7.1}$ at 4.1 K. The inset shows magnetization values at low H in an extended scale.

obtained is of the order 300 A/cm^2 at 4.1 K and 10 kOe with an error of 20% due to the uncertainty of d. This value is three orders of magnitude smaller than values obtained for undoped $YBa_2Cu_3O_7$ samples.

The temperature dependence of the susceptibility M/H for the superconducting material at 15 kOe is shown in Fig. 4. In the normal state at $T > 20$ K the susceptibility is higher than that of the quenched sample. This reduction for the latter compound is attributed either to the general decrease of the Curie-Weiss component with the decreasing oxygen content[19] leading to the decrease of M/H in the quenched sample, or to antiferromagnetism discussed above, which would mean that even at that magnetic field some part of spins remain coupled.

We now focus attention on the glassy behavior of the superconducting features. Figure 6 exhibits several typical ZFC-FC curves with different applied magnetic fields. The FC magnetization versus temperature at 50 Oe reaches 54% of the ZFC magnetization. This indicates that the Meissner effect (the flux expulsion) is incomplete and flux is pinned during the cooling process. When the magnetic field is increased to above H_{c1} an upturn in the shielding curve is observed at low temperatures as shown for the 500 Oe curve. For high-magnetic fields, both FC and ZFC are positive although the sample is still in the superconducting state. Above T_c all the susceptibility curves, measured either *at low or high* magnetic fields coincide into one curve which is well characterized by the Curie-Weiss law [Eq. (1)]. This is in contrast to different susceptibility curves obtained for the quenched sample (Fig. 4). At 300 Oe we derive $\chi_0 = 1.4 \times 10^{-5}$ emu/mol Oe and $C = 0.40$ emu/mol Oe K and $\Theta = 2.8$ K. These values agree perfectly with Ref. 20, and if the Curie constant of pure $YBa_2Cu_3O_7$ ($C = 0.14$ emu/mol Oe K) (Ref. 21) is subtracted from the above value, one obtains an effective moment $P_{eff} = 3.2\mu_B$/Fe ion.

To explain the upturn at magnetic fields higher than H_{c1} and the positive susceptibility obtained at magnetic fields higher than 2000 Oe, we assert that the susceptibility M/H is composed of a diamagnetic contribution due to part of the sample which is superconducting and a paramagnetic contribution from the rest of the sample mainly due to Fe^{3+} ions. The effective magnetic field acting above H_{c1} on the iron ions is just the external applied field, and the magnetic data for the paramagnetic component are those obtained from the temperature dependence of the susceptibility at $T > T_c$ given above.

The temperature dependence of the diamagnetic susceptibility of the superconducting part in the ZFC branch is proportional to an empirical volume equation

$$V_{sc} = V_0[1 - T/T_c(H)]^{\alpha} , \quad (2)$$

and the total susceptibility is given by

$$M/H = -V_{sc}/4\pi d + (1 - V_{sc})[\chi_0 + C/(T - \Theta)] , \quad (3)$$

where V_0 is the volume fraction of the sample which is superconducting at 0 K, d is the measured density, and V_0, T_c, and α are free parameters. The ZFC curve measured at 1000 Oe is shown in Fig. 7 together with the calculated susceptibility (solid line) and the diamagnetic contribution (dashed line) calculated by least-square fits of Eq. (3) and Eq. (2), respectively. The free parameters obtained are $V_0 = 0.017(1)$, $T_c = 64(1)$, and $\alpha = 1.9(1)$. T_c obtained here as a free parameter of Eq. (3) is lower than 69(1) K measured at 5 Oe; it is therefore impossible to calculate from these values the field dependence of the higher critical field H_{c2}. The positive susceptibility curves obtained at higher magnetic fields (Fig. 6) are due to the reduction in the diamagnetic contribution. Equation (3) holds only for the ZFC branch, since, in the FC process

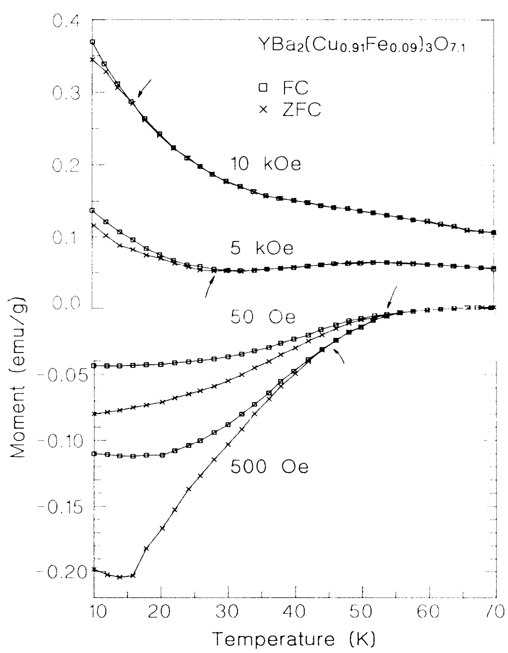

FIG. 6. ZFC and FC magnetic susceptibility curves at different magnetic fields for the superconducting $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{7.1}$. Note the upturn in the ZFC curve at 500 Oe and the positive values obtained at high H. The arrows indicate the inflection temperatures $T_g(H)$.

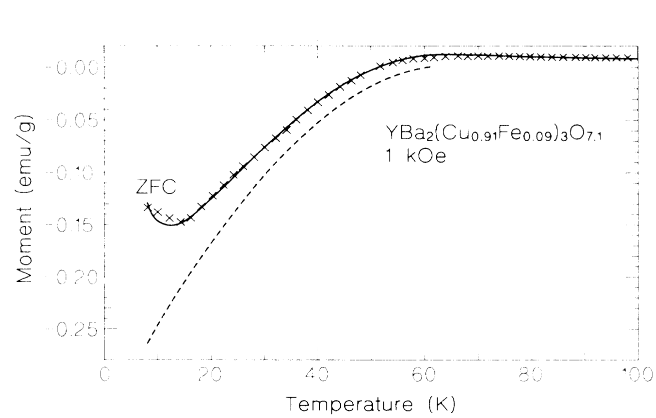

FIG. 7. The temperature dependence of the ZFC susceptibility of the superconducting sample at 1000 Oe. The solid line is the fitted susceptibility (see text) and the dashed line is the pure diamagnetic contribution to the susceptibility.

additional unknown factors (such as the amount of trapped flux) have to be considered. It should be mentioned that the present treatment differs slightly from that given in Ref. 2.

The difference between the FC and the ZFC branches vanishes at T_c (indicated by arrows in Fig. 6). The field dependence of T_g is shown in Fig. 8, yielding a rather weak dependence of T_g on H at low fields. From the linearity of $\log_{10}[T_g(0)-T_g(H)]$ vs $\log_{10}H$ plot (inset of Fig. 8) we obtained for the slope 0.25 ± 0.02. The fact that T_g scales with the applied field as $H^{1/4}$ stands in contrast to other high-T_c superconductors which exhibit superconducting glassy phenomena[3] and clearly differs also from the regular magnetic spin-glass state where the temperature of the irreversibility shows a strong-field dependence[22] of the form $T_g(0)-T_g(H)\propto H^{2/3}$.

The specific heat of the superconducting sample over the temperature range 4–70 K is shown in Fig. 9. Figure 10 shows the specific heat of the 10 at.% Fe-doped sample together with the pure $YBa_2Cu_3O_7$ phase, in the usual presentation of C_p/T vs T^2. At very low temperatures, the strong upturn in C/T, common for these materials, is observed in both samples.

From the nearly linear variation of the data in Fig. 10 we extrapolate a finite C/T value at $T=0$ which we denote with γ^* in order to distinguish between this term and the normal electronic specific-heat coefficient γ. For our undoped samples γ^* varies between 2.5 and 17 mJ/mol K^2, whereas T_c remains almost constant. In this connection we note that γ^* values of 5 and 9 mJ/mol K^2 for tetragonal and orthorhombic $YBa_2Cu_3O_{7-\delta}$ single crystals have been observed respectively.[23] Substitution of Cu by 10 at.% Fe significantly increases γ^* up to 73 mJ/mol K^2. This appears to be a systematic trend in $YBa_2Cu_3O_7$ when Cu is replaced by either Zn or Ni (Refs. 18, 23, and 24), and cannot simply be attributed to normal-state electronic heat capacity of an impurity phase. Otherwise, the second phase containing Fe would be detectable in the Mössbauer or x-ray measurement and should exhibit an unreasonably high value, comparable to that of a heavy-fermion system. This systematic trend is also observed in $La_{1.85}Sr_{0.15}CuO_4$ on substituting Cu by

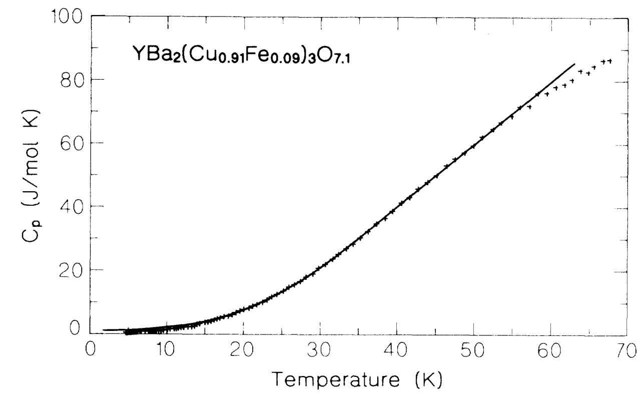

FIG. 9. C_p vs T curve for the superconducting Fe-doped sample.

Zn: γ^* rises by a factor of 4 upon 2.5% Zn doping.[18] Under the assumption of an almost linear increase of γ^* upon replacement of Cu by Zn or another transition metal, we derive for the La-Sr-Cu-O and the Y-Ba-Cu-O system $d\gamma^*/dx=3$ and 2.1 mJ/Cu-mol K^2 per mol% Zn, respectively. Here mJ/Cu-mol K^2 was used in order to have comparable values for both systems. With the latter value (multiplied by a factor of 3 to obtain mJ/mol K^2) we estimate γ^* for the 9 at.% Fe-doped sample to be 73.7 mJ/mol K^2 which is in good agreement with the experimental value. These findings together with arguments concerning the Zn substitution presented recently[18] indicate that the finite γ^* term is an intrinsic property of these materials and may arise from unpaired nonsuperconducting carriers and/or from a vanishing gap at certain regions of the Fermi surface.

In conclusion, we provide the first static magnetic measurements of the antiferromagnetic nature found in the nonsuperconducting system using Fe dopant as a microscopic probe. The mechanism for the magnetic behavior of this system, particularly in low and high magnetic fields is still not clear. We reject the assumption of magnetic spin-glass behavior of Fe ions at low temperatures in both superconducting and quenched samples, which was deduced from Mössbauer studies.[10–12] The broadening in the Mössbauer spectra at low temperatures is due to slow

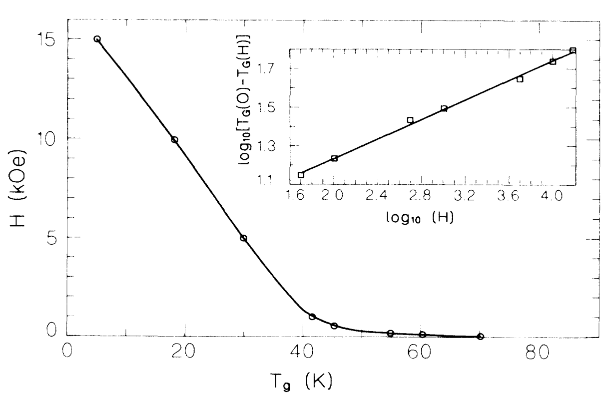

FIG. 8. The glassy temperatures (T_g) as a function of the applied field H. The inset shows the linear fitting to the $\log_{10}$-$\log_{10}$ plot.

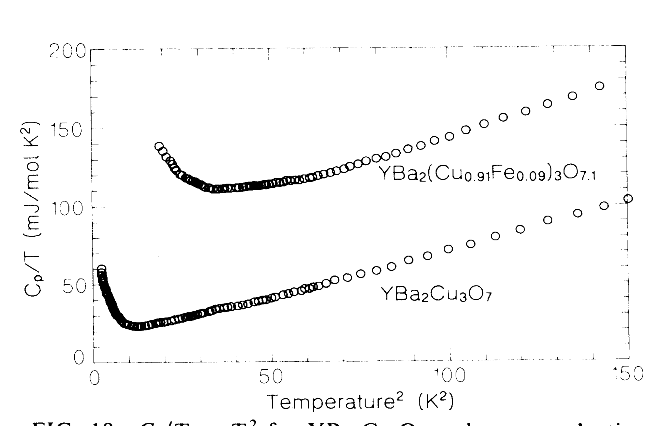

FIG. 10. C_p/T vs T^2 for $YBa_2Cu_3O_7$ and superconducting $YBa_2(Cu_{0.91}Fe_{0.09})_3O_{7.1}$ at low temperatures.

spin-relaxation rate phenomena. Regarding the glassy phenomena of superconducting transition we show that T_g scales with the applied field as $H^{1/4}$. The linear term in the specific heat for the superconducting sample is extremely high. Further work on magnetic properties and specific-heat measurements as a function of Fe concentration and of other dopants like Co and Zn is underway.

ACKNOWLEDGMENTS

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[1]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).

[2]Y. Yeshurun, I. Felner, and H. Sompolinsky, Phys. Rev. B **36**, 840 (1987).

[3]K. A. Müller, K. W. Blazey, J. G. Bednorz, and M. Takashige, Physica B **148**, 149 (1987).

[4]G. Deutscher and K. A. Müller, Phys. Rev. Lett. **59**, 1745 (1987).

[5]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

[6]I. Felner, I. Nowik, and Y. Yeshurun, Phys. Rev. B **36**, 3923 (1987).

[7]M. Mehbod, P. Wyder, R. Deltour, Ph. Duvigneaud, and G. Naessens, Phys. Rev. B **36**, 8819 (1987); Y. Maeno, T. Tomita, M. Kyoguku, S. Awaji, Y. A. Aaki, K. Hoshino, A. Minami, and T. Fujita, Nature (London) **328**, 512 (1987).

[8]E. R. Bauminger, M. Kowitt, I. Felner, and I. Nowik, Solid State Commun. **65**, 123 (1988).

[9]P. Bordet, J. L. Hodeau, P. Strobel, M. Marezio, and A. Santoro, Solid State Commun. **66**, 435 (1988).

[10]X. Z. Zhou, M. Raudsepp, Q. A. Pankhurst, A. H. Morrish, Y. L. Luo, and I. Mattrtense, Phys. Rev. B **36**, 7320 (1987).

[11]S. Nasu, H. Kitagawa, Y. Oda, T. Kohara, T. Shinjo, K. Asayama, and F. E. Fujita, Physica B **148**, 484 (1987).

[12]Z. Q. Qiu, Y. W. Du, H. Tang, J. C. Walker, W. A. Bryden, and K. Moorjani, J. Magn. Magn. Mater. (to be published).

[13]Q. A. Pankhurst, A. H. Morrish, M. Raudsepp, and X. Z. Zhou, J. Phys. C **21**, L7 (1988).

[14]B. D. Dunlap, Physica C **153–155**, 1100 (1988).

[15]Y. Oda, H. Fujita, H. Toyoda, T. Kaneko, T. Kohara, I. Nakada, and K. Asayama, Jpn. J. Appl. Phys. **26**, L1660 (1987).

[16]J. M. Tranquada, D. F. Cox, W. Kunnmann, H. Moudden, G. Shirane, M. Suenaga, P. Zolliker, D. Vaknin, S. K. Sinha, M. S. Alvarez, A. J. Jacobson, and D. C. Jonston, Phys. Rev. Lett. **60**, 156 (1988).

[17]I. Nowik, M. Kowitt, I. Felner, and E. R. Bauminger, Phys. Rev. B **38**, 6677 (1988).

[18]G. Hilscher, N. Pillmayr, R. Eibler, E. Bauer, K. Remschnig, and P. Rogl, Z. Phys. (to be published).

[19]M. Tokumoto, H. Ihara, M. Hirabayashi, K. Murata, N. Terada, T. Matsubara, and Y. Kimura, Physica B **148**, 436 (1987).

[20]E. Sakagami, Y. Oda, Y. Yamada, T. Kohara, and K. Asayama (unpublished).

[21]J. R. Thompson, D. K. Christen, S. T. Sekula, B. C. Sales, and L. A. Boatner, Phys. Rev. B **36**, 836 (1987).

[22]P. Monod and H. Bouchiat, J. Phys. (Paris) Lett. **43**, L45 (1982).

[23]S. von Molnar, A. Torreseen, D. Kaiser, F. Holtzberg, and T. Penny, Phys. Rev. B **37**, 3762 (1988).

[24]Chan-soo Jee, S. Rahman, A. Kebede, D. Nichols, J. F. Crow, T. Mihalisin, and P. Schlottmann, Bull. Am. Phys. Soc. **33**, 465 (1988).