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Disorder and non-linear magnetic response of high T_c superconductors

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Abstract

We measure the low frequency magnetic response of $YBa_2Cu_3O_{6.7}F_x$ ($0 \leq x < 0.2$) ceramics in a wide range of a.c. fields (10^{-7} T $\leq \mu_0H_0 \leq 10^{-4}$ T). When changing the amount of disorder (varying x) on the microscopic level we find the same non linear response with field amplitude H_0 as in granular conventional superconductors. The real part of the susceptibility appears as a universal function of $H_1(T)/H_0$ where $H_1(T)$ is the field of first flux penetration. The power law dependence found for $H_1(T)$ can be understood in the framework of the coherence transition of granular superconductors with random couplings.

1. Introduction

Granularity is an important characteristic of high T_c superconductors ; it plays an essential role in determining their magnetic properties [1-5]. In previous papers we have shown that the low a.c. magnetic field behaviour of $YBa_2Cu_3O_{7-\delta}$ sintered samples [6] and powdered samples [7] was similar to that of conventional granular superconductors made of weakly coupled low T_c grains (grain size $a \gg \xi_s$, the superconducting coherence length)[8]. These systems display a second order phase transition from paracoherence to coherence in the phases of the order parameter of the grains at a critical temperature $T_c < T_{cg}$, the superconducting critical temperature of the grains.

We report here experimental results on the diamagnetic response of fluorinated ceramics to very low fields. The purpose of this paper is to show that the disorder on the microscopic level due to the insertion of fluorine atoms in the oxygen vacancies of the structure leads to the same critical properties as random couplings in granular superconductors. We measure the field of first flux penetration H_1 as a function of temperature and show that this quantity follows the same power law independently of the origin (granular or otherwise) of disorder. A model assuming random couplings between grains has been extended to account for the temperature dependence of H_1 [9]. Furthermore we obtain a universal law for the non-linear susceptibility as a function of the temperature and of the amplitude of the applied field.

2. Experimental procedure and results

$YBa_2Cu_3O_{6.7}F_x$ (x = 0, 0.03 and 0.14) compounds have been synthesized using a non-aggressive solid/gas reaction : $YBa_2Cu_3O_{6.7}$ ceramics are prepared using techniques developped by the chemistry laboratory at Rennes. They are then held at 300°C under diluted NF_3 (3% NF_3 in N_2) during 5 or 20 min. Previous neutron diffraction studies on fluorinated $YBa_2Cu_3O_{6.7}$ powder samples [10] have shown an insertion of fluorine into the structure in (0 1/2 0) sites, the occupancy increasing with x for x <0.2 (the range of this work). At $x \geq 0.2$ this remains practically constant, while the occupancy of the (1/2 0 0) sites increases.

The experimental method used here has been described in earlier papers[8]. We apply a low frequency a.c. field $H = H_0 \sin\omega t$ and record the voltage V from a pick-up coil closely wound around the cylindrical samples. We measure the response of the initial sample (x = 0) and of the two fluorinated samples. They display qualitatively the same typical behaviour as that shown in Fig.1. For different values of the magnetic field amplitude H_0 we plot the normalized quantity :

$$\frac{V}{n\mu_0H_0S\omega} = 1+\chi' \qquad (1)$$

as a function of T. χ' is the real part of the apparent susceptibility, n the number of turns of the coil and S its area.

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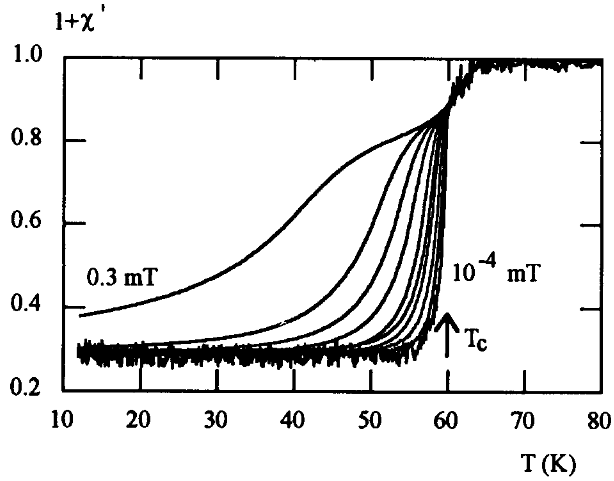

Figure 1. Real part of the apparent susceptibility of the non-fluorinated sample measured at different a.c. magnetic field amplitudes against temperature.

The response of the coil V/μ_0H_0 at a given T when varying H_0 is shown in Fig.2. As in conventional granular superconductors[8] the response is proportional to H_0 in two domains : for very low field amplitudes when no vortex enters the sample during the ac cycle and for large field amplitudes when the induction is close to the value it would have in the case of uncoupled grains.

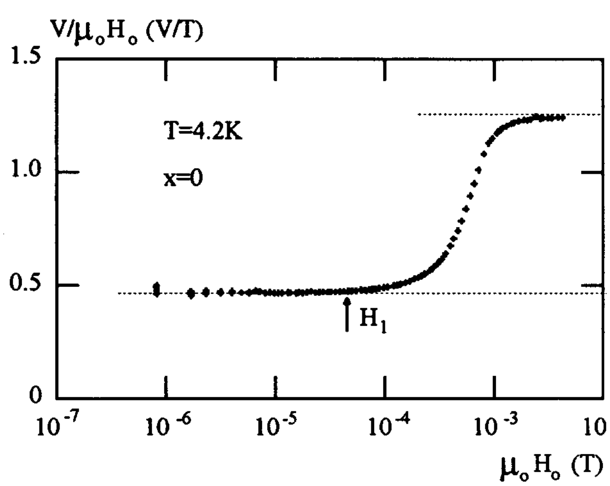

Figure 2. Normalized signal from the pick-up coil V/μ_0H_0 as a function of the field amplitude.

The signal from the pick-up coil is proportionnal to the induction B in the granular material. For $H_0 << H_{c1g}$, the intragrain penetration field, we have $B = \mu H + M_c$, where M_c is the coherent magnetization corresponding to the intergrain screening currents and μ the diamagnetic permeability of the granular medium with uncoupled grains. As we have already seen, weakly coupled granular superconductors display a type II behaviour. For $H_0 << H_{c1c}$ the field of first penetration, Josephson supercurrents will develop in a region of the order of the coherent penetration depth λ_c and $M_c \propto H$, while for $H_{c1c} << H_0 << H_{c1g}$, $M_c \cong 0$.

From measurements in very low fields we determine H_1, the field of first flux penetration in the sample, as the limit above which the response V/μ_0H_0 is no longer constant (Fig.2). The experimental results $H_1(T)$ are plotted as a function of the reduced temperature $(T_c-T)/T_c$ in log-log coordinates (Fig.3). T_c is obtained from the kink of Fig.1 that has been ascribed to the onset of the coherence transition.

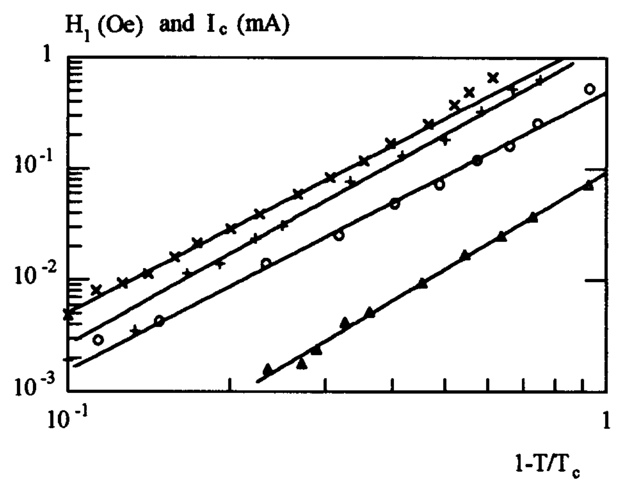

Figure 3. Field of first flux penetration H_1 of $YBa_2Cu_3O_{6.7}F_x$ ceramics (+ : x = 0, o : x = 0.03, Δ : x = 0.14) and critical current I_c of a Nb granular superconductor (×) against reduced temperature.

These plots give straight lines corresponding to the law :

$$H_1(T) = H_1(0)(1 - T/T_c)^{\varphi} \quad (2)$$

with $\varphi = 2.7 \pm 0.2$ for the initial sample and the two fluorinated ones. These results are reported in Table 1 together with the extrapolated value $H_1(0)$. As expected $H_1(0)$ and T_c decrease when x is increased.

Table 1. Characteristic data obtained on $YBa_2Cu_3O_{6.7}F_x$ ceramics.

x	T_c (K)	φ	$H_1(0)$ (Oe)
0	60	2.7	1.32
0.03	58.7	2.5	$4.65\ 10^{-1}$
0.14	54.9	2.9	$9.36\ 10^{-2}$

This power-law dependence on temperature for H_1 suggests the existence of scaling laws and therefore some universal behaviour. In fact, $1 + \chi'$ appears to be a universal function of the variable $H_1(T)/H_0$. This is illustrated in Fig.4a which represents the experimental results for the initial sample when T is varying from 10K to T_c and $\mu_0 H_0$ from 10^{-7} T to 10^{-4} T. The same typical curve is obtained for all the values of x explored. No adjustable parameters are used to obtain these curves. Another example is shown in Fig.4b.

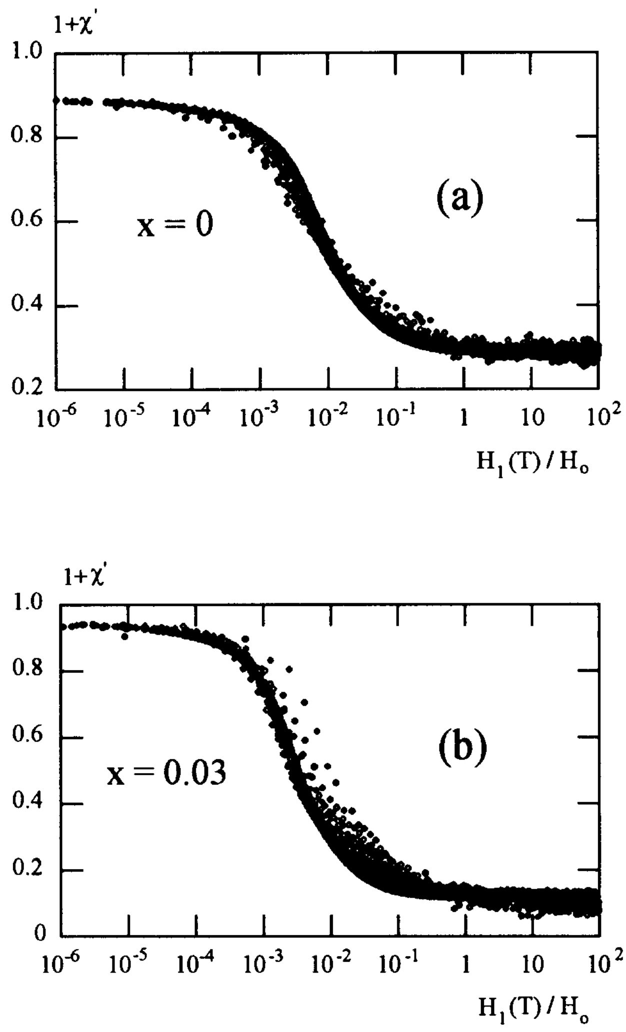

Figure 4. Universal behavior of the susceptibility in $YBa_2Cu_3O_{6.7}F_x$ ceramics (a) x = 0 and (b) x = 0.03 as a function of $H_1(T)/H_0$.

3. Discussion

The use of a varying-amplitude a.c. field only, instead of a superposition of constant a.c. and varying d.c. fields insures that at least for $H_0 < H_1$ the processes we observe are reversible. For $H_0 > H_1$ the experimental results are "artificially" non-hysteretic, hysteresis effects on dissipation appearing in the imaginary part of the susceptibility. The universality of curves in Fig. 4a and 4b is thus quite remarkable, since they concern only non-equilibrium phenomena. In any case, they show the similarity of the effects of intramolecular disorder ($x \neq 0$) and granularity. Indeed, the sample $YBa_2Cu_3O_{6.7}$ is quite probably granular, while oxygen vacancies contribute to disorder at the microscopic level. But it is only the latter type of disorder which is modified in fluorinated samples. Furthermore, the two-plateau behaviour displayed in Fig.2 has been shown by the INSA group [8] to be typical of weakly coupled granular superconductors. In this context, it is worth pointing out that the critical current of the latter follows a law $I_c \sim (1-T/T_c)^{2\beta+\nu}$, with $\beta \cong 0.7$ and $\nu \cong 1.33$ [8,11]. In other words, the exponent of the critical current numerically equals that of H_1. This is an example of Silsbee's rule [12], stating that the current becomes critical when it creates on the sample surface the field of first flux penetration.

One is tempted to apply the classical formulae [12]

$$H_{C1} = \frac{\Phi_0}{4\pi\lambda^2\mu} \ln\frac{\lambda}{\xi} \tag{3}$$

and

$$H_s \cong \frac{\Phi_0}{4\pi\lambda\xi\mu} \tag{4}$$

for the first penetration field in an infinite medium and across a surface, respectively, with $\lambda \sim (1-T/T_c)^{-\beta}$ and $\xi \sim (1-T/T_c)^{-\nu}$. In the Ginzburg-Landau theory, $\beta = \nu = 1/2$, which is clearly not our case. If the exponents found in random granular superconductors are temptatively used, $\varphi = 2\beta \cong 1.4$ in the first case and $\varphi = \beta+\nu \cong 2.0$ in the second, which is still far from our experimental results. A fractal model which solves the discrepancy is described elsewhere in this Conference [9].

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