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CRITICAL FIELD OF HIGH T_c SUPERCONDUCTORS

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Upper critical fields of $YBa_2Cu_3O_{7-\delta}$ have been measured in pulsed magnetic fields up to 27 T. The destruction of the superconductivity when the Y is partially replaced by Pr is confirmed by a decrease of the critical field slope near T_c. We have also studied the effect of applying a high mechanical pressure (8 kbar) during the sample preparation on the critical field as well as the transport properties.

1. Introduction

One of the main problems concerning high T_c superconductivity is the small critical current density ($<10^3$ A/cm^2) found in larger polycrystalline samples at liquid nitrogen temperature. On the other hand, the critical current density derived from magnetization measurements ($\sim 10^6$ A/cm^2) is much larger than the current density obtained from direct transport measurements [1]. The origin of this difference is the granular character of the ceramic samples. The superconducting electrons in neighbouring grains are weakly coupled, probably by the Josephson effect. Near T_c, magnetic fields as small as 0.001 T can destroy the weak superconducting links and a finite resistance appears. In high magnetic fields and for temperatures close to T_c, one can measure the upper critical field B_c where 50% of the normal-state resistance is recovered. From the linear temperature dependence of the critical field, typical of type II superconductors, one infers a coherence length comparable to the size of the unit cell in these high T_c superconductors [2].

Here, we present a detailed study of the critical fields in $YBa_2Cu_3O_{7-\delta}$ compounds. In a first experiment, Y was partially replaced by Pr. Pr is a rare earth element having a stable tetravalent state while most of the rare earth elements have only a stable trivalent state. Using resistivity measurements in zero field, Soderholm et al. [3] have shown that for a Pr concentration larger than 0.5, these compounds become insulators. At the same time, T_c gradually decreases from 90 K to 0 K when the Y concentration decreases from 1 to 0. In the samples with a 0.5 Pr concentration, the Pr^{4+} adds one extra electron to the structure and completely removes the presence of Cu^{3+}. The situation is very similar to the formation of insulating $YBa_2Cu_3O_{7-\delta}$ when the oxygen deficiency $\delta = 0.5$ [4]. In order to improve the intergrain coupling and increase the critical current density, we have also prepared very compact samples of $YBa_2Cu_3O_{7-\delta}$ by sintering this compound under high pressure at elevated temperatures. Different annealing treatments in oxygen which influence the transport properties in a complex way, have been studied in detail.

2. Sample preparation

The $Y_xPr_{1-x}Ba_2Cu_3O_{7-\delta}$ samples, with $x = 1$, 0.96, 0.91, 0.88, and 0.80, were prepared at Argonne National Laboratory, U.S.A. We refer to Soderholm et al. [3] for the details of the sample preparation. The high pressure samples were obtained in our laboratory. Pellets of stoichiometric mixtures were ground and then sintered for 2 h at 550°C under a mechanical pressure of 8 kbar. The temperature was increased to 700°C for 7 h while keeping the

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pressure constant. On cooling, the pressure again remained constant and the temperature was stabilized for 2 h at 550°C and for 10 h at 430°C. Finally, the pressure was reduced to atmospheric pressure and the sample was furnace cooled. After preparation, the samples were cut into bars with typical dimensions 10 mm × 2 mm × 1 mm. The bars were annealed at 400°C, 600°C, 650°C, 720°C, 900°C, and 1050°C in a pure oxygen atmosphere for 17 h followed by a furnace cooling at a rate of 100°C/h.

3. Experimental results and discussion

The experimental results for the critical fields of the $Y_xPr_{1-x}Ba_2Cu_3O_{7-\delta}$ as a function of the reduced temperature T/T_c are shown in fig. 1. The slope of the critical field near $T/T_c = 1$ clearly decreases with increasing Pr concentration. The coherence length calculated from these measurements gradually increases from $\xi(T \to 0) = 1.4$ nm for $x = 1$ towards $\xi(T \to 0) = 2.3$ nm for $x = 0.8$. This increase of the coherence length cannot be explained as a consequence of a reduced elastic mean free path. In a dirty type II superconductor, a smaller mean free path implies larger B_c values. The replacement of Y by Pr rather decreases the superconducting coupling and consequently increases the intrinsic coherence length which varies inversely proportional to the superconducting transition temperature. This is in agreement with the observation of Soderholm et al. [3] that T_c rapidly

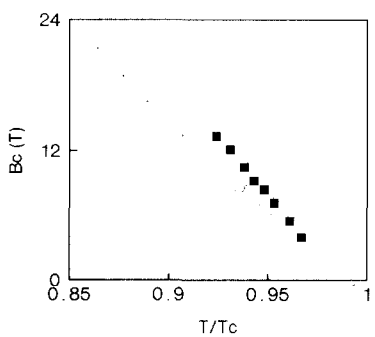

Fig. 1. The critical field B_c versus T/T_c for $Y_xPr_{1-x}Ba_2Cu_3O_{7-\delta}$: $x = 1$ (○), $x = 0.94$ (■), $x = 0.91$ (△), $x = 0.88$ (★), $x = 0.80$ (□).

decreases with increasing Pr concentration. This would also indicate that the metal–insulator transition in the Pr compounds is driven by electronic correlation effects rather than by disorder-induced Anderson localization.

Next, we discuss the experimental results obtained for samples prepared under high pressure. After each annealing, the mass of the sample was carefully determined. Up to an annealing temperature of 900°C no measurable change of the sample mass could be detected. This indicates that no additional oxygen is absorbed by the ceramic structure. The initial sample density was close to 94% of the density for orthorhombic $YBa_2Cu_3O_{7-\delta}$ and was not affected by the annealing procedure up to 900°C. Only after annealing at 1050°C is an appreciable amount of oxygen absorbed. At the same time, the sample becomes more porous and the relative density falls to 0.86, as indicated in table I.

In table I we have also listed the resistivity ρ at 130 K and the T_c value for the different annealing temperatures. Since T_c does not change significantly by the annealing at the lower temperatures, this implies that the superconducting coupling is not affected by the annealing. On the other hand, the sample resistivity decreases by more than an order of magnitude after the annealing at 720°C. We may therefore conclude that the decrease of the sample resistivity is mainly due to an annealing of defect structures. An important part of the defects is probably concentrated near the grain boundaries. This is confirmed by the improved intergrain coupling after annealing as indicated by the increase of the critical current density [5] at $T/T_c \simeq 0.87$ (see table I). The temperature dependence of the upper critical field B_c is shown in fig. 2 for the different annealing temperatures. For annealing up to 900°C, the critical field slope remains unaffected. This result indicates that the coherence length $\xi(T \to 0)$ is a constant and is not influenced by the elastic mean free path. This confirms the results for the samples containing Pr impurities from which we concluded that the small coherence length of the ceramic superconductors is an intrinsic property and is not determined by disorder effects.

Table I
The annealing temperature, the relative sample density, the critical temperature T_c, the resistivity ρ at 130 K, the critical current density J_c at $T/T_c = 0.87$ and the critical field slope dB_c/dT of the $YBa_2Cu_3O_{7-\delta}$ sample prepared under a mechanical pressure of 8 kbar

Ann. temp. [°C]	Rel. dens.	T_c [K]	ρ(130 K) [mΩ cm]	$J_c(T/T_c \simeq 0.87)$ [A/cm^2]	dB_c/dT [T/K]
400	0.94	77	5.70	1.8	−1.0
600	0.94	78	0.55	–	–
650	0.94	79	0.42	17	−1.1
680	0.94	78	0.28	15	−1.0
720	0.94	79	0.12	15	–
900	0.94	77	0.59	–	–
1050	0.86	89	6.20	73	−3.3

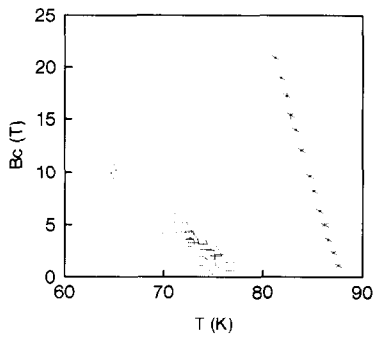

Fig. 2. The critical field B_c versus T for the $YBa_2Cu_3O_{7-\delta}$ sample prepared under a mechanical pressure of 8 kbar. The different symbols refer to the annealing temperature: 400°C (○), 650°C (□), 680°C (△) and 1050°C (★).

After the final annealing treatment at 1050°C, the resistivity jumps to a much higher value. At the same time, the critical current density as well as the critical field slope increase markedly (see table I). As already indicated by the mass measurement, the sample absorbs an important amount of oxygen so that the classical 123 phase with $T_c \simeq 90$ K is recovered. The oxygen absorption causes a change of the electronic band structure near the Fermi level, implying an important change of the normal and superconducting properties. Finally, we can conclude that the preparation of the samples under a high mechanical pressure at 700°C does not improve the critical current density of the high T_c phase since the required annealing at 1050°C destroys the compact sample structure.

Acknowledgements

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