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# High field magnetization of $\text{YBa}_2(\text{Cu,Zn})_3\text{O}_{7-\delta}$ superconductors

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## Abstract

The high field magnetization of sintered  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds was investigated. By means of Bean's critical state model it is derived from magnetization measurements that the slope of critical current density vs. external magnetic field curve becomes less negative and the flux-pinning force diminishes significantly with increasing zinc content. The magnetic field corresponding to the maximum of the pinning force decreases sharply with increasing zinc content. It is thought that the destruction of superconductivity in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  compounds by zinc doping is due to the change in electronic structure in the  $\text{Cu-O}_2$  planes as well as the reduction in orthorhombic distortion.

**Keywords:** High  $T_c$  superconductors; High field magnetization

## 1. Introduction

Since the discovery of the oxide superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  [1–3], many studies on the effect of substitution of copper by transition metal elements have been performed to investigate the superconducting mechanism of the oxide superconductors [4–6]. A prime result of these investigations is that the superconductivity occurs in the  $\text{Cu-O}_2$  planes [6]. It has been a prevailing point of view [5] that the  $\text{Cu-O}_2$  planes ( $\text{Cu}(2)$  sites) are the crucial assemblies in sustaining the high  $T_c$  superconductivity, whereas the existence of the one-dimensional  $\text{Cu-O}$  chains ( $\text{Cu}(1)$  sites), i.e. the oxygen vacancy order structure, plays a much less important role. This is by no means in conflict with the results that  $T_c$  correlates with the number of oxygen ions along the  $\text{Cu-O}$  chains [7,8]. The one-dimensional  $\text{Cu-O}$  chains play a role not only in providing the current carriers, but also in coupling  $\text{Cu-O}_2$  planes which form the real motion channels for the current carriers.

In this paper, we have investigated the influence of zinc substitution for copper on the high field magnetization of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  compounds in the superconducting state.

## 2. Experimental procedures

Zn-bearing samples of nominal compositions  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  with  $x = 0-0.60$  were prepared by a conventional solid state reaction method.  $\text{Y}_2\text{O}_3$ ,  $\text{BaCO}_3$ ,  $\text{CuO}$  and  $\text{ZnO}$  powders with appropriate proportions were well mixed and pre-calcined at  $950^\circ\text{C}$  for 48 h in air. After intermediate pulverizing followed by fine grinding in an agate mortar, they were recalcined at  $940^\circ\text{C}$  for 48 h in air. The recalcined powders were pulverized and ground again and pressed into 18 mm diameter  $\times$  2 mm pellets under a pressure of  $2000 \text{ kgf cm}^{-2}$ . The pellets were sintered at  $940^\circ\text{C}$  for 28 h in air and then furnace cooled. Specimens with a size of  $2 \text{ mm} \times 1 \text{ mm} \times 15 \text{ mm}$  were cut from the sintered pellets and annealed at  $535^\circ\text{C}$  for 24 h and then at  $450^\circ\text{C}$  for 7 h in flowing oxygen.

A.c. susceptibility measurements were carried out on all samples between room temperature and liquid helium temperature to assess the superconducting transition temperature. X-ray diffraction with  $\text{Cu K}_\alpha$  radiation was employed to characterize the phase(s) present in the compounds.

High field magnetization measurements at 4.2 K were performed in the 40 T facility at the University of

Amsterdam. Triangular field vs. time profiles were employed with increasing and decreasing field rates of  $42 \text{ T s}^{-1}$  for measurements up to 11 and 29 T. After each pulse, the specimen was heated to room temperature in order to remove the trapped flux.

### 3. Results and discussion

Fig. 1 shows the powder X-ray diffraction patterns of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  samples ( $x=0-0.48$ ). It is demonstrated clearly that all samples are of single "123" phase with orthorhombic structure. However, the splittings of the (100)–(010), (012)–(102), (103)–(013) and (200)–(020) reflections and the intensities of the (010), (102), (020) reflections become weaker with increasing zinc content. This illustrates that the orthorhombic distortion of the samples is reduced with increase in zinc content. In addition, the intensity of the (005) reflection at  $38.5^\circ$  decreases and even becomes weaker than that of the (113) reflection at  $40.4^\circ$  as the zinc content increases. This reflects that zinc mainly plays a role in the  $\text{Cu-O}_2$  planes, i.e. the bivalent  $\text{Zn}^{2+}$  ions preferentially occupy the Cu(2) sites in the  $\text{Cu-O}_2$  planes [9,10]. The systematic

decrease in the orthorhombic distortion indicates the influence of zinc doping on the  $\text{Cu-O}$  chains. This also implies that the electronic structure of the sample is changed by zinc substitution for copper, which results in the attenuation of the superconductivity.

The diffraction patterns of the undoped samples before and after annealing in flowing oxygen are shown in Fig. 2. It can be seen that the intensities of the (013) reflection at  $2\theta = 32.8^\circ$  are increased after annealing. This indicates that the superconductivity of the samples is increased as a result of the enhancement of the orthorhombic distortion after annealing.

The temperature dependence of the a.c. susceptibility for various  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds is given in Fig. 3. It is apparent that the superconducting transition temperature diminishes significantly with increasing zinc content. As mentioned above, the superconducting links of 123 phase compound should be in the  $\text{Cu-O}_2$  planes. Since Zn ions preferentially occupy the Cu(2) sites [5,6,9,10], zinc doping will severely destroy the superconducting links in the 123 phase compound as it alters the electronic structure of the Y-based oxide superconductors and reduces the orthorhombic distortion in these compounds.

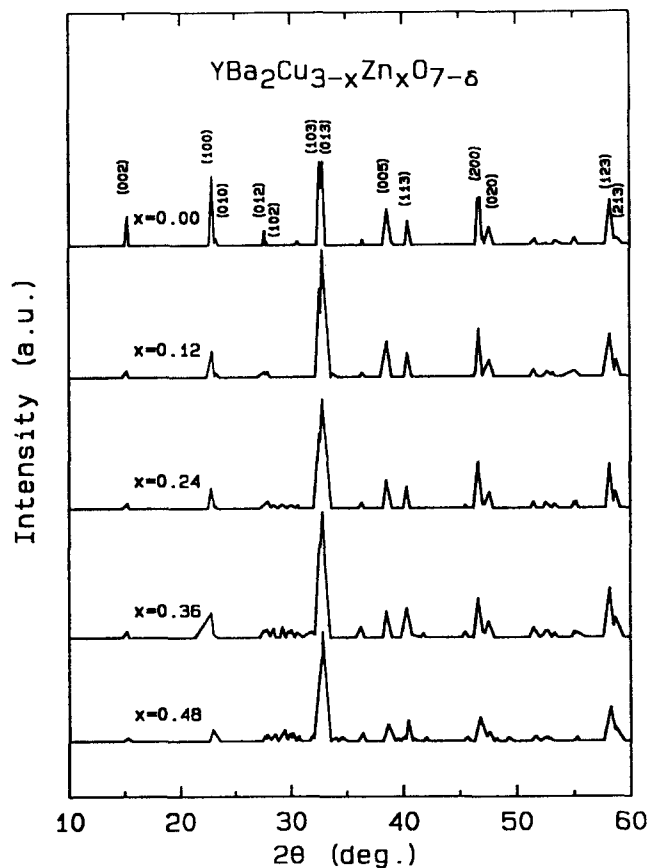


Fig. 1. Powder X-ray diffraction patterns of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds with  $x=0-0.48$ .

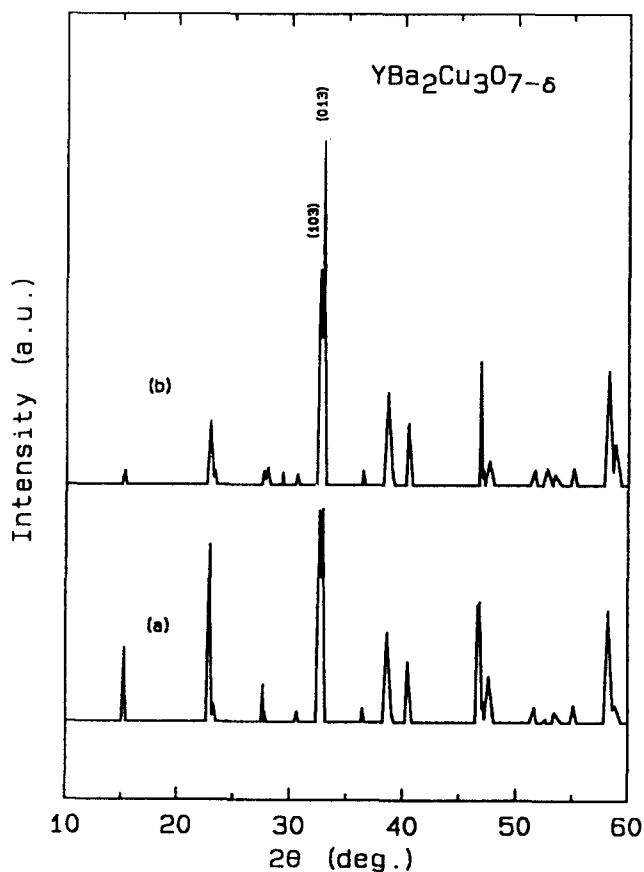


Fig. 2. X-ray powder diffraction pattern of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  before and after annealing in flowing oxygen: spectrum a, before annealing; spectrum b, after annealing.

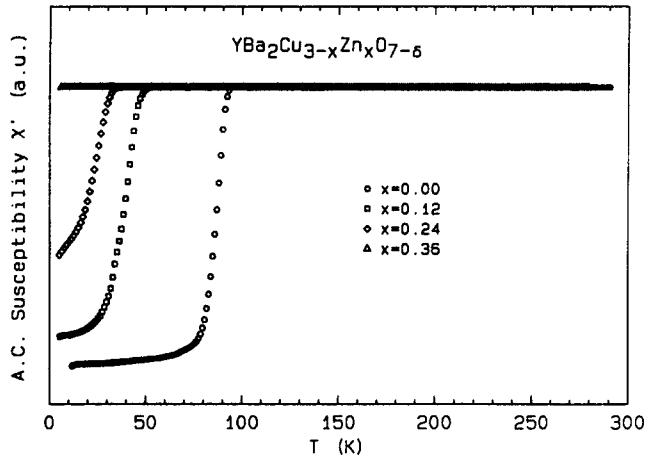


Fig. 3. Temperature dependence of the a.c. susceptibility of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds with  $x = 0-0.36$ . The critical temperature decreases significantly with enhancement of Zn content.

Fig. 4 exhibits the irreversible magnetization curves of various  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-x}$  compounds at 4.2 K. The dependence of the magnetization on the external magnetic field shows that the magnetization is drastically reduced with enhancement of Zn content. This result is also consistent with the observations in Fig. 3, that the superconducting transition temperatures of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds decrease markedly with increasing Zn content.

The critical current density  $J_c$  of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds vs. the external magnetic field  $B$  at 4.2 K is presented in Fig. 5.  $J_c$  was calculated by means of Bean's critical-state model [11]. In this model,  $J_c = \Delta M/Vd$ , where  $\Delta M$  represents the difference between the magnetic moments measured in increasing and decreasing magnetic fields as shown in Fig. 4,  $V$  the effective volume of the specimen and  $d$  the thickness. Since the length of the sample is much larger than its thickness, the demagnetization effect of the specimen may be neglected. From Fig. 5, it can be

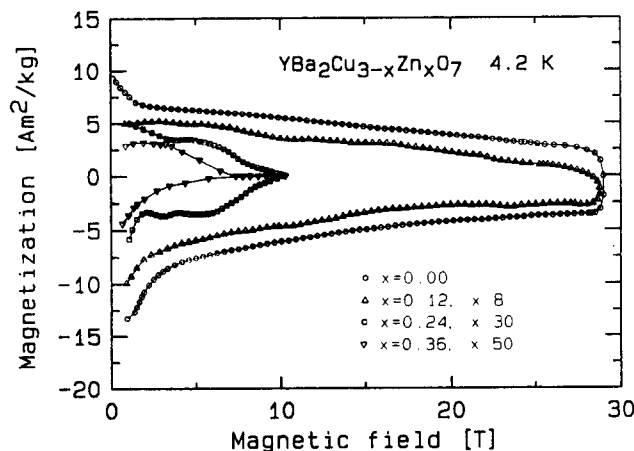


Fig. 4. Field dependence of the magnetization of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds with  $x = 0-0.36$  at 4.2 K.

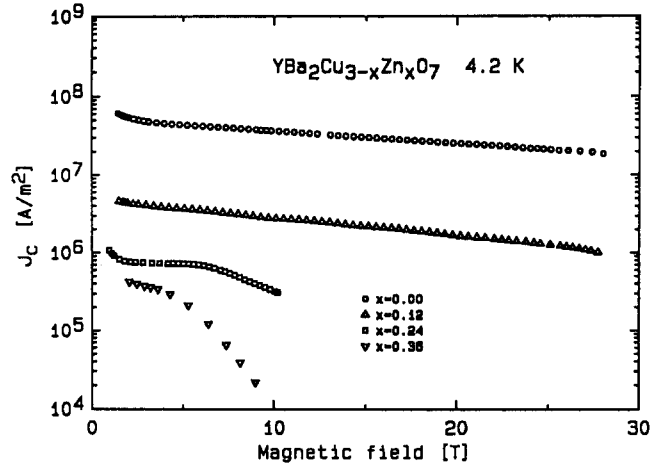


Fig. 5. Dependence of the critical current density of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds with  $x = 0-0.36$  on the applied external magnetic field at 4.2 K.

seen that  $J_c$  decreases with increasing applied magnetic field. However, the slope of the  $J_c$  vs.  $B$  curve is mild for zinc content  $x \leq 0.12$ .

The volume density of the flux-pinning force  $F_p$  was calculated with the formula  $F_p = J_c \times B$ . The field dependence of  $F_p$  in  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds at 4.2 K is shown in Fig. 6. It is apparent that  $F_p$  also decreases remarkably with increase in zinc content. This demonstrates that zinc replaces copper in the 123 phase, resulting in the weakening of retardation of flux motion due to the reduction in the lattice distortion.

For all specimens, the pinning force initially increases to a maximum value and then drops with further increasing external magnetic field. Considering the validity of Bean's critical-state model, more and more flux is trapped with increasing external magnetic field as long as the flux-trapping capacity of the sample is not saturated, so that the macroscopic flux-pinning force increases. However, when the saturation of flux-

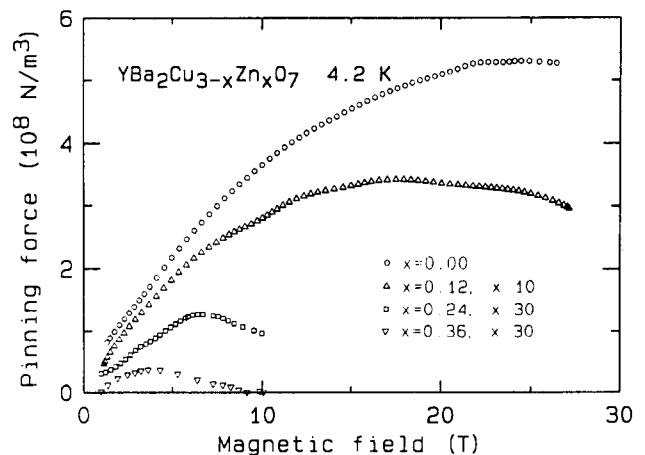


Fig. 6. Field dependence of the flux-pinning force of  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  samples with  $x = 0-0.36$  at 4.2 K.

trapping capacity is reached, the trapping is in equilibrium with the depinning. After that, the amount of flowing flux increases and the pinning force decreases with increasing external magnetic field. From Fig. 6 it can be seen that the maximum of the pinning force decreases notably with increasing zinc content.

#### 4. Conclusions

(1) The superconductivity of the sintered  $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_{7-\delta}$  compounds is severely destroyed by Zn doping owing to a reduction in orthorhombic distortion and the change in electronic structure in the Cu–O<sub>2</sub> planes.

(2) The slope of the  $J_c$  vs.  $B$  curve varies with the amount of Zn doping. For  $x \leq 0.12$ ,  $J_c$  is little affected by an increase in external magnetic field.

(3) The flux-pinning force increases to a maximum value and then drops with increasing external magnetic field. The maximum flux-pinning force decreases remarkably with increasing zinc content.

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