# Transport properties and magnetic hysteresis of the high $T_c$ superconductor EuBa<sub>2</sub>(Cu<sub>1-x</sub>Fe<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub>

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### Abstract

The compounds  $\operatorname{EuBa}_2(\operatorname{Cu}_{1-x}\operatorname{Fe}_x)_3\operatorname{O}_{7-\delta}$  have been prepared in order to study the effect of partial replacement of copper by iron on the transport properties, superconductivity and magnetic behaviour of this oxide system. The normal state conduction gradually changes from metallic to semiconductor like as the iron concentration increases. From the hysteresis loops recorded at 10 K, the values of the lower critical field and magnetization critical current densities are determined.

### 1. Introduction

As a result of the research initiated by the work of Bednorz and Muller [1], three families of materials exhibiting superconductivity with critical temperatures above 77 K have been discovered to date. If the compositions derived from the Bi-Sr-Ca-Cu-O [2] and Tl-Ba-Ca-Cu-O [3] systems have the highest  $T_c$  (80-110 K and 120 K respectively), the Y-Ba-Cu-O system [4] (with  $T_c = 90-94$  K) is today the best known and seems the nearest to industrial application.

The problem of partial substitution of copper by a magnetic ion has been well studied in the 1-2-3 system. The situation is complicated by the presence of two non-chemically equivalent sites for Cu(1) and Cu(2); the dopant can occupy one of the two sites, or even the two sites at the same time [4]. Therefore experiments on the substitution of copper in  $YBa_2Cu_3O_{7-\delta}$  produced interesting results. Tarascon et al. [5] substituted copper by different concentrations of aluminium, iron, cobalt, nickel and zinc. They observed an orthorhombic-tetragonal transition in the case of aluminium, iron and cobalt, and also a strong correlation between the oxygen proportion, the peculiarity and the quantity of the dopant. Strobel et al. [6] found that substitution by a non-magnetic ion such as zinc has a strong effect on the superconducting properties.

In the present work, the results of transport and magnetic properties with hysteresis cycles at high field of  $\text{EuBa}_2(\text{Cu}_{1-x}\text{Fe}_x)_3\text{O}_{7-\delta}$  with x = 0, 0.025, 0.05 and 0.075 will be discussed.

## 2. Preparation and experimental techniques

The Eu<sub>2</sub>O<sub>3</sub>, BaCO<sub>3</sub>, CuO and Fe<sub>2</sub>O<sub>3</sub> powders (purity of 99.99%) were mixed in the desired proportions and calcined at T = 920 °C in an alumina crucible. The product was ground, compacted and heated at 900 °C. The product was ground a second time, compacted and reheated at 900 °C. The pellets were after that annealed at 400-500 °C under oxygen for 48 h. The last annealing was followed by slow cooling to room temperature.

The electrical measures were performed by the Van der Pauw method [7] using both a continuous current and an alternating current at 80 Hz; the temperature was measured with a carbon probe (below 77 K) or a platinum probe (between 77 K and 300 K). For the electrical contact, indium was welded by an ultrasonic process.

The magnetization M = f(H) was measured at T = 10 K with H = 0-15 kOe by using a vibrating magnetometer.

### 3. Results and discussion

The fact that the majority of classical superconductors with  $T_c < 40$  K lose their superconductivity when a weak quantity of magnetic impurity is introduced (Arbrikovsov and Gorkov's theory [8]) led us to consider the substitution of copper by iron. In Fig. 1 it is possible to see the characteristics of the resistivity for all the prepared samples. A change from metallic to semiconductor properties can be seen.



Fig. 1. The temperature dependence of the electrical resistivity for  $EuBa_2(Cu_{1-x}Fe_x)_3O_{7-\delta}$ .



The  $T_c$  value (R = 0) decreased continually with the increase in iron (Fig. 2) with  $T_c = 92.7$  K for x = 0 to  $T_c = 47$  K for x = 0.075. Our results agree perfectly with those of refs. 9 and 10; we think that the gradual change in resistivity may come from the localization of the charges or the decrease in the carrier density [11].

The measurements of the magnetization M = f(H)were also performed. The cycles, namely high field cycles, as opposed to low field cycles (the results of the latter will be published soon), were obtained by cooling the samples systematically in zero field, going from a temperature  $T > T_c$  to the measurement temperature. They reveal three characteristic fields [12, 13]  $H_{c_1}$ ,  $H_m$ and  $H_{c_2}$ .  $H_{c_1}$  and  $H_{c_2}$  have the usual meanings; they indicate the beginning of the entry of the vortex for  $H_{c_1}$ and the transition to the normal state for  $H_{c_2}$ .  $H_m$ defines the field for which the magnetization is the highest.

Only  $H_{c_1}$  and  $H_m$  will be discussed, because the determination of  $H_{c_2}$  needs measures of the magnetoresistance [14] in pulsed fields.

The magnetization data at T = 10 K of sintered pellets for x = 0, 0.025, 0.05 and 0.075 as a function of H are shown in Fig. 3. As the field is increased, a



Fig. 3. The magnetization hysteresis loops at 10 K for  $EuBa_2(Cu_{1-x}Fe_x)_3O_{7-\delta}$  with  $0 \le x \le 0.075$ .

diamagnetic signal appears as a result of the shielding effect. The diamagnetism is linearly proportional to the applied field up to  $H_{c_1}$  after which it deviates from the linear behaviour owing to penetration of the field in the interior of the samples [15, 16].

The  $H_{c_1}$  and  $H_m$  values are gathered in Table 1. We can see that, as the concentration of iron increased, these fields decreased, which is in agreement with the results of ref. 17. The tendency shown on our cycles confirms also the decrease in the diamagnetic moment as it is replaced by paramagnetic moments [18], which

TABLE 1.  $T_c$ ,  $H_{c_1}$  and  $H_m$  values for various concentrations x

$T_{\rm c} \ (R=0) \ ({\rm K})$	$H_{c_1}$ (Oe)	$H_{\rm m}$ (Oe)
92.7	300	2000
81.5	115	765
56.7	73	430
47.6	40	110
	$T_{c} (R = 0) (K)$ 92.7 81.5 56.7 47.6	$T_{\rm c} \ (R=0) \ ({\rm K})$ $H_{\rm c_1} \ ({\rm Oe})$ 92.730081.511556.77347.640

demonstrates the coexistence of the superconductivity independently of the paramagnetism [10].

The correct theoretical evaluation of the critical current value in ceramic compounds seems to be very difficult, according to the inhomogeneity of the magnetic field trapped in the volume and to the shape of the grains. Until now, Bean's model [19] has been used as the theoretical basis. Experimentally, this model can be used by taking the  $M^+$  and  $M^-$  curves of the hysteresis cycles. The critical current density  $J_c$  is evaluated as proportional to  $(M^+ - M^-)/r$ , where r can be taken as the grain dimension, but as we are just concerned with the relative variation in  $J_c$ , we have plotted  $\delta M = f(H)$ for different concentrations at T = 10 K (Fig. 4) and also  $\delta M = f(x)$  with H = 2 kOe at T = 10 K (Fig. 5).

A decrease, as expected, in  $J_c$  with increasing field that is almost exponential [20] can be seen. We agree with Grover *et al.* [21] that the behaviour of  $J_c$  is due to the shortness of the coherence length, considering the doping with iron, the latter decreasing still further the pinning strength.



Fig. 4. The temperature dependence of  $\delta M$  at 10 K for EuBa<sub>2</sub>(Cu<sub>1-x</sub>Fe<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub>.



Fig. 5. The dependence of  $\delta M$  on the substituent concentration x at 10 K (H = 2 kOe).

## 4. Conclusion

The superconductivity and magnetic properties of the superconductor  $EuBa_2(Cu_{1-x}Fe_x)_3O_{7-\delta}$  were examined. The transport measurements revealed the prominent part taken by the copper, substituted partially by a magnetic ion. The presence of a high irreversibility regime arising from the pinned vortex in the grain was also shown up by the cycles at high field. The use of Bean's model allowed us to see the real variation in the critical current in the field range 0–15 kOe at 10 K, and its variation with the dopant concentration.

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