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Remarkable influence of heat treatment on AC irreversibility line and flux pinning properties in $(Y_{1-x}Eu_x)SrBaCu_3O_{6+z}$

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Polycrystalline samples $(Y_{1-x}Eu_x)SrBaCu_3O_{6+z}$ have been synthesized by solid state reaction method with two distinguished heat treatment. The irreversibility line and flux pinning proprieties (pinning force density) has been investigated by AC susceptibility measurements. The experimental data for irreversibility line fit well with the power law relation: $H=(1-T/T_c)^n$. Results show that the effect of annealing under argon flux leads to an enhancement of irreversibility line and an increase of pinning density force for high applied magnetic field which is justified by the improvement of crystallographic quality and the intergranular coupling of the samples heated under argon flux [AO].

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1. Introduction

Recent progress in the melt-processing of bulk high-temperature superconductors has created a basis for various engineering applications. For later, high critical current density J_c and irreversibility field B_{irr} are substantial. The irreversibility field, B_{irr}, is an important parameter representing the maximum applicable magnetic field for a practical use of superconductor at which critical current density, J_c, is reduced to zero and which is determined in terms of a criterion of the critical current density J_c. With respect to an effective cooling, the most interesting temperature for applications is that of liquid nitrogen, 77.3 K. Therefore, high J_c and B_{irr} at this rather high temperature are extremely desired.

X Ray diffraction (XRD) and AC susceptibility ($\chi_{ac} = \chi' + i\chi''$) are very useful for characterizing high T_c superconductors. χ' reflect the shielding ability while χ'' is measure of the magnetic irreversibility.

AC susceptibility is a non destructive technique that plays an important role in studying the high temperature superconductors. The complex AC susceptibility provides information about several physical properties of high T_c superconductors [1-3]. We cite the irreversibility properties (pinning properties). A high irreversibility field B_{irr} is very important as far as the practical applications of the materials are concerned. The irreversibility field is the line in the H-T plane that divides the irreversibility region from reversible region [4].

Mechanism of pinning vortex in type II of superconductors was found to be more dependent on the granular characteristic of those compounds [5]. High treatment has been shown to be effective in fabricating dense YBCO bulk superconductors with good alignment of grains and minimum weak links between grain boundaries.

We reported in our previous work [6], that for the high-T_c superconductors $(Y_{1-x}Eu_x)(SrBa)Cu_3O_{6+z}$, the [AO] treatment increases the orthorhombicity ε and the critical temperature T_c for $x \geq 0.45$, as well as the shielding effect S (amplitude of $\chi'(T)$) for $x > 0.5$. In the normal state, the [AO] treatment reduces the linear resistivity parameters, indicating a diminution in the interaction between carrier charges and phonons.

For further investigation of the effect of heat treatment on the $(Y_{1-x}Eu_x)(SrBa)Cu_3O_{6+z}$, we focus in this study on the AC irreversibility line and flux pinning properties. The preparation steps were explained in details. The AC magnetic susceptibility measurements were carried out for x=0, 0.2, 0.4, 0.5, 0.6, 0.8, 1.0.

2. Experimental techniques

The polycrystalline samples $(Y_{1-x}Eu_x)(SrBa)Cu_3O_{6+z}$ were prepared using solid-state sintering of the respective oxides and carbonates. The chemicals were of 99.999% purity except in the case of $BaCO_3$ which it was 99.99% pure. Eu_2O_3, $SrCO_3$, $BaCO_3$ and CuO were thoroughly mixed in required proportions and calcined at 950°C in air for a period of 12-18 hours. The resulting product was ground, mixed, pelletized and heated in air at 980°C for a period of 16-24 hours. This was repeated twice. The pellets were annealed in oxygen at 450°C for a period of 60-72 hours and furnace cooled. This was denoted as sample [O] for each x.

XRD data of the samples were collected with Philips diffractometer fitted with a secondary beam graphite monochromator and using CuKα (40 kV/20 mA) radiation. The angle 2θ was varied from 20° to 120° in steps of 0.025°and the counting time per step was 10 sec. The XRD specters were refined with Rietveld refinement.

Superconducting transitions were checked by measuring both the real (χ') and the imaginary (χ'') parts of the AC magnetic susceptibility as a function of temperature under a field of 0.11 Oe and at a frequency of 1500 Hz. In addition, χ' and χ'' were measured in a static field ($0 < H=H_{dc} < 150$ Oe) superimposed on the AC field of H_{ac} =0.11 Oe.

The resistivity ρ(T) was measured using the Van Der Pauw method. T_c was checked by measuring χ' (T) and confirmed by the measure of the ρ(T).

For each x, the same sample [O] was then heated in argon at 850 °C for about 12 h, cooled to 20 °C and then oxygen was allowed to flow instead of argon and the sample was annealed at 450 °C for about 72 h. This sample is denoted as [AO]. Resistivity and AC susceptibility measurements were done on a part of this sample.

3. Results and discussion

The XRD patterns of all the samples allowed clear identification of the orthorhombic splitting and observation of some weak unidentified impurity peaks in case of [O] samples. These impurity peaks were eliminated in the samples [AO]. This indicates an improvement of crystallographic quality after heat treatment under argon [6].

A magnetic field harmonically varying in time (to probe the sample) and the lock-in technique (to register the sample's induced magnetic response sensed by a pick-up coil) are often used to study the electromagnetic properties of superconductors. The measure of the temperature dependence on the complex AC susceptibility is the most common performed experiment. Below T_c, the sharp decrease in the real part $\chi'(T)$ indicates the manifestation of diamagnetic shielding, whereas the peak T_p in the imaginary part $\chi''(T)$ represents the A.C. losses, from which we obtain the irreversible magnetic field.

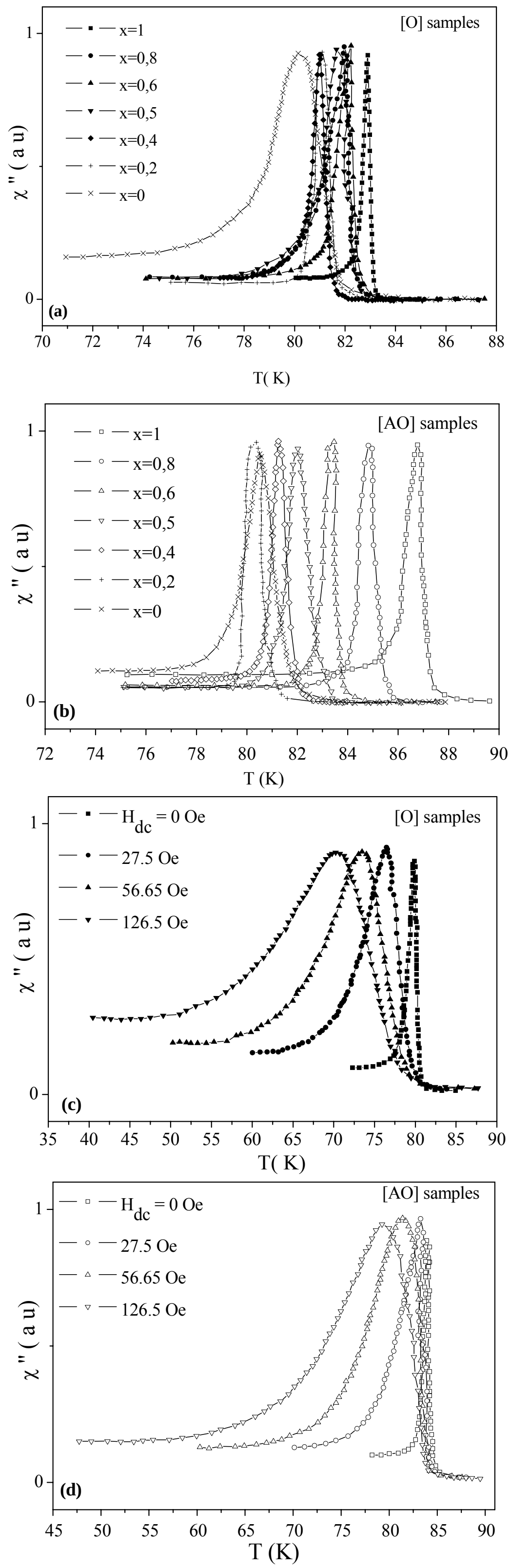

Fig. 1. (a,b) χ'' of $Y_{1-x}Eu_xSrBaCu_3O_{6+z}$ as a function of the temperature and heat treatment and (c,d) χ'' of de $EuSrBaCu_3O_{6+z}$ (x=1) as a function of the temperature and heat treatment at four fields H_{dc} ($0 < H_{dc} < 126.5$ Oe).

As can be seen in Fig. 1, the effect of [AO] heat treatment on T_c was remarkable. The temperature at which the diamagnetism sets in is taken as T_c and it was found to be dependent on both x and the heat treatment employed. Since the same sample was used for both heat treatments, one can compare the hysteresis losses response (the pick T_p and area under $\chi''(T)$) and note that the motion of intergranular vortices of the [AO] sample increased considerably compared to that of the [O] sample for each x (see for example the case x=1in Fig. 1b and Fig. 1d).

The intergranular peak is due to losses associated with flux penetrating within the grains boundaries. This peak can also be called the coupling peak and can be explained by considering the weak links or Josephson junction (JJ) coupling among adjacent superconducting grains. This peak is expected to be observed at low magnetic fields because of the weak JJ coupling in polycrystalline superconductor. The intergranular peak at T_p indicates that the maximum hysteresis losses is due to the motion of intergranular vortices [7]. When x increases, T_p shifts to high temperatures indicating the very weak intergranular flux pinning as observed in our [AO] samples [8].

In order to investigate the field dependence of the intergranular peak T_p, we performed the measurements of AC susceptibility in the field lying between 0Oe and 126.5Oe. Fig. 1c and Fig. 1d depicts the obtained results.

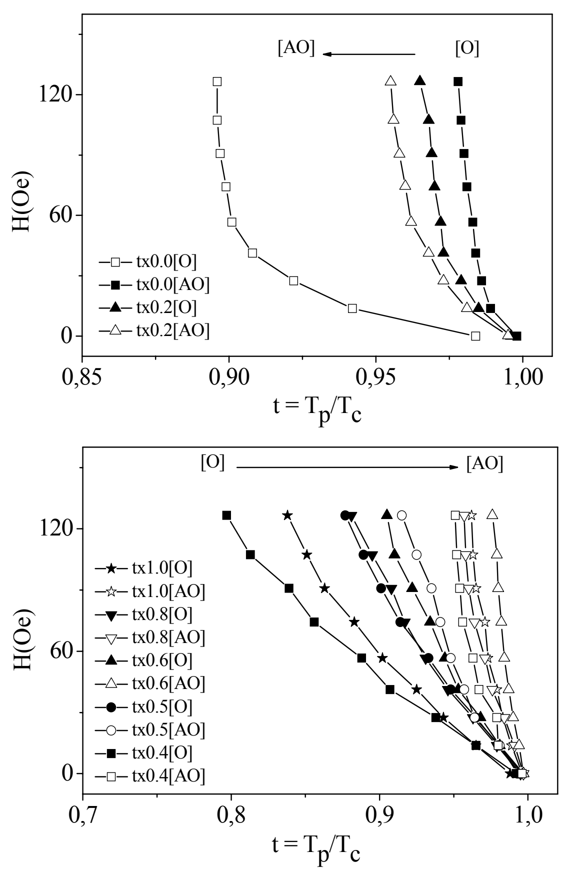

Fig. 2. Variation of $t = T_p/T_c$. as function of x(Eu), H_{dc} and the heat treatment of $Y_{1-x}Eu_xSrBaCu_3O_{6+z}$.

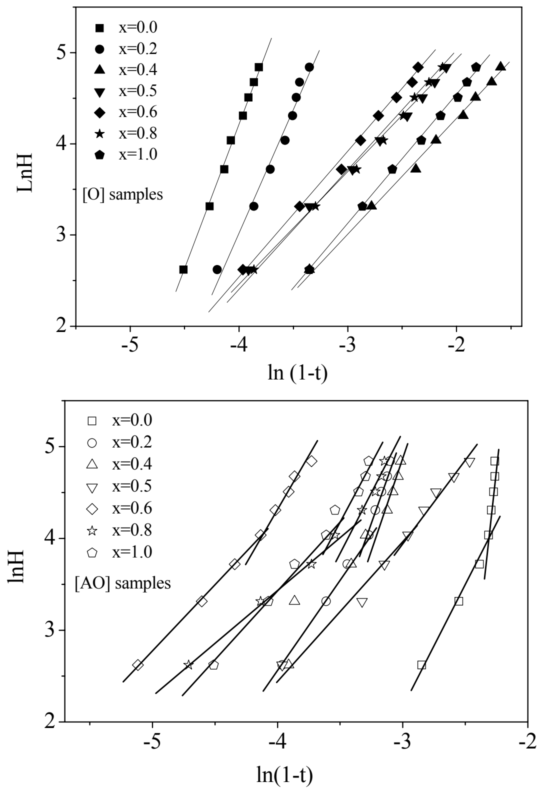

Fig. 3. The double logarithmic plots of irreversibility field versus $[1-(T_p/T_c)]$ for all samples. [O] samples: a), [AO] samples: b). The slope of the line gives the 'n' and K' value for each sample.

Looking to the imaginary part of the AC susceptibility $\chi''(T)$ of the sample $EuSrBaCu_3O_{6+z}$ (x=1), we can see that the width of transition for all fields is smaller in the case of [AO] samples, and the peak T_p shifts less with the applied field compared to that of the sample [O]. In fact, when H is plotted as a function of $t = T_p / T_c$ (Fig. 2), an enhancement of the irreversibility line was observed after argon treatment for $x \geq 0.45$. The obtained curves can be fitted using the following equation [6].

$$H = K'(1-t)^n \tag{1}$$

K' et n, are constant, K' may be interpreted as the field necessary to reduce the intergranular critical cur-rent to zero in the limit of T_p=0.

To simplify the processing, we use the double logarithm of the equation (1). Fig. 3 illustrates the results. Two values of the parameters K' and n were observed in the case of [AO] samples.

Table 1 shows the exact measured values of the superconducting parameters for each sample as a function of the heat treatment. In case of x=1, the value of K' was estimated to be equal to 1600 Oe for [O] samples, while it is increased to reach K_1'=16400 Oe and K_2'=150000 Oe for [AO] samples. We note that the argon treatment considerably increases the value of K' and n, indicating an improvement in the pinning properties (Fig. 4).

Table 1. Superconducting, structural and magnetic parameters of $Y_{1-x}Eu_xSrBaCu_3O_{6+z}$.

x	0		0.2		0.4		0.5		0.6		0.8		1.0	
h Treat	[O]	[AO]	[O]	[AO]	[O]	[AO]	[O]	[AO]	[O]	[AO]	[O]	[AO]	[O]	[AO]
T_c (K)	83	81.7	82.35	80.6	82.17	81.84	82.1	82.47	82.45	83.38	82.05	85.07	81.1	87.05
T_p (K)	82.9	80	82.08	80	81.73	81.25	81.6	81.9	82.2	83.12	81.6	84.8	80.1	86.7
ε (10^{-3})	8.23	9.90	7.89	9,32	6.33	8.79	5.72	8,35	5.49	7.71	4.18	7.62	3.1	6.97
K′ [O], K_1′,[AO](Oe)	2,7E7	1,9E4	9E5	4,8E4	8,6E2	9,9E4	1,5E3	3,5E3	2,9E3	2,1E4	1,8E3	2,9E3	1,6E3	1,6E4
K_2′ [AO](Oe)	-	1,3E16	-	1,5E8	-	3,3E5	-	3,3E4	-	5,4E5	-	1,6E5	-	1,5E5
n[O] , n_1[AO]	3.22	2.56	2.68	2.06	1.24	2.16	1.19	1.56	1.19	1.44	1.12	1.34	1.19	1.5
n_2[AO]	-	14.29	-	4.51	-	2.66	-	2.15	-	2.22	-	2.19	-	2.19

The T_p dependence on external magnetic field has been investigated to check the effect of partial substitution on the intergranular pinning force. Using the critical state model, Muller obtained the following equation [5]:

$$\left(1-\frac{H_{ac}}{H_j}\right)^2 = 1+\frac{2d\cdot f_j(T)}{\mu_0\mu_{eff}(T)H_f^2} \qquad (2)$$

Where H_j is characteristic field of the Kim-Anderson critical state model, d the sample thickness, f_j is the pinning force density and μ_{eff} the effective permeability of grain medium.

The temperature dependence of the pinning density force can be described by the following equation [7]:

$$f_j(T) = f_j(0)(1-t)^q \qquad (3)$$

Where $f_j(0) = \dfrac{K^2\cdot\mu_0\mu_{eff}}{2d}$ and $q = 2n$

The intergranular pinning force density $f_j(T)$ (Eq 3) of our samples fits with the power lower (Eq 1):

Fig. 4a shows that when x increase, the indice n decrease to 1.5 given by Muller for YBaCuO [5]. For each x, the [AO] heat treatment increase n_2[AO], and n_1[AO] for x > 0.35. In Fig. 4b, K′ follows the same allure than n. A maximum of increase was observed in K'_2[AO] for x=0. These results indicate a remarkable enhancement of the irreversibility line due to the argon heat treatment for x ≥ 0.4. We note that the argon treatment considerably increases the value of K′ indicating an improvement in the pinning properties. These results are justified by our XRD spectra, with Rietveld refinement, which means that an improvement of crystallographic quality occurs after argon traitement [AO] [4]. That means an enhancement of the quality of the grains and intergrain contacts and coupling by Josephson junctions. This argument is justified by the fact that the difference between inter- and intragrain currents vanishes and the two peaks in χ″(T) merge [9] as in our case in Fig. 1.

In our previous study in [6], we observed that when x increase from 0 to 1, ε decreases with T_c[O]. However, T_c[AO] decreases with ε until x = 0.2 and after it increases by 6 K to 87 K for x =1 [AO] (Fig. 5). The crystalline parameter b is constant but a (and c) increase indicating an increase of the number of oxygen atoms by chain (NOC) along a axis leading to a decrease of ε (T_c[O]) from orthorhombic toward tetragonal structure for x=1 [O]. Whereas for each x, the [AO] treatment increases the orthorhombicity ε = (b-a)/(b+a) (for 0≤ x ≤1), T_c (for x ≥ 0.4) and the distance d[Cu(1)-O(1)] (decrease T_c) for x< 0.45 and decrease it (increase T_c) for x> 0.45 (Fig. 6). This enhance the transfer of the holes from chains along b axis to the superconductiong plans $Cu(2)-O_2$ and hence increase T_C.

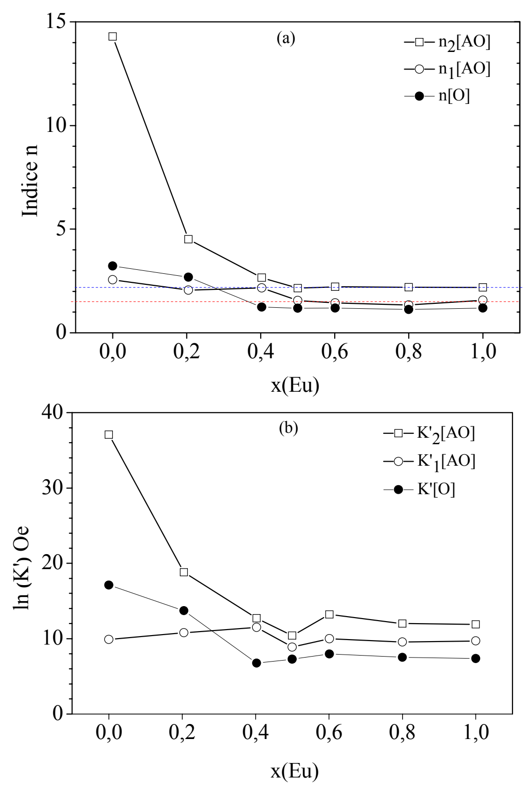

Fig. 4. (a) The exposant n, (b) the field K' as a function of x and heat treatment of $Y_{1-x}Eu_xSrBaCu_3O_{6+z}$.

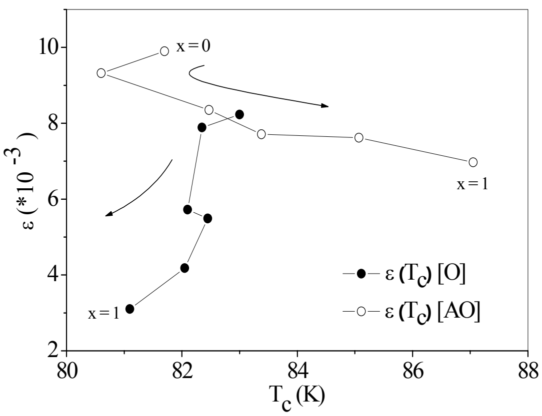

Fig. 5. Variation of the orthorhombicity ε as a function of T_c and heat treatments of $Y_{1-x}Eu_xSrBaCu_3O_{6+z}$.

For each x, the [AO] heat treatment makes decreases a and increases b [6]. This increases the (NOC) along b axis (decrease the cationic disorder, of Y/Sm on the Ba/Sr site, along c axis and increase the anionic order in the basal plane) leading to an increase of the number $p_{sh}(x)$ of hole s by Cu(2)-O_2 superconducting planes (deduced from the under saturation zone of the universal relation T_c/T_{cmax} (p_{Sh}) [10]) and T_c for $x \geq 0.4$. For each x, the increase of d[Cu(1)-O(1)] distance (for $x \geq 0.45$) (Fig. 6) can be seen as an argument in the increase of the formation of the Cooper pairs via the increase of p_{sh} (for $x \geq 0.4$) and increase of T_c. At low temperatures in the vortex state, pinning of the flux lines may be very effective. It weakens, when approaching the peak T_P, where depinning may lead to dissipation. For each x, the [AO] treatment increase the number of vacant sites O(5), along a axis in the basal planes, which act like pinning centers of the vortex and increase the area under the irreversibility line in (H,t) phase diagram (Fig. 2) for the samples [AO].

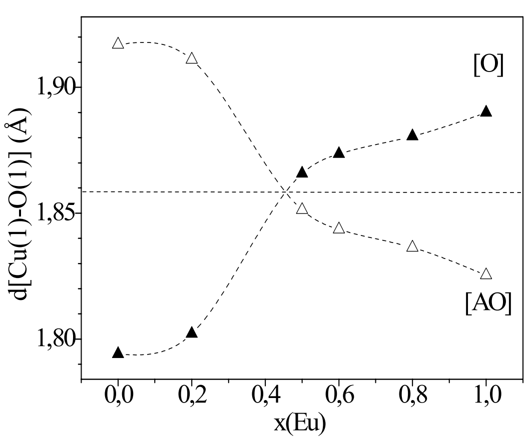

Fig. 6. The inter atomic distance d[Cu(1)-O(1)] as a function of x and heat treatment of $(Y_{1-x}Eu_x)SrBaCu_3O_{6+z}$

The two arguments (cationic and anionic disorders) are justified here by the five remarkable correlations observed between $T_c(x)$, the volume of the unit cell V(x) and the number $p_{sh}(x)$, and on the other hand, between $\delta T_c(x) = T_c[AO]-T_c[O]$ and $\delta\varepsilon(x)$ and between $\delta T_c(x)$ and $\delta K'(x)$ in [6]. So the structural, electrical and superconducting properties are correlated with the effect of argon heat treatment.

3. Conclusions

In the samples [AO], the remarkable improvement in the irreversibility line (for $x \geq 0.4$) is explained by the improvement of the quality of the grains and intergranular coupling and to the improvement of the pinning properties and crystallographic quality of these samples. These results are the outcome of interplay between cationic disorder along the c axis and oxygen disorder in basal plane. A combination of several factors such as decrease in d[Cu(1)-O(1)] for x>0.4; increase in cationic and chain oxygen ordering; p_{sh} and in-phase purity for the [AO] samples may account for the observed increase of T_c for $x \geq 0.4$.

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