Effect of partial substitution of Ca by 4f elements on dissipative processes in Bi:2223 superconductors

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Abstract. (Bi,Pb):2223 superconducting samples were obtained by using solid state reaction method. The effect of partial substitution of Ca by R=Sm, La on the structural and magnetic properties of \((Bi_{1.6}Pb_{0.4})(Sr_{1.8}Ba_{0.2})(Ca_{1-x}R_x)_{2}Cu_{3}O_y\) with \(0\leq x \leq 0.05\) were investigated by XRD and ac magnetic susceptibility measurements. The intergranular critical current density function of temperature \(J_c(T)\), were obtained and agree with the SNS behavior of grain boundaries.

1. Introduction
Superconductivity of Bi:2223 system may be directly influenced for substitution in the CuO₂ plane itself, or indirectly for substitution in the Ca or Sr positions (affecting the charge transfer to or from CuO₂ planes).

The partial substitution of divalent cation Ca by trivalent cations Y or rare earth was found to induce nanodefects which decreases the hole concentration and affect the normal and superconducting properties. The macroscopic critical current density \(J_c\) is limited by the intergranular vortex pinning force at the grain boundaries (\(J_c\) is much smaller than the critical current density \(J_{ci}\) inside the grain). The addition of Sm and Yb on the bulk Bi:2212 lead to increase the critical current density with seven times higher than that of pure sample[1]. Recently was fabricated a Bi:2223 cylindrical bulk current lead (\(I_c\) higher than 670 A/cm²) for use in cryocooler –cooled superconducting magnets [2]. Studies of partial substitution of divalent cation Ca by trivalent cations Y or rare earth (Re) ions in Bi:2223 system are in limited number in literature[3-5].

In this paper we report the effect of the partial substitution for Ca by R=La and Sm up to 5% on the purity of samples and superconducting properties of \((Bi_{1.6}Pb_{0.4})(Sr_{1.8}Ba_{0.2})(Ca_{1-x}R_x)_{2}Cu_{3}O_y\) by measurements of the AC magnetic susceptibilities function of temperature.

2. Experimental
Polycrystalline samples with nominal composition \((Bi_{1.6}Pb_{0.4})(Sr_{1.8}Ba_{0.2})(Ca_{1-x}R_x)_{2}Cu_{3}O_y\) with R=La, Sm and \(0\leq x \leq 0.05\) were prepared by the conventional solid state reaction of appropriate amounts of the metal oxides and carbonates of 99.99% purity [6].

The partial substitution of Sr by Ba was used to induce the reduction of the modulation period. Appropriate amounts of Bi₂O₃, PbO, SrCO₃, BaO, Ca CO₃, La₂O₃, Sm₂O₃ and CuO were mixed in agate mortar and calcinated at 800°C for 36 hours. The calcinated powder was pressed into pellets and presintered at 845°C for 200 hours and finally sintered at 850°C for 150 hours.
The influence of partial substitution of Ca by La, Sm on the structural and magnetic properties of (Bi,Pb):2223 bulk superconducting compound were investigated by XRD and ac magnetic susceptibility measurements. The phase purity was determined by Brucker X-ray diffractometer with Cu-Kα radiation.

The real ($\chi'$) and imaginary ($\chi''$) parts of the a.c. susceptibility were simultaneously collected with a Lake Shore Model 7000 a.c. susceptometer in the temperature range from 77K to 110K, by using frequencies $f$ and a.c. field amplitudes $H_{ac}$ situated in the ranges from 20 Hz to 1000Hz and from 4 A/m to 800 A/m, respectively.

The temperature $T_p$ for the imaginary part $\chi''(T)$ of the complex magnetic susceptibility performed at various field amplitudes is different influenced by the concentration of 4f element.

### 3. Results and discussion

Fig.1 shows the X-ray diffraction pattern for $x = 0.00 \ x=0.02$ and $x=0.05$ La and Sm samples. The results have shown that the sample with $x=0.00$ has a single (2223) phase.

![Figure 1. Powder XRD pattern for (Bi$_{1.6}$Pb$_{0.4}$)(Sr$_{1.8}$Ba$_{0.2}$)(Ca$_{1-x}$R$_x$)$_2$Cu$_3$O$_y$ with R=La, Sm and 0≤ x ≤0.05, with each peak labeled according to the crystallographic direction that produced it for 2223 phase. Stars marked 2212 phase.](image)

With increasing $x$ up to $x=0.05$ La, peaks corresponding to (2212) phase have increased in number and intensities. In the samples with $x≥0.02$ Sm the amount of (2212) phase increase from 10%vol. for $x=0.02$ to 80%vol. for $x=0.05$ (Table 1). XRD shows that only (2212) phase were found by increasing $x$ up to 0.05 [7,8].

| Sample   | $T_c$ [K] | Phase content %vol |
|----------|-----------|---------------------|
|          |           | 2223 | 2212          |
| $x=0.00$ | 109.0     | 98   | traces-       |
| $x=0.02$La | 107.5  | 95   | 5             |
| $x=0.05$La | 106    | 90   | 10            |
| $x=0.02$Sm  | 106.5  | 80   | 20            |
| $x=0.05$Sm  | 103.5  | 20   | 80            |

Table 1. The phase content of the crystalline in studied samples.
Figure 2 shows the real $\chi'$(T) and imaginary $\chi''$(T) susceptibilities for $x=0.00;0.01;0.02$ and $0.05$ La and Sm samples, by using for a. c. field an amplitude $H_{ac}=100$ A/m and a frequency $f=1000$ Hz.

With increasing temperature, the real part $\chi'$(T) shows a two step behavior for La and Sm substitute, characterizing the flux penetration in the intergranular matrix and in the grains respectively. The end of the upper step (the end of the superconductor diamagnetism) corresponds to the intragrain critical temperature. With increasing La and Sm concentration the intergrain drops shift to lower temperatures.

$\chi''$(T) exhibit a single peak at temperature $T_p$ indicating a maximum hysteresis losses due to the motion of the intergranular (Josephson) vortices. With increasing the alternative field amplitude $H_{ac}$, the $\chi''$ signal shifts to lower temperatures. This result shows that with increasing the ac field amplitude the intergranular coupling decrease. The value of temperature $T_p$ for maximum of $\chi''$ is influenced different by the nature of 4f ions. The increase of $T_p$ with increasing $x$ up to $x=0.01$ Sm, suggest the increase of intergranular coupling.

To estimate the intergranular critical current density $J_{cJ}$ from the $\chi''$(T) data we used the magnetic formulas for isotropic superconductors of cylindrical geometry in the Bean critical state model [9].

Figure 3 shows the calculated intergrain critical current densities $J_{cJ}$ function of temperature for La and Sm samples.

Figure 3. The intergrain critical current density function of temperature for for $x=0.00;0.01;0.02$ and $0.05$ La and Sm samples
Best fit of the data from Fig.3 is described by:

\[ J_{cl}(T) = J_{cl}(0) \left( 1 - T/T_{p0} \right)^2 \]  

The fitting parameters \( J_{cl}(0) \) and \( T_{p0} \) are presented in Table 2.

| Sample | \( J_{cl}(0) \) [A/cm\(^2\)] | \( T_{p0} \) [K] | \( J_{cl}(77K) \) [A/cm\(^2\)] |
|--------|-------------------------------|----------------|-----------------|
| x=0.00 | 7748                          | 104            | 522             |
| x=0.01 La | 8168                        | 106            | 613             |
| x=0.02 La | 6512                        | 105            | 463             |
| x=0.05 La | 3243                        | 104            | 220             |
| x=0.01 Sm | 7900                        | 105.5          | 590             |
| x=0.02 Sm | 3336                        | 105            | 243             |
| x=0.05 Sm | 11856                       | 85.5           | 118             |

Table 2. The fitting parameters for intergranular critical current density \( J_{cl} \).

At lower temperatures the superconducting grains will be weakly coupled by the proximity effect superconductor-normal–superconductor (SNS) junctions or by Josephson effect in superconductor-insulator–superconductor (SIS) junctions. The dependence for \( J_{cl} \) near \( T_c \) described by Eq.1 is expected for superconductor-normal–superconductor (SNS) junctions [10,11].

The critical current density \( J_{cl}(x) \) obtained from \( \chi^\prime (T) \) data shows a maximum value for \( x=x_c \) near 0.01, and suggest that a critical low concentration \( x_c \) of 4f element create nanopinning centers at grain boundaries of monophasic Bi:2223 samples, which lead to a maximum value of intergranular critical current density. The \( J_{cl}(0)=8168 \) A/cm\(^2\) in 0.01 La sample is larger that \( J_{cl}(0)=7748\)A/cm\(^2\) in 0.00 sample. The value for the maximum current density at 77K, \( J_{cl}(T=77K) \), is influenced by the nature of ion which substitute Ca. The temperature dependence of critical current density, \( J_{cl}(T) \), agree with the SNS behaviour of grain boundaries.

The effect of partial substitution of Ca by \( x=0.02 \) La, lead to a strong decrease of \( J_{cl}(0) \). This result and the calculated values for \( J_{cl} \) (77K) (Table 2) suggest that the intergranular pinning energy is influenced by the partial substitution of Ca by \( x=0.02 \) La.

4. Conclusions

The influence of partial substitution of Ca by La and Sm on the structural and magnetic properties of (Bi,Pb):2223 bulk superconducting compound were studied.

X-ray diffraction measurements shown that the substitution of Ca by La and Sm influenced different the volume fraction of 2223 phase.

The temperature \( T_p \) for the imaginary part \( \chi^\prime \) of the complex magnetic susceptibility performed at various field amplitudes is different influenced by the concentration of 4f element.

The critical current density \( J_{cl}(x) \) obtained from \( \chi^\prime\prime(T) \) data shows a maximum value for \( x=x_c \) near 0.01 for La and Sm substituent and suggest that a critical low concentration \( x_c \) of 4f element create nanopinning centers at grain boundaries of monophasic Bi:2223 samples, which lead to a maximum value of intergranular critical current density. The value for the maximum current density at 77K, \( J_{cl}(T=77K) \), is influenced by the nature of ion which substitute Ca. The temperature dependence of critical current density, \( J_{cl}(T) \), agree with the SNS behaviour of grain boundaries.

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