Effect of partial substitution of Ca by La on intergranular processes in (Bi,Pb):2223 superconductors[♣]

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The influence of partial substitution of Ca by La on (Bi,Pb)2223 phase purity and intergranular dissipative processes is characterized by XRD and a.c.magnetic susceptibility function of temperature and a.c. field amplitude. The intergranular phase diagram Hp function of temperature and the intragrain contributions of effective volume fraction of the grains f_g were estimed.

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1. Introduction

Intense studies were made in order to improve the superconducting properties of Bi-based superconductors with general formula Bi_2Sr_2Can-1Cu nOy, where n = 1, 2, and 3. This system holds some advantage, because its oxygen stoichiometry is relatively invariant with respect to cationic dopings when the samples are prepared in identical conditions [1,2].

In Bi:2212 compound, the substitution of rare earth elements in place of Ca are of much interest because it leads to structural stability and helps in understanding the nature of charge carriers as well as the effect of variation of carrier concentration of the system [3-6].

AC susceptibility measurements is a useful tool to investigate the magnetic and superconducting

Properties and for making the distinction between intergrain and intragrain properties. Two loss peaks are usually present in the imaginary part of susceptibility data; a broad peak at low temperature due to the motion of intergranular (Josephson) vortices [7] and a narrower peak due to the motion of intragranular (Abrikosov) vortices inside the superconducting grains near T onset [8]. These two peaks depend on the sample processing variables as well as the samples composition for the Bi-2223 system [9,10]

The increase of J_{transc} in the samples by increasing cooling rates may be caused by the increase of grain sizes and by improved coupling between superconducting grains [11].

In this article we report on the effects of partial substitution of Ca by Sm in Bi-2223 superconductor on AC losses as well as the field and temperature dependence of the intergranular critical current density. The effect of Sm concentration on the intergrain properties of Bi-2223 superconductors was studied.

2. Experimental

Polycrystalline samples with nominal composition $(Bi_{1.6}\,Pb_{0.4})(Sr_{1.8}\,Ba_{0.2})(Ca_{1-x}La_x)_2Cu_3$ Oy with $0\leq x \leq 0.05$ were prepared by the conventional solid -state reaction. Appropriate amounts of Bi_2O_3, PbO, $SrCO_3$, BaO, CaCO3, La2O3 and CuO were mixed in agate mortar and calcined at 800°C for 36 hours. The calcinated powder was pressed into pellets and sintered at 845°C for 200 hours. The pellets were grinding, pressed and resintered for 60 hours at 850°C. Slabs tickness 2d=3mm were cut from the sintered samples and used for a.c. susceptibility measurements. The phase purity was determined by Brucker X-ray diffractometer with Cu-Kα radiation. The results have shown that the sample with x≤0.01 has a single (2223) phase. With increasing x up to x=0.02, peaks corresponding to (2212) phase have increased in number and intensities.

The real (χ') and imaginary (χ") 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 frecquencies f and a.c. field amplitudes Hac situated in the ranges from 20 Hz to 1000Hz and from 20 A/m to 800 A/m respectively.

3. Results and discussion

In Fig. 1, we display the AC susceptibility versus temperature graph for x=0.00, 0.°2 and 0.05 La samples at Hac=100A/m field amplitudes. It is evident that La substitution has an effect on both AC loss peaks and shielding behavior of the superconducting BSCCO samples.

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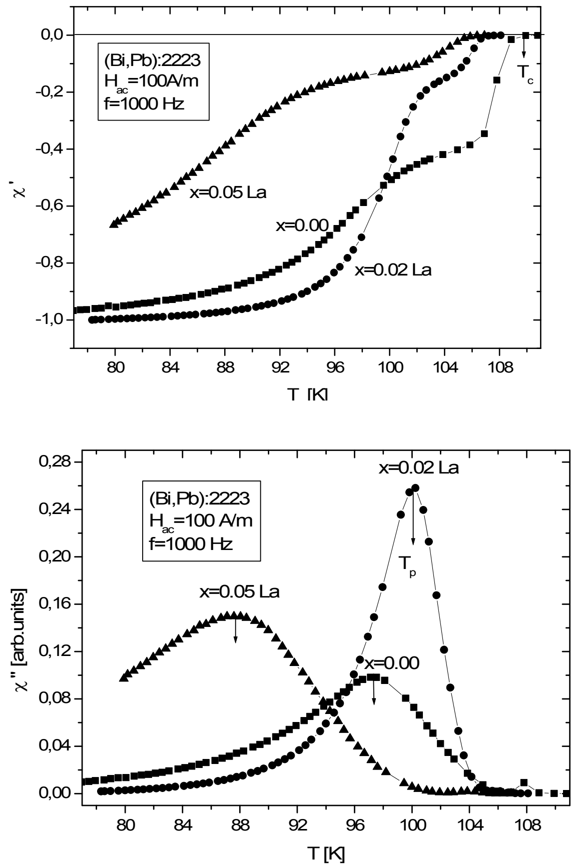

Fig. 1. AC susceptibility real (χ') and imaginary (χ'') parts as function of temperature at Hac = 100 A/m and f = 1000 Hz for samples with x = 0.00,0.02 and 0.05 La..

When the samples are at just below Tc, the superconducting grains first shield the applied magnetic field. This is measured as a negative χ'.The diamagnetic onset temperature, T_c , decreaseas from 110K to 107 K by increasing La concentration from x= 0.00 to x=0.02. This temperature remains constant at different values of AC field amplitude. As shown figure 1,the real part of AC susceptibility, χ' versus T shows for all samples two step process which reflects the flux penetration into and between the grains. At low enough temperature, intergranular component of χ' appears. At low temperatures, the entire volume of the sample is expected to be shielded by the supercurrent circulating around the sample along the grains and through the matrix and hence the When the samples are at just below Tc, the superconducting grains first shield the applied magnetic field. This is measured as a negative χ'(T)curve saturates.

In Fig. 1, χ''(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 amplitude Hac of alternative field, the χ'' signal shifts to lower temperatures and broadens. This result show that the intergranular coupling decrease with increasing the ac field amplitude. The value of temperature T_p for maximum of χ'' is influenced different by the concentration x of La. By increasing x from x=0.00 up to x=0.02 La, the peak at temperature Tp increases. The increase of x above x=0.02 lead to the decrease of Tp. The amount of the shift as a function of the field amplitude is proportional to the magnitude or strength of the pinning force. The width of each peak is manifestation of temperature exponent p of the intergranular critical current density in the expression:

$$\mathbf{J_{cJ}} = \frac{\alpha_0}{\mathbf{B^n}}\left(\mathbf{1}-\frac{\mathbf{T}}{\mathbf{T_c}}\right)^{\mathbf{p}}$$

where α0 is the pinning strength parameter at T = 0, T_c is the transition temperature for matrix and Bn the field dependence of the intergranular (matrix) current density [12].

Fig. 2 shows the semi-log plot of the AC loss peak temperatures, T_p, as a function of various AC field amplitudes, for our studied samples. The peaks shift to the higher temperature by increasing La concentration up to x=0.02 resulting in a increase of intergranular critical current density. By increasing x to 0.05 the peak shift to lower temperature, suggesting the decrease of intergranular pinning force.

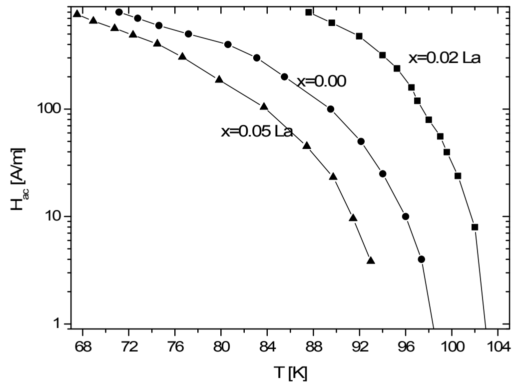

Fig. 2. The variation of Hac as a function of peak temperature.

To separate the intra- and intergrain contribution, one has to know the effective volume fraction of the grains fg. Method of calculations for fg is reported by several workers In reference [13] the experimental AC susceptibility data χ''(T) were plotted versus χ'(T) and the value of fg is obtained.. Applying the same procedure in Fig. 3, we obtained the values of f_g =0.39; 0.15 and 0.12 for x=0.00; 0.02 and 0.05 La , respectively.

It should be noted that partial substitution of Ca by La change the values of f_g.

We can attribute the variation in the parameter fg to the multiphase and granular structure of these ceramic superconductors, and the increase of Tp for x=0.02 to the reduction of modulation structure by La and the increase of intergranular coupling.

The study of La influence on intergranular dissipation were studied from electrical resistivity function of temperature measurements.

Fig. 4 show that above the excess conductivity region, in the 200K-300K temperature range, our samples are characterised by a linear temperature dependence of the electrical resistivity:

$$\rho = \rho_0 + \alpha * T$$

where ρ0 is the residual resistivity and α is the slope of resistivity in the normal state.

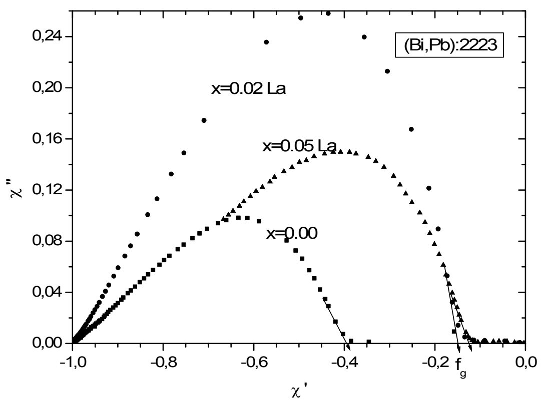

Fig. 3. χ'' versus χ' for x=0.00;0.02 and 0.05 La. The value of fg is obtained by the linear extrapolation of χ'' versus χ' dependence.

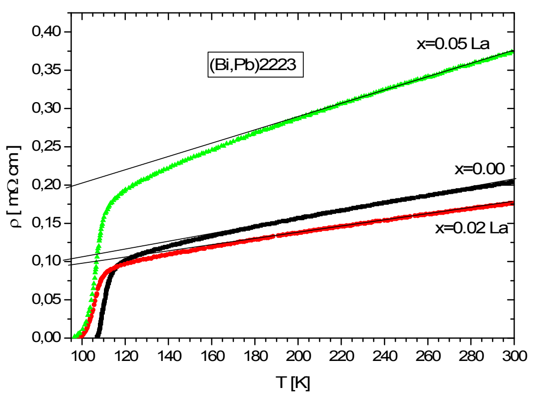

Fig. 4. Electrical resistivity versus temperature for x=0.00;0.02 and 0.05 La doped samples.The straight lines shows the extrapolation for linear dependence of ρ(T).

As shown in Table1, ρ_0 and α values are sensitive function of x. A partial substitution of Ca by x=0.02La decreases residual resistivity ρ_0 , which suggest that the number of scattering centers in the intra-and intergrain regions decrease.This result is in agreement by the AC magnetic measurements (the decrease of T_p) , which suggest the increase of intergranular pinning force.For x=0.05 La, residual resistivity and α increases, in agreement with the decrease of inetergranular coupling obtained from AC data.The granular microstructure and the nature of contacts between the grains strongly influenced the residual resistivity. Structural defects and inhomogenities force the current to meander through the cuprate enhancing the normal resistivity. The measured normal resistivity in bulk samples is related to the intrinsic resistivity in CuO_2 (a-b) plane of a single crystal ,by the relation [14,15]:

$$\rho = p(\rho\, ab + \rho\, ct).$$

The coefficient p account for a mean percolative lengthening of the conduction paths and for the mean shrinking of the current cross-section, ρ ab is the in plane resistivity for a single crystal and ρct is associated with the contact resistance between the grains. The in plane resistivity for a single crystal is: ρ_{ab} = ρ0i + αi*T , where ρ 0i is the intrinsic residual resistivity and αi = dρab/dT is the intrinsic slope of linear dependence of $\rho_{ab}(T)$.

The coefficient p(x) was extracted for each sample by assuming that αi = dρab/dT in x=0.00 single crystal is weakly affected by a small concentration of La. We obtain p(x) = α(x)/ αi , therefore the increase of parameter p for x=0.05 sample and the decrease for x=0.02 sample [Table 1]. For Bi:2223 single crystal the intrinsic value is around αi = 0.5 μΩ cm K-1 [12].

Table 1 shows other parameters sensitive to Ca substitution for La as: the midpoint critical transition temperature (T_c), the transition width and the temperature for zero resistivity ($T_c(\rho=0)$), respectively.

Table 1. Parameters of undoped and La doped samples obtained from resistivity and XRD data.

Sample	Tc [K]	ΔTc [K]	Tc(ρ=0) [K]	ρ(0) [μΩ.cm]	α=dρ/dT [μΩ.cm/K]	Phase content %vol		
						2223	2212	2201
x=0.00	109.5	4	107.0	71	4.8	98	traces-	--
x=0.02 La	107,5	5	107	67	4.1	98	urme	-
x=0.05 La	106.5	5	100	114	8.7	92	8	

The dependence of Tc(ρ=0) function of x may be in relation by phase content and intergrain dissipation processes.

4. Conclusions

The partial substitution of Ca by La (x=0.00;0.02;0.05) in $(Bi_{1.92}Pb_{0.44})Sr_2$ $(Ca_{1-x}La_x)2Cu_3$.Oy bulk superconductor was performed.

By increasing x up to x=0.02, peaks corresponding to (2212) phase have increased in number and intensities.

The intergranular pinning force is different influenced by the partial substitution of Ca by La in (Bi,Pb):2223 samples.

χ' versus T shows for all samples two step process which reflects the flux penetration into and between the grains.

χ"(T) exhibit a single peak at temperature Tp indicating a maximum hysteressis losses due to the motion of the intergranular (Josephson) vortices.

Semi-log plot of the AC loss peak temperatures, Tp, as a function of various AC field amplitudes, for our studied samples, shows that La influence different the intergrain coupling, (which increase for La concentration up to x=0.02 and decrease for x=0.05).

The partial substitution of Ca by La decreases the effective volume fraction of the grains, f_g.

In normal region all samples show a linear temperature dependence of electrical resistivity function of temperature. The decrease of residual resistivity in sample x=0.02 La agree with the increase of intergranular critical current density.

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