JOURNAL OF MATERIALS SCIENCE **34** (1999) 2813–2819

Effect of Ba and Zn doping in $Bi_2Pb_{0.6}Sr_2Ca_2Cu_3O_\delta$ superconductors using ac susceptibility measurements

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Ac complex susceptibility, $\chi = \chi' - i\chi''$, measurements were done on the samples doped with barium and zinc ($Bi_2Pb_{0.6}Sr_2Ca_{2-x}M_xCu_3O_\delta$, M = Ba, Zn and $x = 0.02$ and 0.10). The data of χ' shows that coupling of the grains in Zn-doped samples are weaker than that of Ba-doped samples and hence it could be concluded that Zn-doped samples are dominated by the S-I-S type of weak links, whereas the Ba doped samples are dominated by the S-N-S weak links. Calculated values of I_0 is three times higher in the Ba doped samples such that the values of Josephson coupling energy, E_j is four times that of Zn doped samples. Analysis based on the sensitivity of the data of $d\chi'(T)/dT$ versus temperature furnished further information on the two-step transitions related to the coupling of the grains in both systems. © *1999 Kluwer Academic Publishers*

1. Introduction

Ac susceptibility technique is now commonly used in the characterization of superconducting materials [1–5]. This technique offers additional advantage because it avoids the need for contacts as in the case of resistivity measurements. It also avoids ambiguity in the determination of the onset temperature of diamagnetism even for a small volume of sample. Intrinsic and extrinsic properties related to the nature of grains, grain boundaries, couplings and phases in connection to superconducting properties have been studied extensively using ac susceptibility measurements. There are numerous reports related to the properties of superconductors, such as magnetic relaxation [6], vortex dynamics, flux distribution and creeps [7], penetration depth [8], transport current densities [9], magnetic reversibility [10] and granularity [11] based on ac susceptibility measurements.

In this paper, ac susceptibility measurements were performed on Ba and Zn doped $Bi_2Pb_{0.6}Sr_2Ca_{2-x}M_xCu_3O_\delta$, (M = Ba, Zn and $x = 0.02$ and 0.10) superconducting system, as a function of temperature and ac fields and its effect on the coupling of the grains were discussed.

2. Experiment

The ceramic samples of $Bi_2Pb_{0.6}Sr_2Ca_{2-x}M_xCu_3O_\delta$, M = Ba, Zn and $x = 0.02$ and 0.10 were prepared from appropriate mixtures of high purity (99.9%) Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$, $BaCO_3$, ZnO and CuO powders. The powders were ball-milled for 24 hours. The mixture was first calcined at 800 °C for 24 hours and was later presintered at 830 °C for 14 hours with intermediate grindings in order to ensure good homogeneity. Then the powders were pressed into pellets and sintered at 855 °C for 150 hours. The samples prepared were identified as sample 1 (for Ba doped $x = 0.02$); sample 2 (for Ba doped $x = 0.10$); sample 3 (for Zn doped $x = 0.02$) and sample 4 (for Zn doped $x = 0.10$).

Ac susceptibility measurements were performed, using a LakeShore Model 7000, to study the flux penetration as samples were heated from 20 to 115 K at 125 Hz with the driving field ranging from 0.1 Oe to 10 Oe. Resistance measurements, using a fourpoint probe method at a constant current of 20 mA, over the range of temperature from 20 to 300 K were also carried out.

3. Results and discussion

The temperature variation of the complex magnetic susceptibility, $\chi = \chi' - i\chi''$, in various ac fields, H, for all samples are shown in Figs 1–4. The curves of the real component, $\chi'(T)$ which provide information on intrinsic and coupling diamagnetic shielding; and the imaginary component, $\chi''(T)$ that displays the features of coupling losses which are common behaviour of sintered ceramics superconductors were observed. However, the peaks associated with the intragranular loss near T_c were not observed for all samples. Unlike the intragranular peaks, the coupling peaks were observed because the intergranular critical current density, J_{cj} is very sensitive to the presence of magnetic field [12]. The onset of diamagnetism for Ba and Zn-doped samples were observed at 107 and 106 K, respectively. All samples show two step transition in χ' as temperature

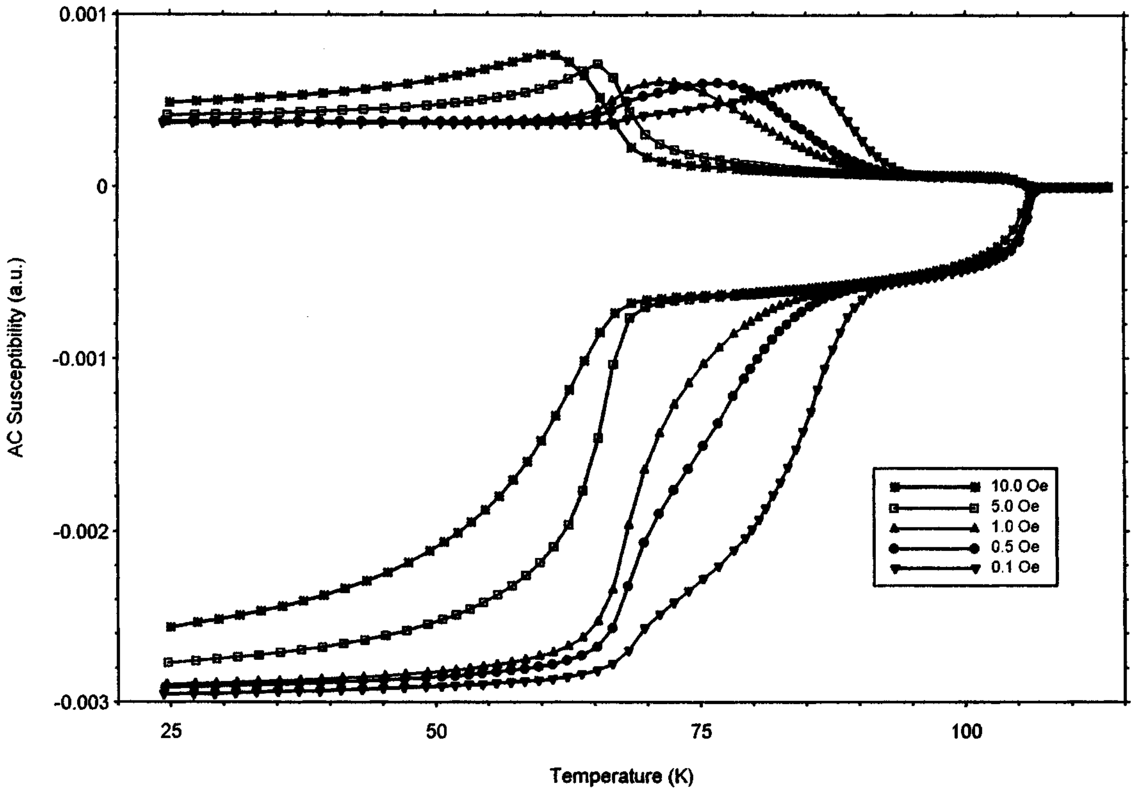

Figure 1 AC susceptibility of $Bi_2Pb_{0.6}Sr_2Ca_{2-x}Ba_xCu_3O_\delta$ sample ($x = 0.02$).

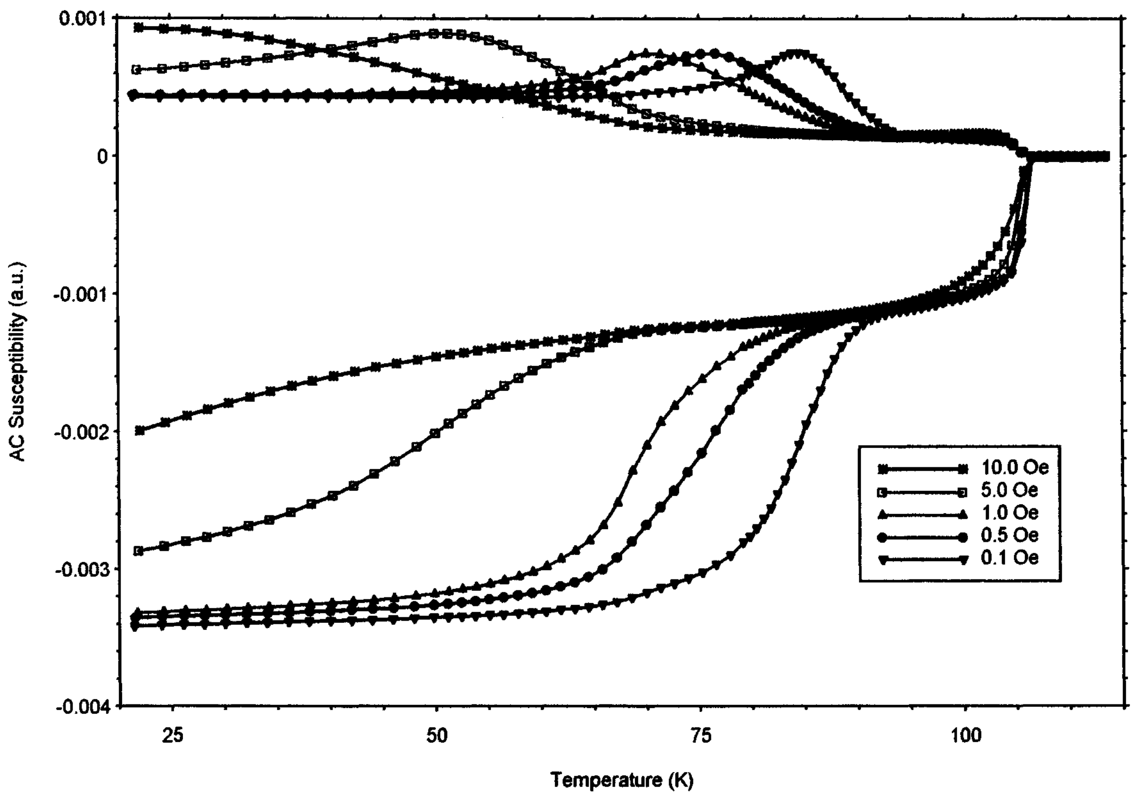

Figure 2 AC susceptibility of $Bi_2Pb_{0.6}Sr_2Ca_{2-x}Ba_xCu_3O_\delta$ sample ($x = 0.10$).

decreases [13, 14]. This feature is due to presence of both low and high T_c phases where the high T_c phases is more dominant in Ba doped samples than in Zn doped samples. Hence Zn doping is less favourable for the formation of 2223 phase than Ba doping. This observation (two step transition) is also consistent with the resistance measurements shown in Fig. 5. Sample 4 shows a T_c ($R = 0$) of 75 K, while for sample 2 T_c ($R = 0$) is 88 K.

The behaviour of flux penetration in the samples shown in the χ'-T curves (Figs 1–4) is field dependent. For samples 1 and 2, at lower applied fields of 0.1, 0.5 and 1 Oe. flux penetration is minimum until 65 K. Beyond this temperature rapid increase of

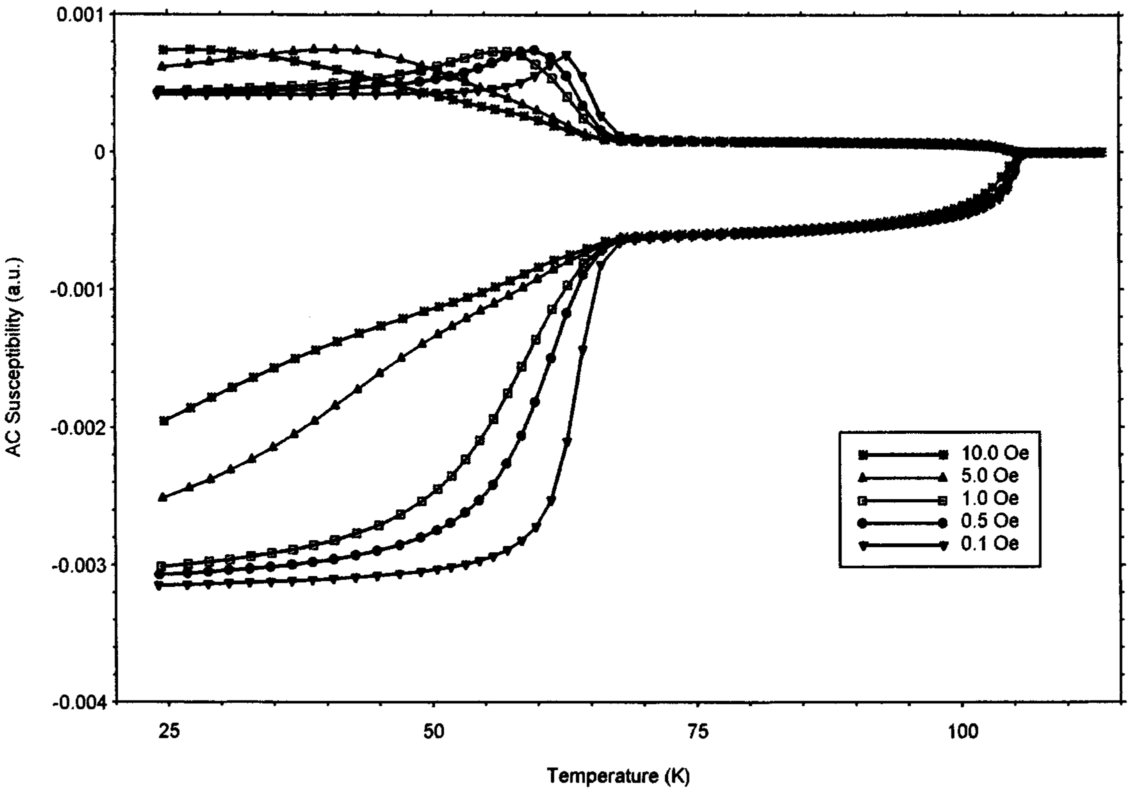

Figure 3 AC susceptibility of $Bi_2Pb_{0.6}Sr_2Ca_{2-x}Zn_xCu_3O_\delta$ sample ($x = 0.02$).

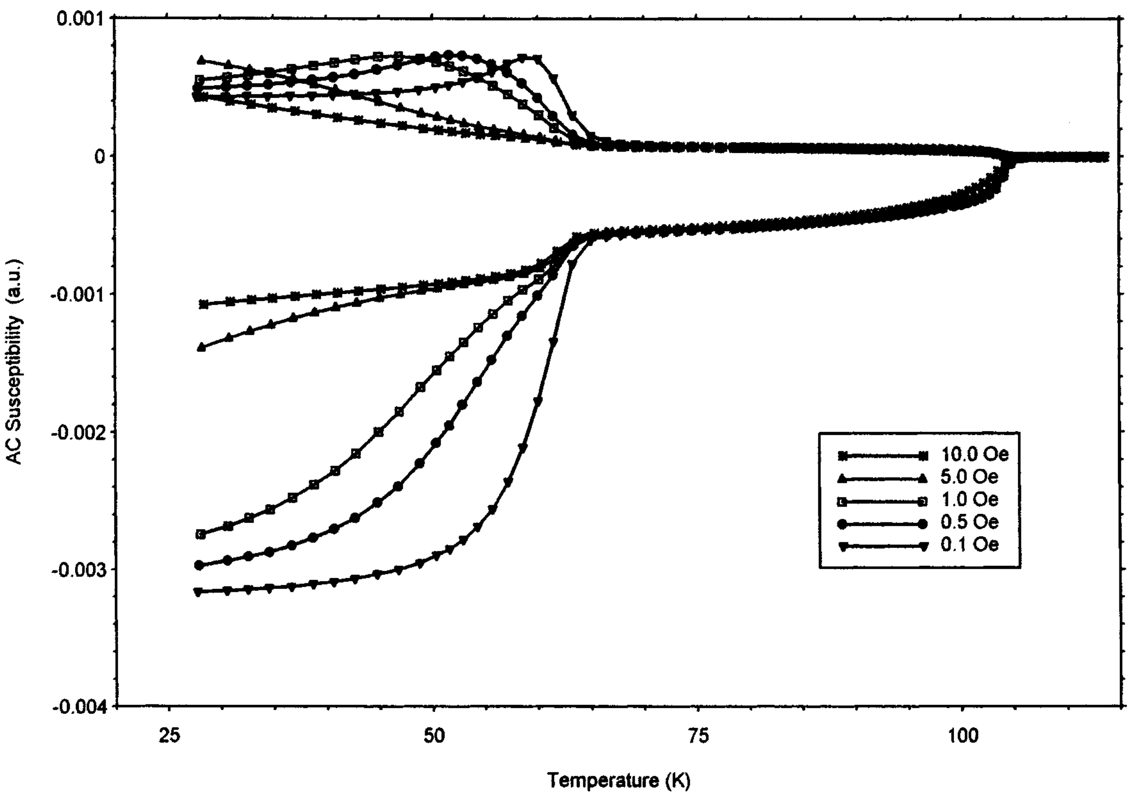

Figure 4 AC susceptibility of $Bi_2Pb_{0.6}Sr_2Ca_{2-x}Zn_xCu_3O_\delta$ sample ($x = 0.10$).

flux penetration were observed until 92 K. Between this temperature and the onset temperature of diamagnetism, it is field independent. For applied fields of 5 and 10 Oe. flux penetration increases exponentially for sample 1 but linearly for sample 2. This happens until the temperature is 70 K. Beyond this temperature until the onset of diamagnetism, flux penetration is field independent. However the behaviour of flux penetration in Zn doped samples differs. At lower applied fields of 0.1, 0.5 and 1 Oe, the field penetrates exponentially until 68 K for sample 3 and 65 K for sample 4. For higher fields the penetration is quite linear until 68 and 65 K for samples 3 and 4, respectively. Beyond these temperatures, flux penetration is field independent until the

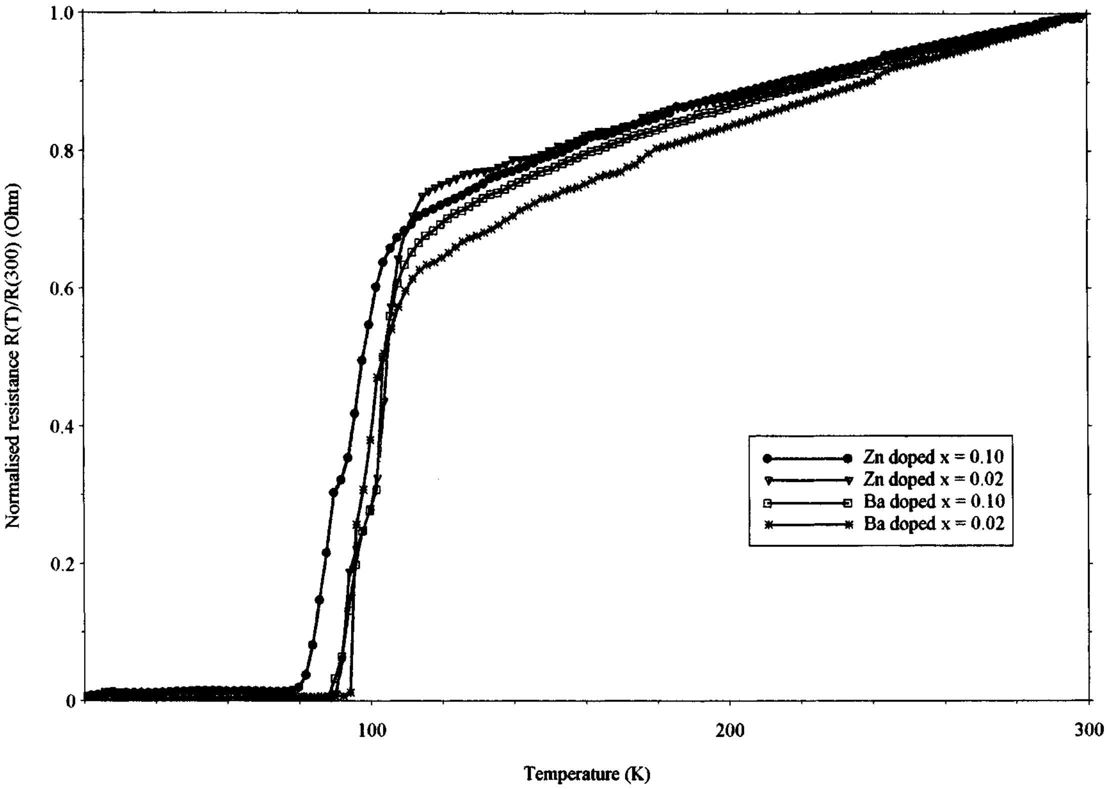

Figure 5 Normalised resistance as a function of temperature for $Bi_2Sr_2Ca_{2-x}M_xCu_3O_\delta$ (M = Ba, Zn) samples.

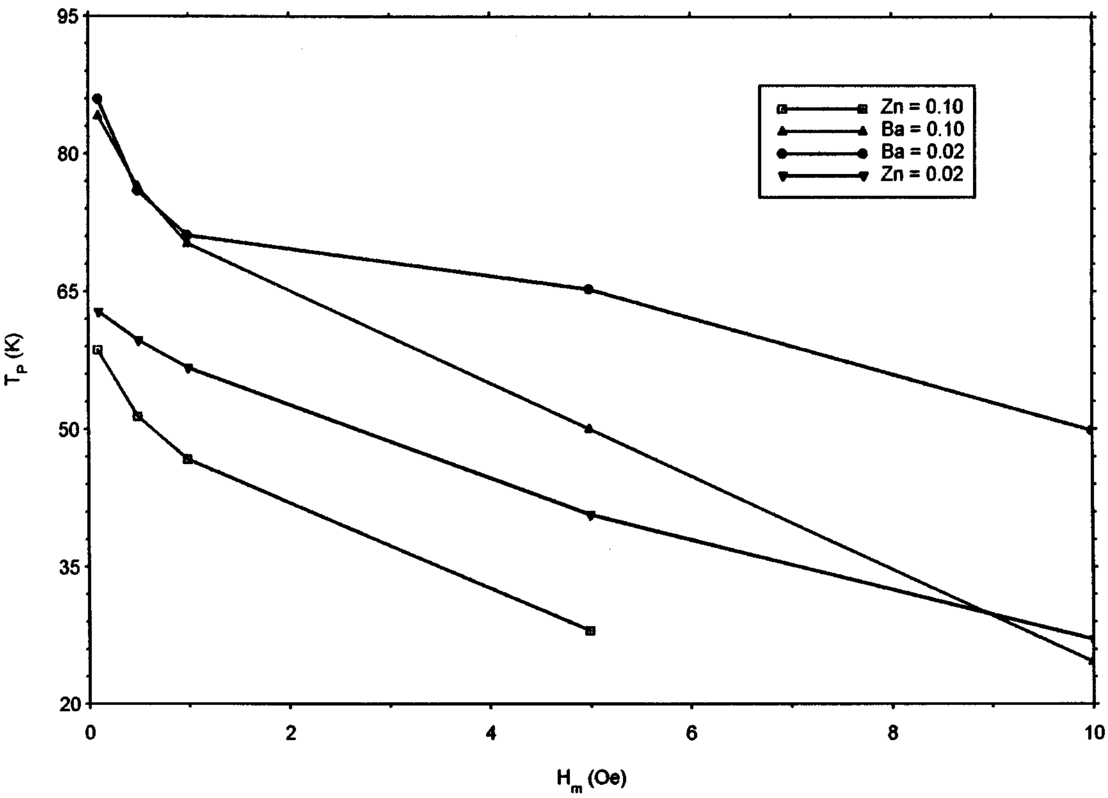

Figure 6 Applied field as a function of the coupling peak temperature.

onset temperature of diamagnetism. These differences observed between the Ba doped and the Zn doped samples might be due to the quality of the grains and the nature of the grain boundaries.

The effect of different applied fields on the quality of the grains, the nature of the grain boundaries and the phases present in the samples is also shown in the χ''-T curves. In all the samples, intrinsic peaks were not resolved even at 10 Oe. because the samples were not strongly coupled. At temperatures well below T_c broad coupling peaks observed were field dependent. Fig. 6 shows the variation of coupling peaks T_p with

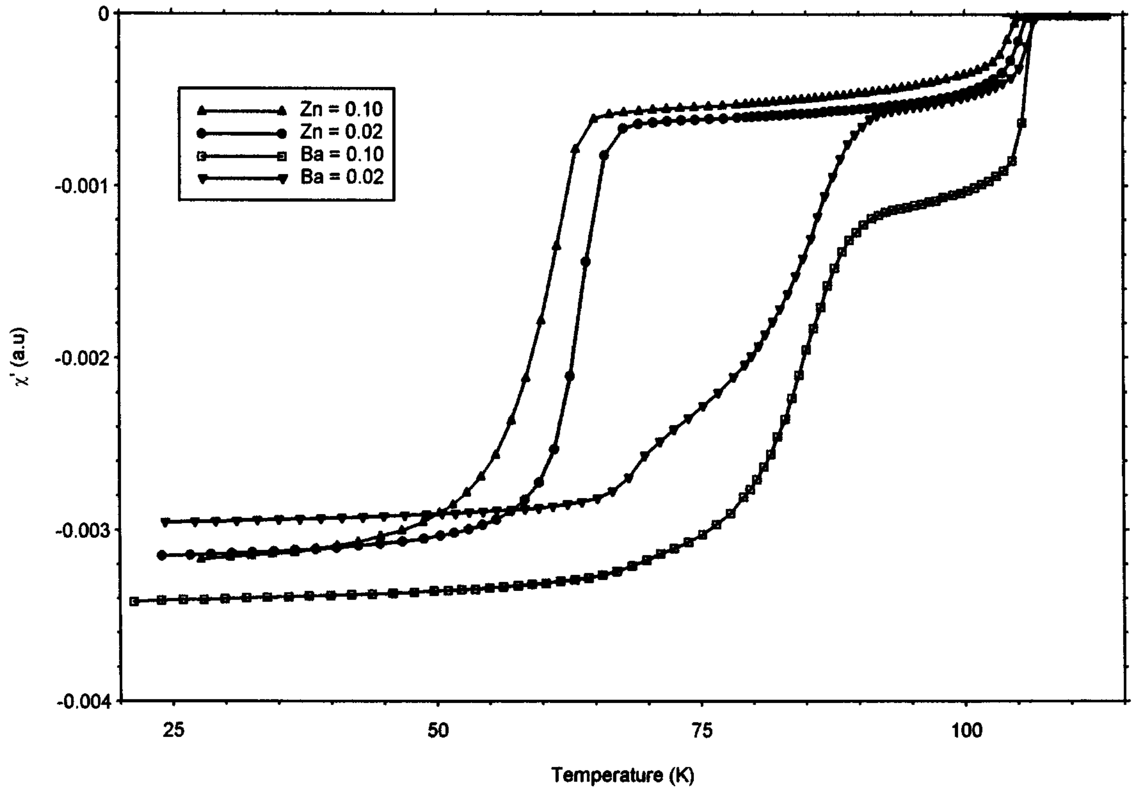

Figure 7 Real part of the AC susceptibility as a function of temperature for $Bi_2Pb_{0.6}Sr_2Ca_{2-x}M_xCu_3O_\delta$ (M = Ba, Zn).

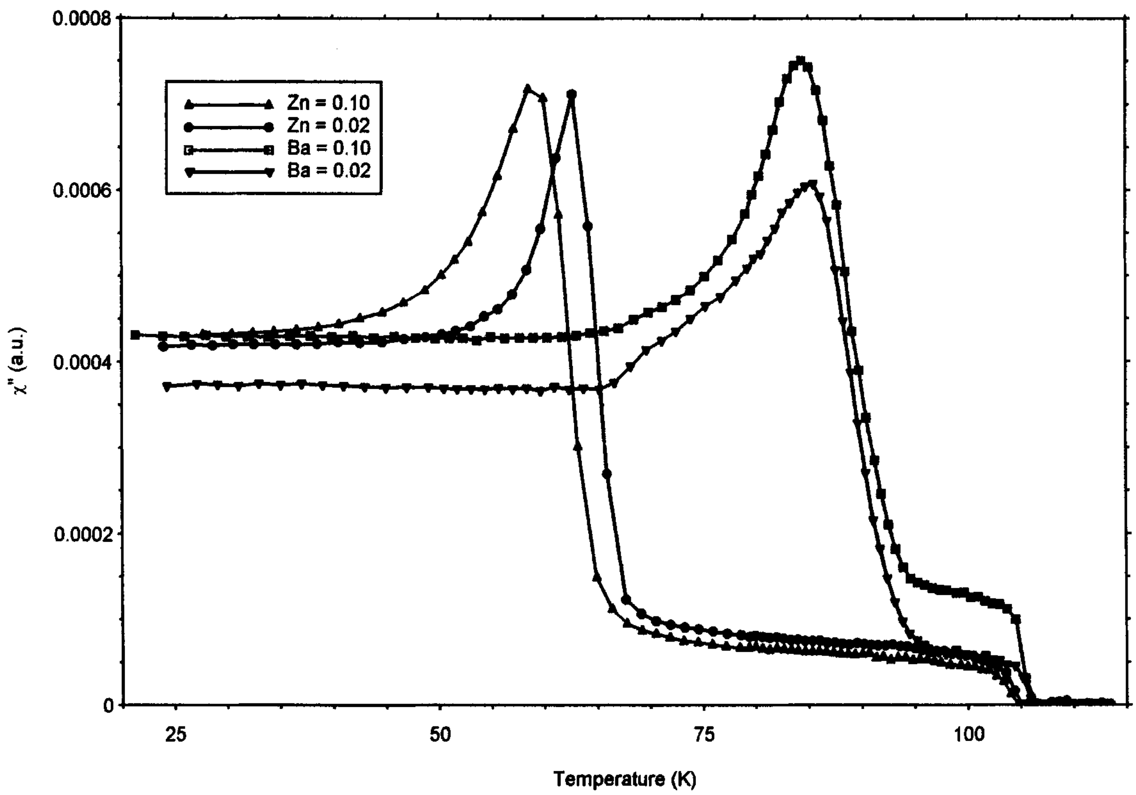

Figure 8 Imaginary part of the AC susceptibility as a function of temperature for $Bi_2Pb_{0.6}Sr_2Ca_{2-x}M_xCu_3O_\delta$ (M = Ba, Zn).

applied fields H. For samples 1 and 2, T_p occurs at almost the same temperatures, but diverge as the field increases. However for samples 3 and 4 T_p diverge at all fields. The coupling peak at 10 Oe was not observed for sample 4 and it could be at a much lower temperature.

The data of $\chi'(T)$ and $\chi''(T)$ as shown in Figs 7 and 8, respectively, can also be used to study the effect of barium and zinc doping in the system. The two-step feature of the transition curve in $\chi'(T)$ shows that Ba doping favours the formation of high T_c phase than Zn doping. For samples 1 and 2, (Ba-doped) the temperatures at the shoulder of the first transition are 94 and 93 K, while for samples 3 and 4 (Zn-doped) the temperatures are 68 and 65 K respectively, hence indicating the

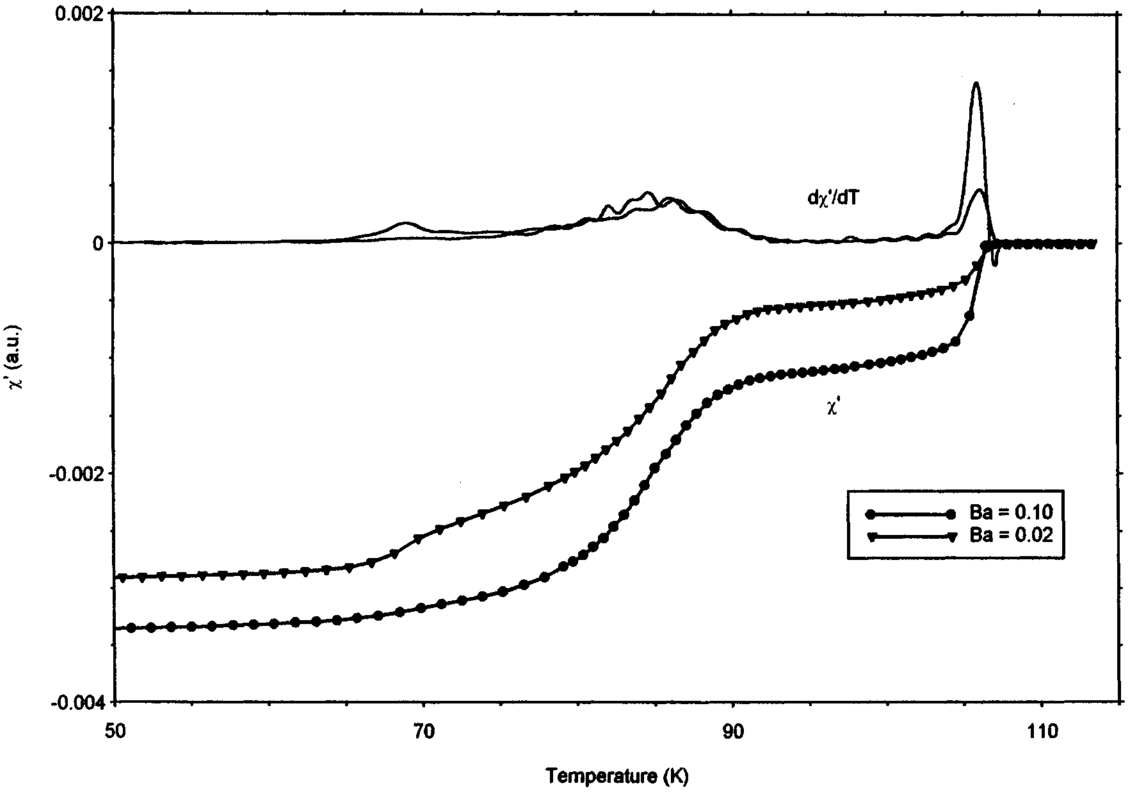

Figure 9 Differential analysis of real part χ' as a function of $Bi_2\ Pb_{0.6}Sr_2Ca_{2-x}Ba_xCu_3O_\delta$ samples.

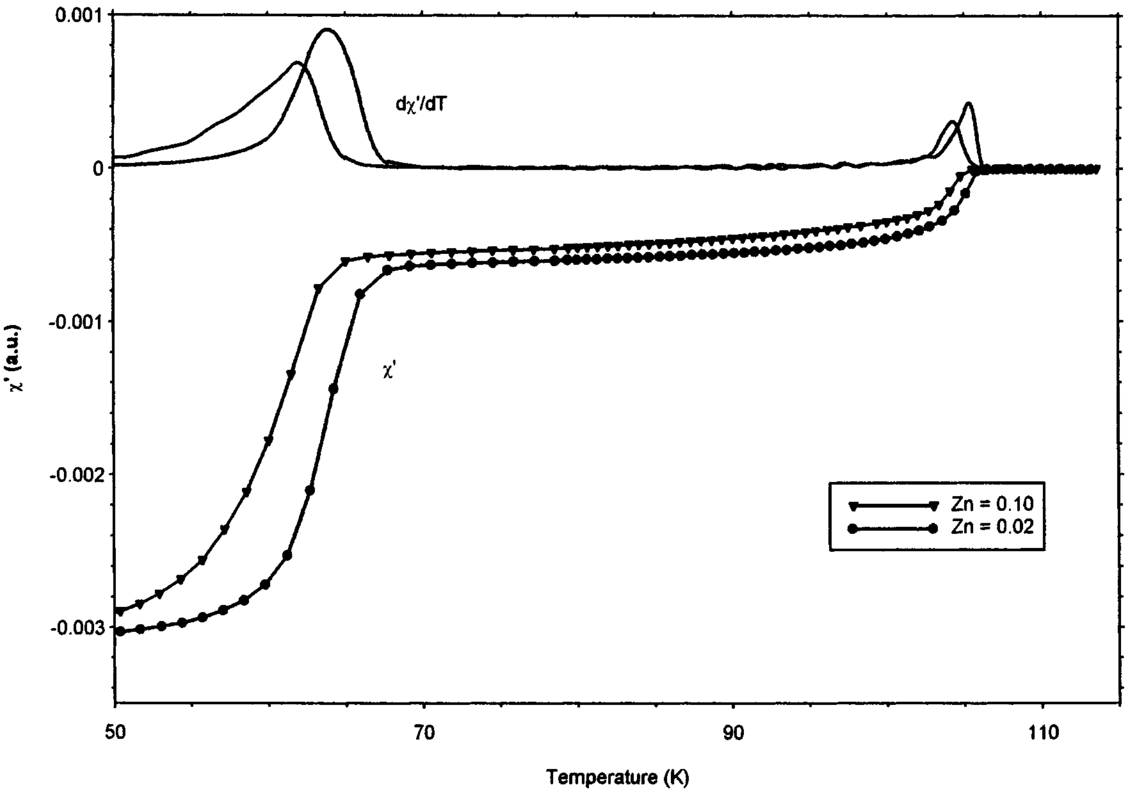

Figure 10 Differential analysis of real part χ' as a function of temperature for $Bi_2Pb_{0.6}Sr_2Ca_{2-x}Zn_xCu_3O_\epsilon$ samples.

dominance of 2223 phase in barium doped samples. Additional analysis based on the data of $d\chi'(T)/dT$ versus temperature could furnish further information on the two-step transitions. Figs 9 and 10 show $d\chi'(T)/dT$ versus temperature for barium-doped and zinc-doped samples respectively. The peaks observed, which correspond to the mid-point of the transition, in the high temperature transition for samples 1 and 2 has a difference of 0.5 K, while for samples 3 and 4 the difference is about 2 K. For the low temperature transition, the separation of the peaks between samples 1 and 2 is 1.5 and 2.5 K for samples 3 and 4. Separation of the low

TABLE I Calculated values of maximum Josephson current and coupling energy in Ba doped and Zn doped samples

Samples	Doping composition	I_0 (Maximum Josephson current) μA	E_j (Josephson coupling energy) $\times 10^{-21}$ J
Ba doped	$X = 0.02$	13.6	4.481
	$X = 0.10$	12.6	4.152
Zn doped	$X = 0.02$	4.44	1.463
	$X = 0.10$	4.29	1.414

and high peaks for sample 1 at 0.1 Oe. is 20.3 K, while for sample 3 the separation is 42 K.

Remarkable differences between both type of doping were also observed in the imaginary component, $\chi''(T)$. Peaks corresponding to maximum hysteresis losses at the grain boundaries, when supercurrents and penetrated flux reach the center of the samples, were seen at much lower temperatures for zinc doped samples as compared to barium doped samples. The shifting of the peaks between samples 1 and 2 is 1.5 and 5 K for samples 3 and 4. Hence it can be concluded that the nature and degree of the weak links, due to Josephson junction coupling, in both types of samples are different. For sample with 2223 phase dominancy, the weak links is probably of the S-N-S type, whereas in the low T_c (2212) phase dominated sample, S-I-S junctions seem to dominate [15]. Thus, in Zn-doped samples the S-I-S type of weak links dominated, while the S-N-S weak links seem to dominate in the Ba doped samples.

Clem [16] assumed that bulk samples of high T_c superconducting ceramic are consisted of superconducting grains separated by layers of non-stoichiometric interface materials and the coupling of the grains occurs through the Josephson currents in the layers. The Josephson coupling energy is given by

$$E_j = (h/4\pi e)I_0 \tag{1}$$

where I_0 is the maximum supercurrent through the junction. According to Ambegaokar-Baratoff theory [17], $I_0(0)$ is given by

$$I_0 = 1.57 \times 10^{-8} T_c^2/(T_c - T_{cj}) \tag{2}$$

where T_{cj} is the phase locking temperature (onset of decoupling of the grains) associated with the lower transition temperature in $\chi'(T)$. The calculated values of I_0 and E_j, based on Equations 1 and 2, for the samples are summarized in Table I. Both values of I_0 and E_j in Ba doped samples are higher than that of Zn doped samples. This tends to agree with the assumption of the nature of weak links in the samples as described earlier.

4. Conclusion

The importance of varying the ac field amplitude has been underlined. In Zn doped samples maximum ac losses occurs at a much lower temperatures as compared to Ba doped samples due to the difference in the dominancy of the high T_c phase. The nature and degree of the weak-links, due to Josephson junction coupling of types S-N-S and S-I-S, in both types of samples are different. For sample with 2223 phase dominancy, the weak links is probably of the S-N-S type, whereas the low T_c (2212) phase dominated sample, S-I-S junctions seem to dominate. The coupling of the grains in Zn-doped samples are weaker than that of Ba-doped samples. Hence it could be concluded that Zn-doped samples are dominated by the S-I-S type of weak links, whereas the Ba doped samples are dominated by the S-N-S weak links as supported by the calculated values of I_0 and E_j in the samples. Analysis based on the sensitivity of the data of $d\chi'(T)/dT$ versus temperature furnished further information on the two-step transitions related to the coupling of the grains.

Acknowledgement

The financial support of the Ministry of Science, Technology and Environment, Malaysia under the Intensified Research In Priority Area (IRPA) vote: 4-07-05-026 (High Temperature Superconducting Materials And Device Applications) is gratefully acknowledged.

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Received 28 July and accepted 23 December 1998