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Investigation of Ni Substitution for Cu in Bi(Pb)-Sr-Ca-Cu-O Superconductor by AC Magnetic Susceptibility Measurements.

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Abstract

In this work we have investigated the effect of doping of magnetic element Ni for copper into the copper site of Bi(Pb)-Sr-Ca-Cu-O superconductor by means of AC magnetic susceptibility measurements. Three samples with composition $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ with $x=0$, $x=0.05$ and $x=0.1$ were prepared using sol-gel method and the measurements of $\chi'(T)$ and $\chi''(T)$ were carried out on these samples in zero and in applied d.c magnetic fields. The $\chi'(T)$ data for the samples in zero d.c field show single drops indicating that the samples had one superconducting phases and the superconducting transition temperatures of the samples were determined to be 108.7 K, 98.9 K and 56.7 K respectively. Obviously superconducting transition temperatures of the samples decrease with increasing Ni concentration. The $\chi'(T)$ data for the samples in d.c magnetic fields show broad superconducting transitions indicating that the magnetic fields quenched superconductivity of the samples. The $\chi''(T)$ data in d.c magnetic fields show broad intergrain peaks associated with energy dissipation inside the samples. In low d.c magnetic fields $\chi'(T)$ data for the $x=0$ and $x=0.05$ samples show two drops associated with inter and intragrain regions of the samples. In high magnetic fields $\chi'(T)$ data show single transitions indicating that the magnetic fields simultaneously penetrated into the superconducting grains and the weak links. The $\chi'(T)$ data for the $x=0.1$ sample shows single drops in low and in high d.c. magnetic fields. The high d.c magnetic fields pushed the superconducting transition temperatures of the samples to lower temperatures and the intergrain peaks in $\chi''(T)$ shifted to lower temperatures with increasing applied d.c magnetic fields.

1. INTRODUCTION

In the copper-oxide based superconductors many studies have been done on the effect on the physical properties of substitutions by 3d elements such as Fe, Co, Ni and

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Zn. It has been reported previously by A. Maeda et.al [1] that the substitutions by these 3d elements for Cu in the Bi-based superconductor results in a decrease in the superconducting transition temperatures T_c of these materials. It has also been reported that doping Ni and Zn into the Cu sites in the lattice of Bi-Pb-Sr-Ca-Cu-O system depresses T_c with the rates of decrease of T_c being the same for both kind of doping [2]. In this work we investigated the effect of Ni substitution for copper in the Bi-Pb-Sr-Ca-Cu-O system by means of ac magnetic susceptibility measurements.

The ac magnetic susceptibility measurements are most useful for characterizing superconducting samples and these measurements can be conveniently carried out using Hartshorn's mutual inductance technique [3]. The ac susceptibility can be written as a complex quantity such that

$$\chi = \chi' + i \chi''$$

where χ' and χ'' are the in-phase and out-of-phase components of the complex susceptibility. In general temperature dependence of in-phase component $\chi'(T)$ of the superconducting sample shows the perfect diamagnetism of the sample below its superconducting transition temperature which indicates that sample exhibits Meissner effect. The temperature dependence of the out-of-phase component $\chi''(T)$ reflects the energy dissipation in the superconductor during the superconducting transition. Usually each drop in $\chi'(T)$ is accompanied by a dissipative peak in $\chi''(T)$. In high T_c superconductors the complex susceptibility is most useful for studying the nature of intergranular and intragranular superconductivity. In a very small ac field a sharp drop is observed in $\chi'(T)$ showing diamagnetism of the sample. However if the ac field is relatively large then typically there will exit two superconductive components in $\chi'(T)$. The component with the higher critical temperature can be regarded as being due to the bulk superconductivity appearing in the inside of the superconducting grains. On the other hand the component with the lower critical temperature and the corresponding peak in $\chi''(T)$ are attributed to the presence of weak coupling between superconducting grains. This two stage behaviour in $\chi'(T)$ is found even more clearly in the presence of an applied dc magnetic field.

The $\chi'(T)$ also determines the onset temperature of the superconducting transition, superconducting transition width and the number of superconducting phases existing in the sample.

2. SAMPLE PREPARATION

Three samples with the composition $\text{Bi}_{1.6} \text{Pb}_{0.4} \text{Sr}_2 \text{Ca}_2 \text{Ni}_x \text{Cu}_{3-x} \text{O}_y$ with $x=0$, $x=0.05$ and $x=0.1$ were prepared. The samples were prepared using sol-gel method in which the appropriate amounts of $\text{Bi}(\text{NO}_3)_3$, $\text{Pb}(\text{NO}_3)_2$, $\text{Ca}(\text{NO}_3)_2$, $\text{Sr}(\text{NO}_3)_2$ and

$\text{Cu}(\text{NO}_3)_2$ and $\text{Ni}(\text{NO}_3)_2$ were mixed in a solution of citric acid, ethylene glycol and distilled water and the solution was heated at 180°C for one day. The resultant powder was first compressed into pellet form and then furnace at 660°C for 24 hours. It was then ground back into powder form and re-compressed into a pellet before being furnace at 825°C for 24 hours. Finally it was reground, pelletised and furnace once again at 844°C for 10 days. All these processes were carried out in air.

3. EXPERIMENTAL TECHNIQUES

The major part of the ac susceptibility apparatus is the ac Hartshorn bridge[3]consisting of a primary coil, two oppositely wound secondaries, mutual inductance box, potentiometer, ac power supply and a two phase lock-in-amplifier. The sample chamber is a long glass tube sealed to the upper plate of the cryostat. The primary and the secondary coils are wound on a tufnel rod which is sealed onto the lower end of the glass tube. The superconducting magnet is connected to the lower part of the cryostat. The sample holder is a thin non-magnetic aluminium plate which was attached to a long stainless steel tube. A sensitive Gold/Iron-chromel thermocouple which was glued on to the one side of the aluminium plate was used to measure the temperature of the sample.

The sample was mounted on one side of the aluminium plate and the stainless steel tube gently inserted into the sample chamber and sealed to the upper plate of the cryostat. The sample chamber was pumped out to a good vacuum. A small ac current was applied to the primary coil. The ac magnetic field produced by the primary coil and the frequency of the ac signal were 0.25 G(r.m.s) and 370 Hz respectively. Initially the bridge was balanced by adjusting the mutual inductance box and the potentiometer so that $\chi'(T)$ and $\chi''(T)$ are zero. Now the sample was cooled down to 4.2 K using liquid nitrogen and liquid helium.

4. THE MEASUREMENTS OF $\chi'(T)$ AND $\chi''(T)$ IN ZERO AND IN APPLIED D.C MAGNETIC FIELDS

The increasing current was applied to the heater coil by a current source and the measurements of $\chi'(T)$ and $\chi''(T)$ were recorded by the two phase lock-in-amplifier while the sample was warming. The corresponding temperatures of the sample in the range from 4.2 K to 120 K were also measured by the thermocouple and recorded by a voltmeter. In this experiment the current source, voltmeter and the lock-in-amplifier were all interfaced to a computer.

In order to carry out the measurements of $\chi'(T)$ and $\chi''(T)$ in d.c magnetic fields the sample was cooled to 4.2 K in zero d.c magnetic field. A d.c magnetic field of 5 G was applied to the sample from the superconducting magnet. Now the sample was slowly warmed and the measurements of $\chi'(T)$ and $\chi''(T)$ and the corresponding temperatures

were recorded in the presence of d.c field. These measurements were repeated for the magnetic fields 15G, 25G, 50G, 100G and 500G. These measurements of $\chi'(T)$ and $\chi''(T)$ are referred to as zero field cooled (ZFC) ac susceptibility measurements.

5. PRESENTATION OF RESULTS

5.1 The measurements of $\chi'(T)$ in zero applied d.c magnetic field

The measurements of $\chi'(T)$ in zero d.c magnetic field on the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ samples with $x=0$, $x=0.05$ and $x=0.1$ are presented in figure 1. As can be seen in the $\chi'(T)$ data for the $x=0$ sample a single sharp drop in the susceptibility was observed which indicates that the sample had only one superconducting phase which is identified as the Bi-2223 superconducting phase. The superconducting transition temperature of this sample was observed to be 108.7 K. From the $\chi'(T)$ data in zero d.c magnetic field for the $x=0.05$ sample the superconducting transition temperature was found to be 98.9 K. This sample also has one superconducting phase. We also observed a small kink in the $\chi'(T)$ data for this sample which begins at a temperature of 97.5 K. This kink is due to the weak link coupling between the superconducting grains. This kink in $\chi'(T)$ for the Bi based superconductors was also previously reported by others [4]-[6]. As can be seen the $\chi'(T)$ data for the $x=0.1$ sample a single drop was observed indicating that the sample had only one superconducting phase. The onset superconducting transition temperature of this sample was found to be 56.7 K.

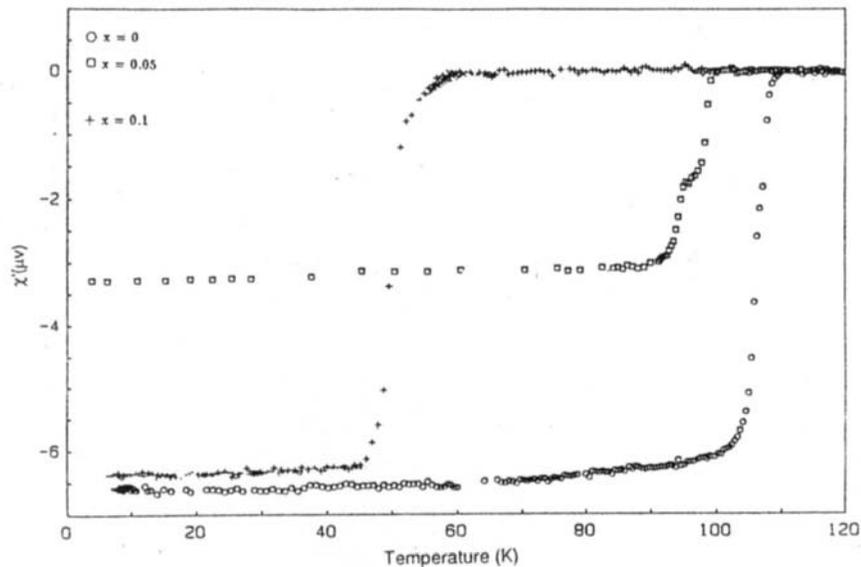


Figure 1. The measurements of $\chi'(T)$ in zero d.c magnetic field for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ samples with $x=0$, $x=0.05$ and $x=0.1$.

5.2 The measurements of $\chi'(T)$ in applied d.c magnetic fields.

The measurements of $\chi'(T)$ for the $x=0$ sample in several d.c magnetic fields which ranged from 0 to 500 G are presented in figure 2. From the experimental results of $\chi'(T)$ it is clear that superconducting transition of the sample became broadened with increasing applied fields which indicates that the penetration of the d.c magnetic field into the sample quenches the superconductivity. The broadening of the superconducting transition of Bi based superconductors in d.c magnetic fields was also reported by others [6]- [9]. The $\chi'(T)$ in low applied d.c magnetic fields show two diamagnetic contributions and, in high magnetic field of 500 G we observe only one single stage transition in $\chi'(T)$. The two drops in $\chi'(T)$ in low magnetic fields are usually attributed to the intergrain and intragrain regions of the sample. These two drops in d.c magnetic fields were also observed by W. Lang et. al [6] for the Bi based superconductors. In high magnetic field these two drops in $\chi'(T)$ disappeared because high magnetic field penetrated simultaneously deeply into the superconducting grains as well as into the weak links. It is clear that the high magnetic fields also pushed the onset of the superconducting transition of the sample to lower temperatures. An applied d.c magnetic field of 500G was able to shift the superconducting transition temperature of the sample by as much as 13.7 K to lower temperatures. The shift of superconducting transition temperatures of BSCCO superconductors to lower temperatures in d.c magnetic fields were also reported by others [6],[8],[9].

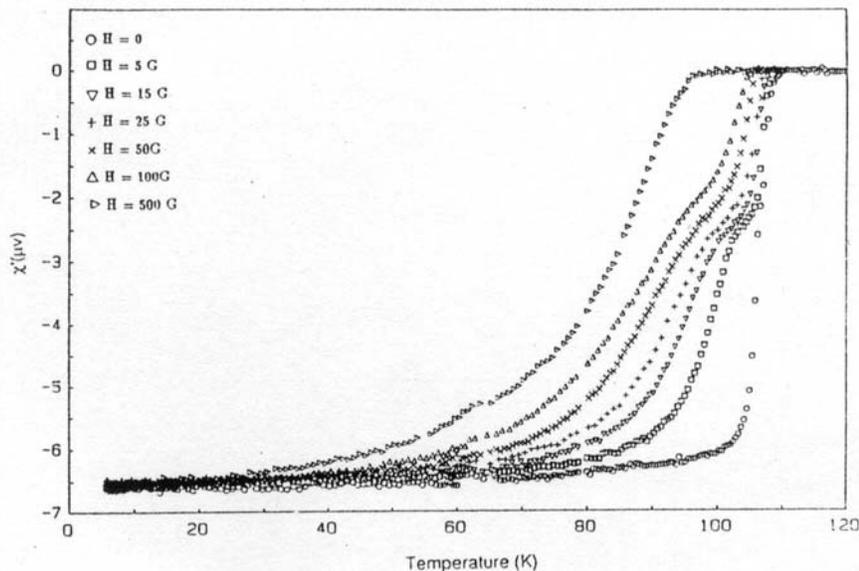


Figure 2. The measurements of $\chi'(T)$ in d.c magnetic fields for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ sample with $x=0$.

The measurements of $\chi'(T)$ for the $x=0.05$ and $x=0.1$ samples in several d.c magnetic fields are presented in figure 3 and figure 4 respectively. The $x=0.05$ sample also shows broad transition and two diamagnetic contributions associated with inter and intra granular regions and in high magnetic fields the sample shows only one transition. It is also clear that the onset of the superconducting transition temperature of this sample was shifted to 13.3 K by the applied d.c magnetic field of 500 G. It is interesting to note that in the presence of low d.c applied magnetic fields the $x=0.1$ sample exhibits only one single drop in $\chi'(T)$. These single drops in $\chi'(T)$ indicate that the ac magnetic field simultaneously penetrated deeply into the superconducting grains and the weak links of the sample in the presence of low and high magnetic d.c fields. It is also clear that the onset of the superconducting transition of this sample was shifted by 10 K to lower temperature by the applied magnetic field of 500 G.

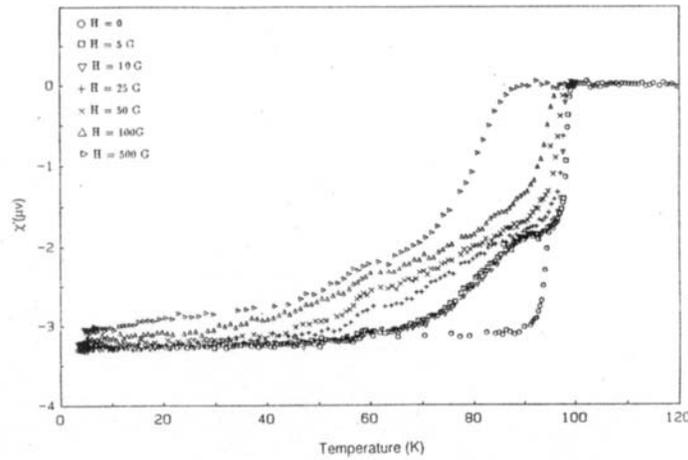


Figure 3. The measurements of $\chi'(T)$ in d.c magnetic fields for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ sample with $x=0.05$.

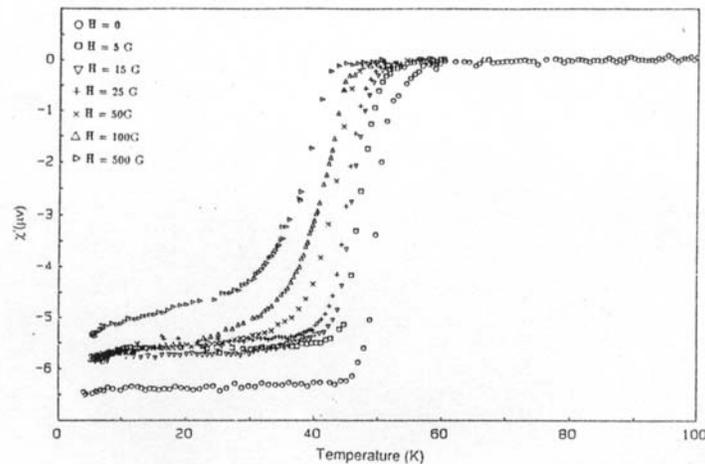


Figure 4. The measurements of $\chi'(T)$ in d.c magnetic field for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ sample with $x=0.1$.

5.3 The measurements of $\chi''(T)$ in zero and in applied d.c magnetic fields.

The measurements of $\chi''(T)$ in zero and in applied d.c magnetic fields for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ samples with $x=0$, $x=0.05$ and $x=0.1$ are presented in figures 5, 6 and 7 respectively. It is clear from the $\chi''(T)$ data in zero d.c field that single sharp peaks associated with energy dissipation in the intergranular regions of the samples are observed. The intergrain peak temperatures for the $x=0$, $x=0.05$ and $x=0.1$ samples were found to be 105.8 K, 94.7 K, 49.3 K respectively and at these temperatures the energy dissipation in the samples are maximum and ac field and shielding current penetrated into the center of the samples.

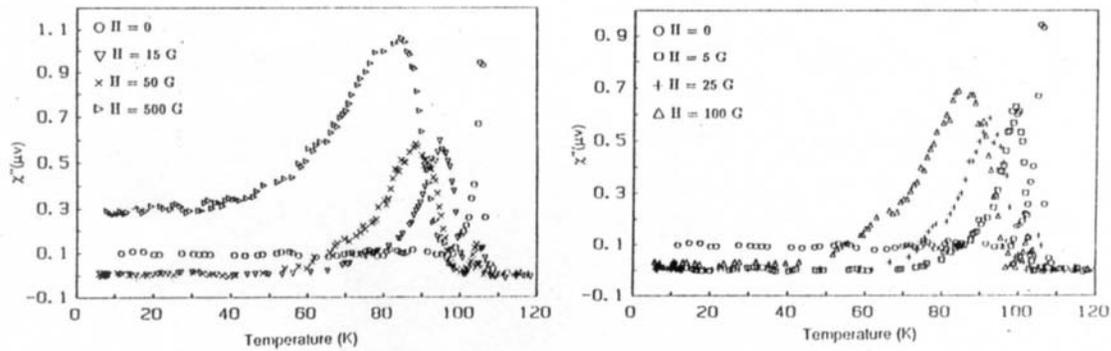


Figure 5. The measurements of $\chi''(T)$ in d.c magnetic fields for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ sample with $x=0$.

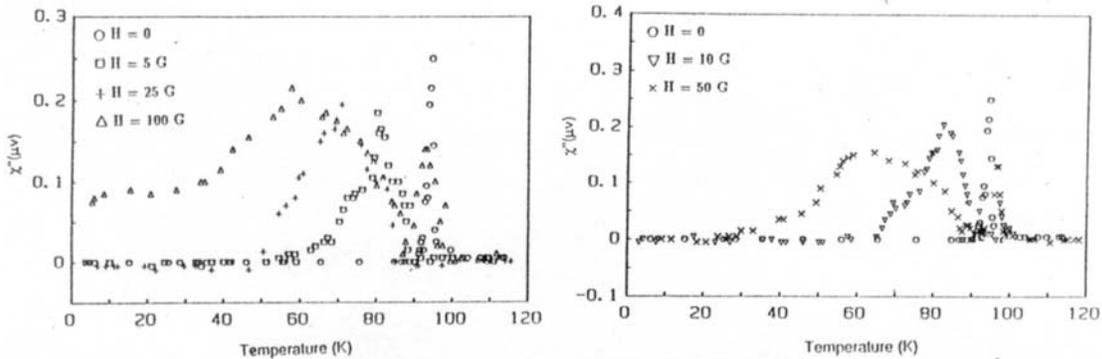


Figure 6. The measurements of $\chi''(T)$ in d.c magnetic fields for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ sample with $x=0.05$

It can be seen in the $\chi''(T)$ data for the $x=0$ sample in several d.c magnetic fields the broad intergrain peaks associated with the energy dissipation in the intergranular regions of the sample. These intergrain peaks in $\chi''(T)$ became broadened in accordance with $\chi'(T)$ data in d.c magnetic fields and shifted to lower temperatures as the applied magnetic field was increased. The intragrain peak shift to lower temperatures in the

presence of dc magnetic fields for the Bi based superconductor was also previously reported [6], [7]. From this data it is clear that the penetration of ac field and the shielding current into the center of the sample take place well below the superconducting transition temperature of the sample in the presence of dc magnetic fields. It can be seen in the $\chi''(T)$ data for the $x=0.05$ and 0.1 samples in several d.c magnetic fields the broad intergrain peaks in accordance with broad superconducting transitions in $\chi'(T)$ and these peaks are associated with the energy dissipation in the intergranular regions of the samples. These peaks shifted to lower temperatures with increasing applied d.c magnetic fields.

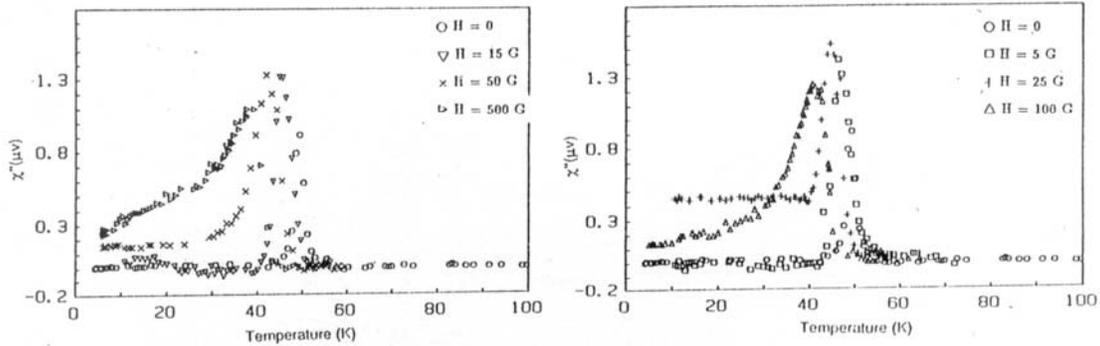


Figure 7. The measurements of $\chi''(T)$ in d.c magnetic fields for the $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ sample with $x=0.1$.

6. CONCLUSIONS

In this work the effect of doping of magnetic element Ni for copper into the copper site of Bi (Pb)-Sr-Ca-Cu-O superconductor was investigated. The ac magnetic susceptibility measurements $\chi'(T)$ and $\chi''(T)$ were carried out on three samples with composition $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Ni}_x\text{Cu}_{3-x}\text{O}_y$ with $x=0$, $x=0.05$ and $x=0.1$ in zero and in applied d.c magnetic fields. The superconducting transition temperatures of the samples were found to be 108.7 K, 98.9 K and 56.7 K respectively. Obviously superconducting transition temperatures of these samples decrease with increasing Ni concentration. The single drops in $\chi'(T)$ suggested that samples have only one Bi-2223 superconducting phase.

The $\chi'(T)$ data for the samples show broad superconducting transition and $\chi''(T)$ data show broad intergrain peaks in applied d.c magnetic fields indicating that the d.c magnetic fields quenched superconductivity of the samples. In low d.c. magnetic fields $\chi'(T)$ data of $x=0$ and $x=0.05$ samples show two drops associated with inter and intra grain regions of the samples. In high magnetic fields $\chi'(T)$ data shows a single transition indicating that the magnetic field simultaneously penetrated deeply into the superconducting grains and the weak links. The $\chi'(T)$ data for the $x=0.1$ sample shows

single drops in low and in high d.c magnetic fields. The high d.c magnetic field pushed the superconducting transition temperatures of the samples to lower temperatures. The intergrain peaks shifted to lower temperatures with increasing applied d.c magnetic fields.

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