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HIGH PRESSURE ELECTRICAL RESISTIVITY OF COMPOSITION CONTROLLED Nd-123 BASED BULK MATERIAL

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High pressure electrical resistivity studies were carried out on the high temperature superconductor $Nd_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ for various calcium concentrations $x = 0.00$, 0.03, 0.06 and 0.10 obtained by the solid-state reaction method. The electrical resistivity study was performed using the four-probe technique with a Bridgman opposed anvil device. All four samples show an initial drastic fall in electrical resistivity up to a pressure of around 3 GPa that remains almost constant up to 8 GPa.

Keywords: High-T_c superconductors; Nd-123; high-pressure; X-ray diffraction; electrical resistivity.

1. Introduction

A strong T_c-dependence on additional charge in the plane has been observed for the Nd-123 superconductor (much stronger than other 123 compounds) and a maximum T_c is reached close to $NdBa_2Cu_3O_{7-\delta}$. The increase or decrease of the negative charge in the planes by doping leads to a decrease of T_c.[1] It is clear from the study made on Ca doped orthorhombic $YBa_2Cu_3O_{7-\delta}$ crystals that, the oxygen content decreases with calcium doping. This is due to a charge balance associated with the substitution for Y^{3+} with Ca^{2+} and also due to the effect of the increasing oxidation state of CuO. The increase of Ca substitution gives a decrease in the orthorhombic strain.[2] This is the main cause for the depression of superconductivity.[3,4] Extensive studies such as hole doping,[5] fluctuation conductivity,[5] flux pinning behavior,[6,7] microstructure[8] and neutron spectroscopy[9] have been made on this system with a view to understanding the homogeneity. It has been reported that the measurement of irreversibility field (B_{irr}), i.e. magnetic measurements made as a function of

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temperature and critical current density (J_c) for Ca substituted Nd-123 have better flux pinning properties than Y-123.[10]

Composition controlled $Nd_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ superconductors have evoked considerable interest mainly because of their wide homogeneity range. The highest T_c value reported for Nd:Ba ratio of 0.5 concentration was 96 K for pure Nd-123.[11] Hence, it is of interest to study the properties of the calcium doped Nd-123 system under high pressure. The transition temperature measurement done on calcium doped Nd-123 shows that $T_{c\,max}$ (maximum transition temperature) decreases with increasing Ca content. This phenomenon is opposite to the observed phenomena in Ca substituted at the yttrium site of the Y-123 system.[12] X-ray powder diffraction studies on Ca doped Nd-123 system have shown that they are crystallized in an orthorhombic structure. Substitution leads to a decrease in the a and b lattice parameters and a small increase in the c lattice parameter. A faster decrease in the b lattice parameter compared to a causes the decrease in the orthorhombicity.[9] Normal state electrical resistivity studies on $Nd_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ have shown improved metallic behavior with increasing calcium doping. In this paper, the variation of electrical resistivity measurements at room temperature for various pressures for different calcium concentrations is presented.

2. Experimental Details

$Nd_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ samples with calcium concentrations ($x = 0.00$, 0.03, 0.06, 0.10) were synthesized by the standard solid-state reaction method.[11] High purity Nd_2O_3, $BaCO_3$, CuO and $CaCO_3$ in stoichiometric composition were ground finely and pressed into pellets. The pellets were then calcined at 1173 K, 1193 K and at 1193 K with intermediate grindings. They were annealed in flowing oxygen at 733 K for three days and the temperature was finally decreased to room temperature at the rate of 12 K/hr. The samples were post annealed by pressing into pellets and sintered at 1213 K for 12 hours and allowed to cool to room temperature.

Energy Dispersive X-Ray Diffraction (EDXRD) studies were carried out on the calcium doped Nd-123 system using white X-rays of the copper target produced by the rotating anode X-ray generator, Rigaku. The systems were indexed with an orthorhombic structure using XRD analysis software. The EDXRD pattern obtained for the parent compound and the calcium doped superconductors are shown in Figs. 1(a)–1(d). The X-ray diffraction pattern of the samples shows the formation of the single phase. The lattice parameters were calculated and are in agreement with the literature.[4,9]

High pressure electrical resistivity studies at room temperature were carried out on the calcium doped Nd-123 system using the standard four-probe method with the Bridgman Opposed Anvil apparatus. The opposed anvils are made of EN 24 alloy steel hardened to RC 60. The working face of the anvil has a diameter of 10 mm and the outer diameter is 100 mm. Pyrophyllite gaskets of dimension 10 mm outer diameter, 2 mm inner diameter and a thickness of 0.25 mm were used. Steatite was

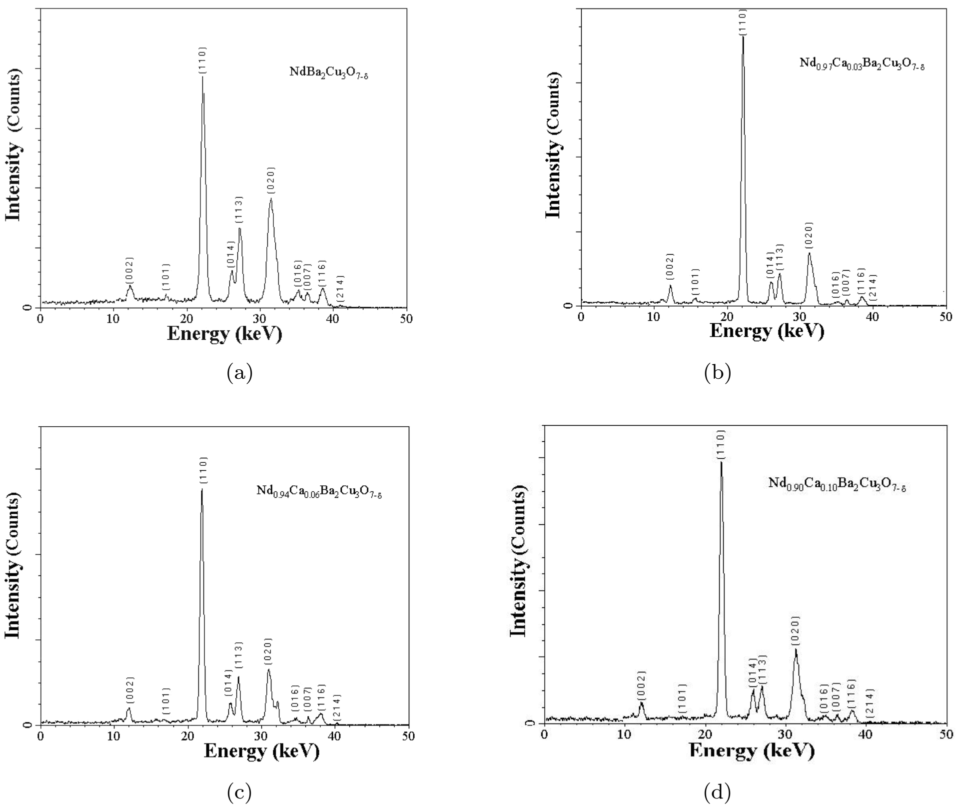

Fig. 1. Energy dispersive X-ray diffraction pattern of $\mathrm{Nd}_{1-x}\mathrm{Ca}_x\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-\delta}$ for $x = 0$, 0.03, 0.06 and 0.10.

the pressure-transmitting medium. Steatite of 2 mm diameter was placed firmly at the center of the gasket. Bismuth was used as the pressure calibrant.

3. Results and Discussion

The normalized electrical resistivity ($\rho/\rho_{0.3\,\mathrm{GPa}}$) versus pressure of orthorhombic $\mathrm{Nd}_{1-x}\mathrm{Ca}_x\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-\delta}$ for the calcium concentrations $x = 0$, 0.03, 0.06, 0.10 measured at room temperature is shown in Fig. 2. All the calcium doped samples including Nd-123 shows an initial monotonous drop up to 3 GPa to 4 GPa above which there is a nearly constant value of relative electrical resistivity up to 8 GPa.

In the normal pressure versus relative electrical resistivity curve, the pressure seems to have very little or comparatively no effect on relative resistivity beyond 3–4 GPa. An increase of pressure above 4 GPa causes the modification of the electron band structure, which leads to increased phonon scattering. The electrical resistivity decreases with increasing calcium concentration at the Nd site, which is in agreement with reported results,[9] confirming improved metallic properties of the Ca doped Nd-123 samples. High pressure studies above 8 GPa would further im-

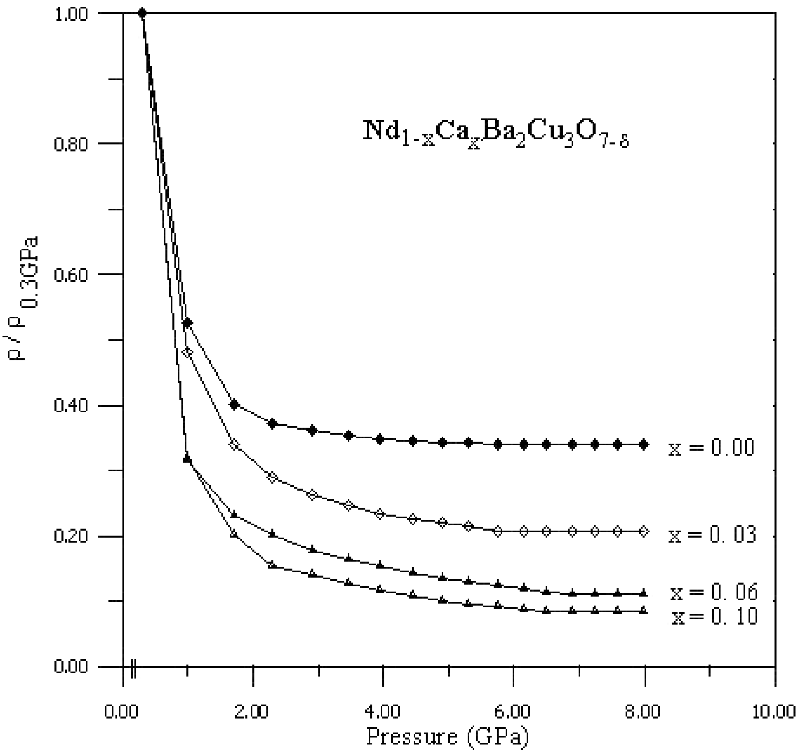

Fig. 2. Variation of Electrical resistivity on $Nd_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ for $x = 0$, 0.03, 0.06 and 0.10.

prove understanding about this system. Generally, for a metal, as lattice scattering controls the electronic conduction and as the compression reduces the amplitude of the lattice vibrations, a moderate decrease in resistance with pressure is a normal process.[12] From Fig. 3, it can be seen that the reduced volume fraction for the sintered samples are 12% at atmospheric pressure and even less at high pressures. Hence, the initial drastic decrease in relative electrical resistivity may be caused partly or to a very small extent due to the role of compaction or rearrangement of voids in these samples.

A release of pressure regained the original value of $(\rho/\rho_{0.3\,\mathrm{GPa}})$ indicating that the behavior is reversible with pressure with small hysterisis, except a decrease in the ρ value for the unloading curve. The loading and unloading curves obtained for the parent compound Nd-123 are shown in Fig. 3. It can be asserted that the initial decrease in the electrical resistivity is not due to the compaction of the sample. From the plot between relative electrical resistivity and calcium concentration shown in Fig. 4(b), it can be seen that the metallicity increases with increasing calcium concentration at the Nd site. Variation of volume with the calcium concentration for $x = 0$, 0.03, 0.06 and 0.10 is shown in Fig. 4(a). This shows a decrease

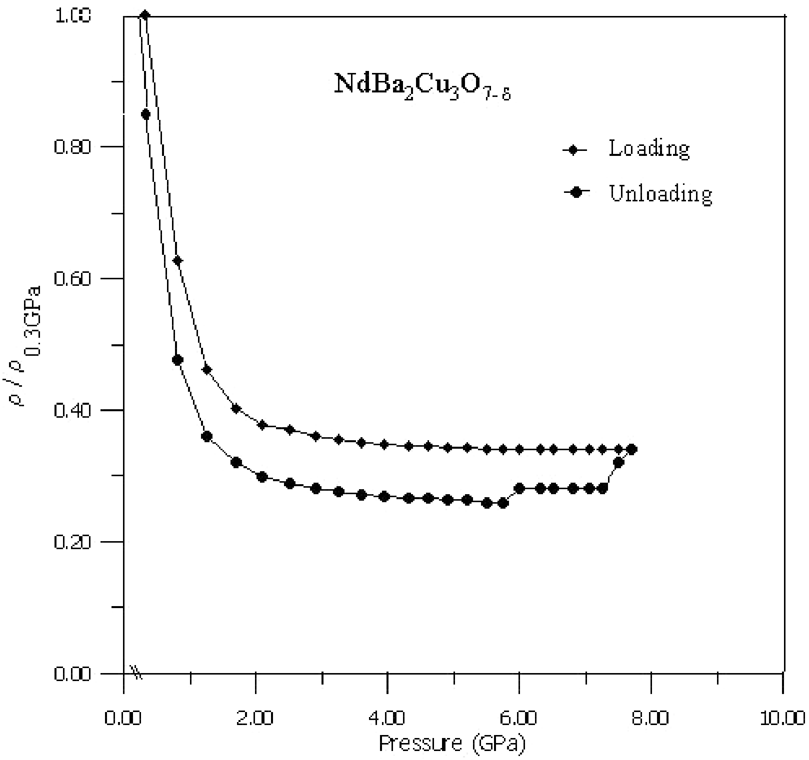

Fig. 3. Loading and unloading curves for Nd-123 measured at room temperature.

in volume with increasing calcium concentration. The decrease in volume is due to the smaller size of the calcium atom compared to neodymium.

The electrical resistivity behavior of the calcium doped Nd-123 system may be consistent with a layered material, which lies between two and three dimensions. This evidence lies in the identification of two different regions of behavior under pressure as shown in Fig. 1. At pressures below 2 GPa, the electrical resistivity decreases strongly with increasing pressure while it saturates at high pressures. The two regions may be attributed to the layered structure. At low pressures, interlayer spacing changes more rapidly than the intralayer bonds. Therefore, the two-dimensional layered structure becomes more three-dimensional under pressure. The decrease in the electrical resistivity in the region below 3 GPa corresponds to the decrease in interlayer spacing. To obtain intralayer changes, pressures greater than 8 GPa are required, since intralayer bonds are stronger.

Table 1 gives the lattice parameter, transition temperature (T_c), transition pressure (T_p) (here, the transition pressure is the maximum pressure required for the two-dimensional structure of the sample to become completely three-dimensional. T_p is obtained from the relative resistivity plot) and the transition width (ΔT_c)

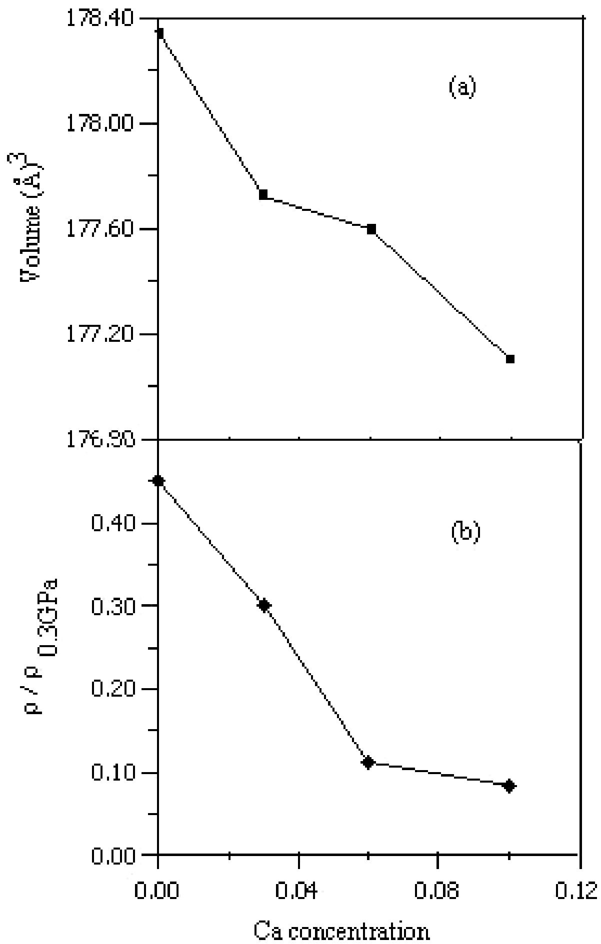

Fig. 4. (a) Variation of volume with calcium concentration of $Nd_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$. (b) Variation of relative electrical resistivity versus calcium concentration in $Nd_{1-x}Ca_x$-$Ba_2Cu_3O_{7-\delta}$.

Table 1. Variation of transition pressure, transition temperature and lattice parameters for the various calcium concentrations compared with pure Nd-123.

	Lattice parameters (Å)			T_c	T_p	ΔT_c
Nd : Ca	*a*	*b*	*c*	(K)	(GPa)	(K)
1.00 : 0.00	3.8724	3.9224	11.742	96	1.4	2.7
0.97 : 0.03	3.8681	3.9170	11.743	93.3	1.3	2.4
0.94 : 0.06	3.8652	3.9105	11.750	94	1.1	1.7
0.90 : 0.10	3.8604	3.9040	11.751	95	1.5	1.0

for various calcium concentrations. The values of the transition pressure, transition temperature and the lattice parameters for the calcium concentrations $x = 0.03$, 0.06 and 0.10 is compared with the parent compound shown in Table 1. Here, the trivalent Nd substituted with divalent Ca causes an increase in the hole concentration at the lattice, which leads to the depression in the transition temperature. With the increase of Ca concentration the hole concentration increases further and hence causes a decrease in the electrical resistivity. Improved electrical properties under high pressure has been reported for Ca doped Nd-123 superconductors. It has been suggested that an enhancement in the transition temperature may be achieved for these samples when pressurized.

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