

# Pressure and temperature effect on the electrical resistivity of $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$

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## Abstract

High-pressure and high-temperature electrical resistivity studies on composition controlled  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  superconductor have been done by four-probe technique using Opposed Anvil High-Pressure Device (OAHPD). The electrical resistivity measurements were carried out up to a maximum pressure of 8 GPa and at various temperatures up to a maximum of 523 K in steps of 50 K using the heating coil arrangement. Simulation of the X-ray diffraction pattern confirms the structure of the sample. A gradual decrease in the electrical resistivity with increase of pressure was observed. The observed decrease in the electrical resistivity on application of pressure is discussed with respect to the structural changes under pressure. Measurements were performed up to a pressure and temperature of 8 GPa and 523 K, respectively. Improved electrical resistivity is observed under pressure. The temperature effect causes an upward shift in the electrical resistivity in the observed range of pressure.

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## 1. Introduction

Early high-temperature solid-phase synthesised superconductors had low density, mechanical characteristics and critical current densities, and so, they degraded fast under ambient conditions. Thus, they were not suitable for practical applications [1]. Improved critical current density, high transition temperature and wide homogeneity range are some of the interesting properties of the Nd-123 superconductor that attracts the researchers. Several workers due to its wide homogeneity have done extensive studies on the properties of the Nd-123-based systems [2–5]. Also, when calcium is incorporated in RE-123 system, it preferentially substitutes at R site [1]. The main interest on doping the divalent calcium ion is to increase the hole concentration, thereby improving the superconducting properties in the Y-123 system. Normal-state electrical resistivity measurements on Nd-123 for various concentrations of Ca at the neodymium site

show improved metallic behaviour, and its critical temperature is found to be higher than in other 123 compounds [6]. The interest in  $\text{NdBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -based superconductors can be explained by the presence of a peak effect, i.e., the increase in critical current density in external magnetic field as compared to its value in self-field [7] and the high irreversible fields greater than that of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Also, the possibility to achieve high rates of melt texturing about 50 times faster than in the case of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  because of the higher solubility of the Nd in the melt as compared with Y led us to study this system. Due to its use in wide areas of applications,  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  has been chosen to study its electrical behaviour under pressure and temperature up to a maximum of 8 GPa and 523 K, respectively. In  $\text{Y}_{0.83}\text{Ca}_{0.17}\text{Ba}_2\text{Cu}_3\text{O}_6$ , two superconducting transitions have been observed under high pressure; the pressure-induced superconductivity at the lower  $T$  and the pressure enhanced one at the higher  $T$ . It was reported that the former is the filamentary superconductivity induced by pressure. The origin of the latter is attributed to the charge redistribution induced by pressure [6]. Hence, It is interesting to study the effects

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of pressure and temperature on the calcium-doped Nd-123 system.

Here, the electrical resistivity study on  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  is performed with respect to pressure and temperature up to a maximum of 8 GPa and 523 K, using the opposed anvil high-pressure device and the results are presented.

## 2. Experimental

The composition controlled  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  were synthesised by solid-state method [7]. Stoichiometric compositions of high purity  $\text{Nd}_2\text{O}_3$ ,  $\text{BaCO}_3$ ,  $\text{CuO}$ ,  $\text{CaCO}_3$  were ground finely and made into pellets. The pellets of the mixture were calcined at 1173, 1193 and at 1193 K with intermediate grindings. They were annealed in flowing oxygen at 733 K for 3 days, and the temperature was finally decreased to room temperature at the rate of 12 K/h.

Energy Dispersive X-ray Powder Diffraction (EDXRD) study is carried out on the  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  system using white X-rays produced by copper target using a rotating anode X-ray generator, Rigaku. The X-ray diffraction measurement confirms the formation of a single phase. The system was indexed using X-ray diffraction analysis (XRDA) software [8], and it is found to crystallise in an orthorhombic structure with space group *Pmmm*. EDXRD simulation has been done by theoretical calculation of intensities using X-ray powder diffraction (XPOW) software [9]. The experimental EDXRD pattern and the simulated pattern for the samples are shown in Fig. 1. The simulated EDXRD pattern confirms the substitution of calcium at the Nd site.

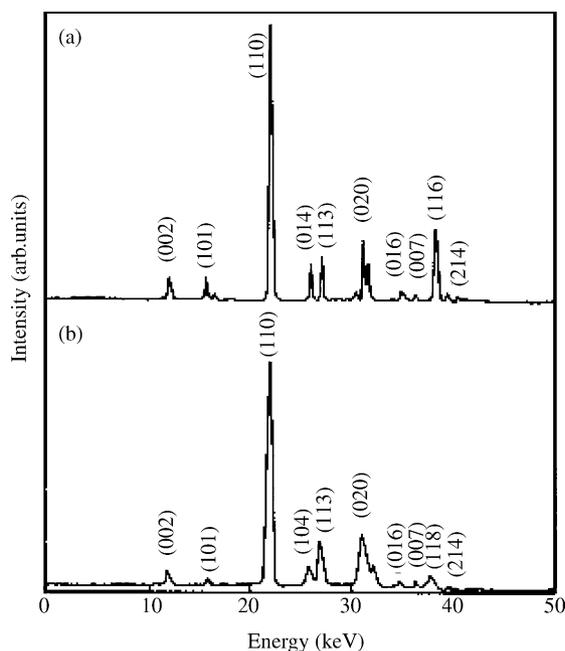


Fig. 1. (a) Experimental and (b) simulated EDXRD pattern of  $\text{Nd}_{0.97}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ .

The electrical resistivity studies at high-pressure and high-temperature were done using Opposed Anvil High Pressure Device which is made up of EN24 alloy steel hardened to Rockwell 60 [10] along with a heating coil attachment which is shown in Fig. 2. The temperature stability obtained was  $\pm 2$  K. The power to the coil is controlled by a variac, and the temperature has been maintained using a temperature controller [11]. In this arrangement, a chromel–alumel thermocouple connected together by spot welding is used as a temperature sensor. The spot-welded junction was hammered and made very thin and flat. Sample of thickness 0.1 mm has been used for the resistivity measurements. Necessary corrections for the effect of pressure on the thermal e.m.f. were made according to Cheng et al. [12,13]. The thermocouples, sample and the heater were assembled for good thermal contact, and the mica sheets and tapes were used for insulating purposes. The position of the sample and the thermocouple in the high-pressure and high-temperature arrangement using heating coil setup is shown in Fig. 2.

## 3. Results and discussion

The EDXRD study shows that the sample is crystallised in orthorhombic structure. The lattice parameters are found to be  $a = 3.87 \pm 0.01 \text{ \AA}$ ,  $b = 3.91 \pm 0.01 \text{ \AA}$ ,  $c = 11.75 \pm 0.01 \text{ \AA}$  and  $V = 177.6 \text{ \AA}^3$ . The X-ray diffraction pattern shows formation of a single phase. The lattice parameters obtained are in agreement with the literature [14,6]. A decrease in volume is observed on substitution of calcium at the Nd site. EDXRD simulation studies confirm the substitution of the calcium at the neodymium site.

The variation of high-pressure and high-temperature electrical resistivity of  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  is shown in Fig. 3. Better contacts were established by giving a small load, which corresponds to 0.3 GPa at the sample site. Hence, resistivity at 0.3 GPa was chosen for the normalisation. The measurements were carried out for both loading and unloading pressures. It was found that the behaviour is reversible

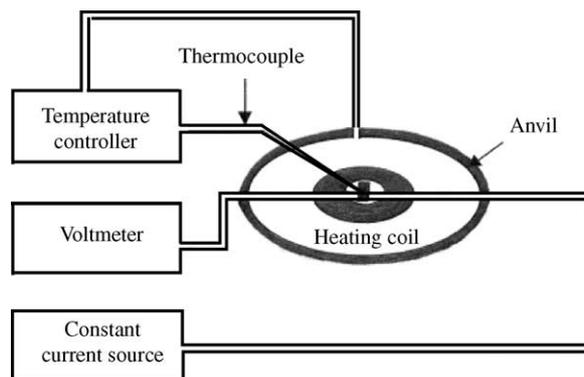


Fig. 2. High pressure–high temperature electrical resistivity setup.

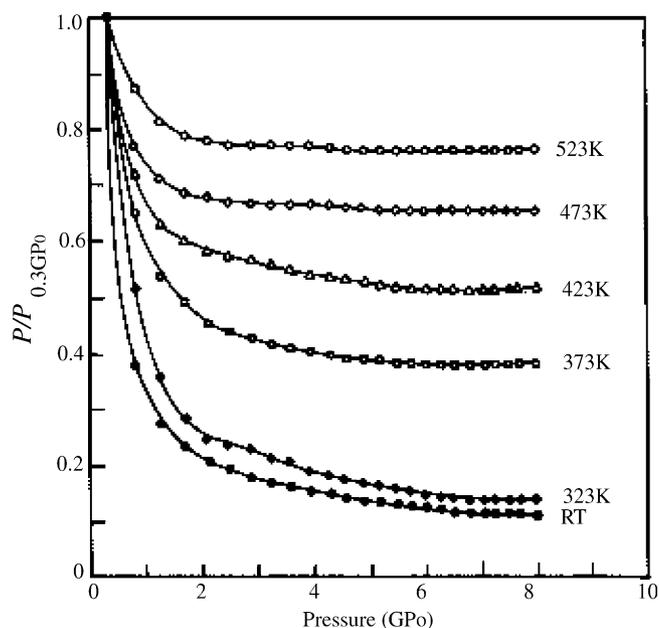


Fig. 3. Variation of electrical resistivity of  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  studied up to 8 GPa at various temperatures;  $RT = 308$  K.

under pressure with small hysteresis, except for a small decrease in  $\rho$  value for the unloading curve. From the loading and unloading behaviour, it is also evidenced that the initial decrease in the resistivity is not mainly due to the compaction of the sample. Thus, it is a real effect caused due to the structural changes in the sample on application of pressure.

At initial pressures, a rapid decrease in  $\rho$  is observed which becomes almost constant at high pressures up to 8 GPa for all the temperatures. A systematic upward shift in resistivity with temperature is observed up to 523 K (maximum temperature studied). Absence of phase transitions under pressure and temperature up to a maximum of 8 GPa and 523 K,

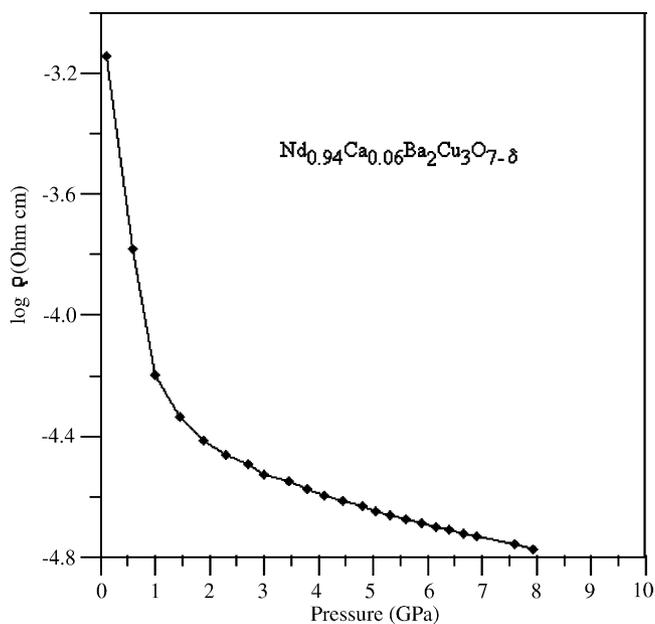


Fig. 4.  $\log \rho$  versus pressure of  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  at room temperature.

respectively, is reported. The behaviour confirms improved metallic nature of the sample up to 523 K and a maximum pressure of 8 GPa.

The variation of  $\log \rho$  versus pressure is plotted and shown in Fig. 4. The two regions (i.e., low and high-pressure region) corresponds to inter- and intra-layer changes, which may be attributed from the figure and confirms the layer like structure. The initial rapid decrease in  $\rho$  of the  $\log \rho$  versus pressure curve shows the effect due to changes in the inter-layer bonds and beyond which the inter-layer is not affected much, which is shown by a nearly constant curve at high pressures. From the high pressure–high temperature studies, it is understood

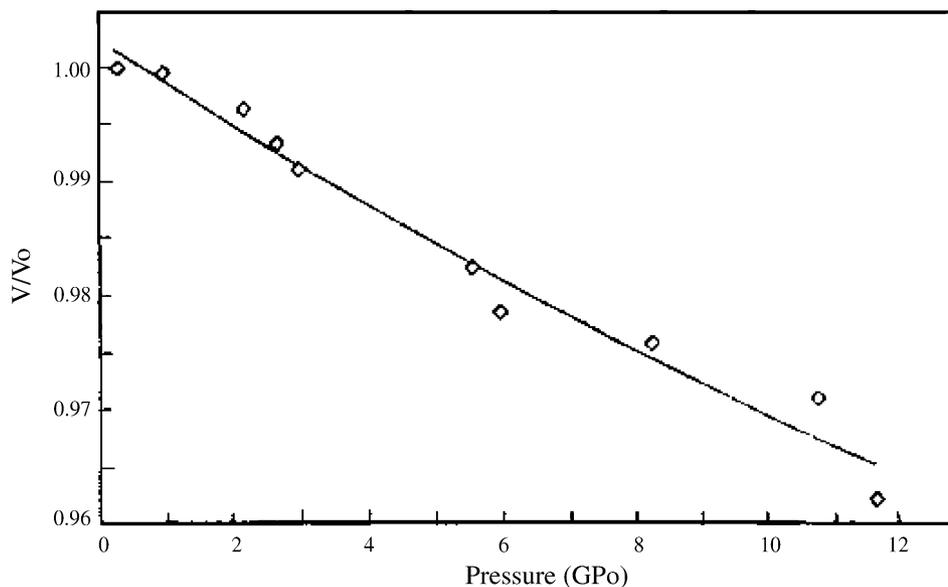


Fig. 5. Variation of  $V/V_0$  versus pressure for  $\text{Nd}_{0.94}\text{Ca}_{0.06}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ ;  $V_0 = 177.6$  Å.

that, the application of even a small pressure in the layered metallic copper oxides increases the carrier concentration between the copper oxide layers, the so-called charge reservoirs, thereby improving the carrier concentration in the Nd-123 superconductor [15]. Alternatively, temperature opposes the interlayer bond breaking effects caused by the pressure. This is the cause for the shift in the resistivity with temperature in the sample studied.

The intra-layer bond changes may be expected at higher pressures greater than 8 GPa since they are stronger than interlayer bonds. The high-pressure X-ray diffraction measurements were employed on the sample using a diamond anvil cell, and the experimental setup is given elsewhere [16]. The plot between the relative volume with pressure is shown in Fig. 5. The changes in the bond distances (caused by the inter-layer bond changes) due to the application of the pressure are confirmed from the X-ray diffraction measurements up to 12 GPa. The bulk modulus decreases with 6% doping of calcium from 377 to 311 GPa. Substitution of calcium at the neodymium site of the Nd-123 superconductor promotes additional holes in the CuO layers, which in turn decreases the resistivity of the material. The chemical substitution itself causes the lowering of puckering effect. This was also confirmed from the bond distances, which were calculated from the X-ray diffraction data. It was observed that the application of pressure reduces the puckering effect strongly in the CuO planes, thus making more holes available for the conduction.

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