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# An electronic structure study of *c*-axis oriented NdBCO (123) thin films using polarized soft x-ray absorption spectroscopy on Cu L<sub>3</sub> and O K edges

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## Abstract

Polarized x-ray absorption spectroscopic (XAS) measurements have been made on Cu L<sub>3</sub> and O K edges on four superconducting thin films of NdBCO with different  $T_c$  values to study the relative importance of the in-plane (Cu 3d<sub>x<sup>2</sup>-y<sup>2</sup></sub> and O 2p<sub>x,y</sub> orbitals) and the out-of-plane (Cu 3d<sub>z<sup>2</sup></sub> and O 2p<sub>z</sub>) orbitals in the mechanism of superconductivity. Our results clearly show that a significant number of out-of plane O 2p<sub>z</sub> and Cu 3d<sub>z<sup>2</sup></sub> do exist in these films, still hardly affecting their  $T_c$ .  $T_c$  is found to depend more upon number of itinerant holes rather than the orientation or substrate of the film. However the out-of-plane holes perhaps do affect the superconducting fraction in the film. O K-edge spectra recorded for  $E \parallel ab$  to measure the density of itinerant holes in plane show a good agreement with the conclusion drawn from Cu L<sub>3</sub> data. The results have been compared with the XRD and the magnetic susceptibility measurements reported on these films earlier and a good qualitative agreement found. From the present study we can confidently state that no direct correlation exists between  $T_c$  and out-of-plane covalent and doping hole densities and the models based on the premise that out-of-plane orbitals, if present, will destroy superconductivity also do not seem to be valid in these systems.

## 1. Introduction

Since the discovery of high  $T_c$  superconducting oxides, tremendous efforts have been made to synthesize thin films of these materials, using various techniques, from both scientific

and technological viewpoints. Thin film techniques have become a key technology for the purposes of electronic and power applications of high temperature superconductors. Amongst others, NdBCO superconductors are promising candidates for high field applications since large critical current densities ( $J_c$ ) are achieved in the high field region when these are fabricated by the oxygen-controlled melt growth process [1–3] and these films have been found to be more suitable for device applications. The materials contain fine Nd rich Nd123 clusters about 10–50 nm in diameter with depressed critical temperature, which can act as field-induced pinning centres, leading to the so-called secondary peak effect [1]. The high  $T_c$  of NdBCO of 96 K and its large  $J_c$  [1, 2] have encouraged the study of the thin films using various methods. The surfaces of NdBCO films are much smoother than those of the high quality PLD (pulsed laser deposition) deposited YBCO [4].

X-ray absorption spectroscopy and other high energy spectroscopy techniques have proved to be of great use in the study of important question of the symmetry of the itinerant doping holes in high temperature superconductors (HTSCs) [5–11]. Most of the theoretical models and the reports agree that O 2p and the Cu 3d holes have a dominant in-plane symmetry, but with a significant contribution of out-of plane O 2p holes. But there is no such consensus on the existence of out-of-plane Cu  $3d_{z^2-r^2}$  orbitals and the estimates of their weight vary from 0 to as great as 20% for different HTSC systems [7, 10, 11]. The question of the presence or absence of the Cu  $3d_{z^2-r^2}$  orbitals and their relevance, if any, to superconductivity continues to hold interest for both theoretical and the experimental studies on these materials.

Many theoretical multi-band models that favour an active role for the Cu  $3d_{z^2-r^2}$  and O  $2p_z$  orbitals and the three band model which uses only the Cu  $3d_{z^2-r^2}$  and O  $2p_{x,y}$  orbitals have been proposed for describing the transport properties both for normal and superconducting states of HTSC materials. Some reports [12] propose an increment of  $T_c$  by the anharmonicity of the vibrations of apical oxygen. Importance of the Cu  $3d_{z^2-r^2}$ –O  $2p_z$  hybrids has been supported by some workers [13], while some others predict that the presence of out-of-plane orbitals will be detrimental to superconductivity.

In the present contribution we report the results of our  $E \parallel ab$  and  $E \parallel c$  polarized soft x-ray absorption measurements carried out at the O K and the Cu  $L_3$  edges on four high quality  $c$ -axis oriented NdBCO (123) polycrystalline thin films with different  $T_c$ , using the fluorescence-yield detection mode. In all these films the  $c$  axis is normal to the plane of the substrate. The purpose of the present study was to study the relative intensity of in-plane and out-of-plane holes in these systems and allow us to comment on the adequacy of the various existing theoretical models in the light of our results.

## 2. Experimental procedure

The preparation of these four NdBCO (123) films has been reported in detail elsewhere [14]. These films were grown by pulsed laser deposition, at 780 °C, in an argon rich atmosphere instead of oxygen (6 mTorr of oxygen for a total pressure of 200 mTorr). At the end of the deposition, the films were cooled to 500 °C over a duration of 10 minutes, in the deposition atmosphere. At 500 °C, the chamber was then filled with 500 mbar of pure oxygen and the films were subsequently cooled to room temperature.

The XRD and ac magnetic susceptibility measurements were made to determine the crystallization quality and  $T_c$  of the films. The x-ray absorption measurements on the NdBCO thin films were carried out at the beamline BL 11 A of the Synchrotron Radiation Research Centre (SRRC), Taiwan, using a bending magnet and a cylindrical element monochromator (CEM). The beamline comprises a horizontal focusing mirror (HFM) and a vertical focussing mirror (VFM) and four gratings, of different grating elements, to focus and monochromatize

the photon beam in different energy regions. The incident photon flux was of the order of  $1.1 \times 10^{11}$ . The experimental set-up yields a resolving power of  $\sim 10\,000$  at the Cu L<sub>3</sub> edge. A high-sensitivity nine-element Ge detector was used to record the spectra in the fluorescence yield (FY) mode. The x-ray absorption spectrum were recorded on the Cu L<sub>3</sub> and O K edge in fluorescence yield (FY) mode while the pressure at the end-station was measured  $\sim 5\text{E}(-10)$  mbar. Primary photon intensity  $I_0$  is recorded by measuring photocurrent from a clean gold mesh to normalize the intensity in the measured spectra.

The samples were mounted on a sample holder which could be rotated in the azimuthal as well as in the polar plane. This allowed the orientation of the plane  $ab$  and  $c$  axis with respect to the electric field vector of polarized incident beam in any desired direction. The in-plane spectra ( $E \parallel ab$ ) were obtained in a normal incidence alignment, while the out-of-plane spectra were recorded by rotating the films about a vertical axis. Such a series of angle-dependent measurements allows for the correction of in-plane misalignment. In order to reach out-of-plane orbitals, the samples were rotated to achieve angles of incidence of 15, 45, 60, 75, 80 degrees with respect to the sample normal.

Self-absorption correction (SAC) is essential for spectra measured in the fluorescence yield mode [15–18]. On their way to the sample surface the fluorescence x-rays are attenuated by the total absorption coefficient  $\mu_{tot}(E_f) = \mu_A(E_f) + \mu_{bac}(E_f)$  at the energy of fluorescence radiation,  $E_f$  for the absorber oxygen atom (oxy). The normalized fluorescence intensity  $\mu_{FY}$  is given by the relation

$$\mu_{FY} \equiv \frac{I_f(E)}{I_0(E)} \alpha \frac{\mu_{oxy}(E)}{\mu_{tot}(E)/\cos\alpha + \mu_{tot}(E_f)/\cos\beta}$$

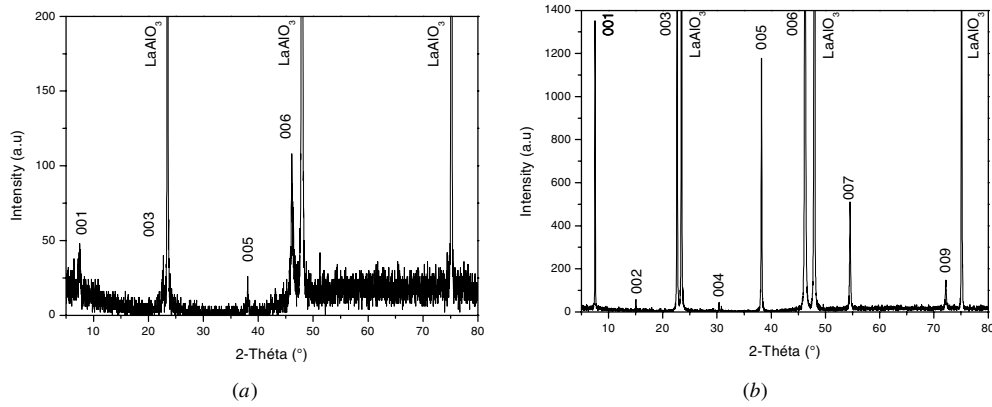
where  $I_f(E)$  is the intensity of the fluorescence radiation detected,  $I_0(E)$  the primary intensity,  $\alpha$  the angle between the incoming beam and the sample normal,  $\beta$  the angle between the sample normal and the outgoing beam and  $\mu_A(E_f)$  and  $\mu_{bac}(E_f)$  are the absorption coefficients of the absorbing and the back-scatterer atoms respectively. However, it is relatively less important for the case of O K edge as error introduced by it is less than about 3%. But, in case of the Cu L<sub>3</sub> white line, it leads to errors up to about 15% [17]. However, we have made a self-absorption correction for both the Cu L<sub>3</sub> and the O K spectra. We first normalized the fluorescence intensity to the intensity variations of the monochromatic light to obtain ( $I/I_0$ ) and finally we applied the self-absorption correction and spectra were normalized to the known cross sections at the 600 eV and at 1000 eV for the O K and Cu L<sub>3</sub> cases respectively as the absorption cross section becomes purely atomic-like at these energy values.

### 3. Results and discussion

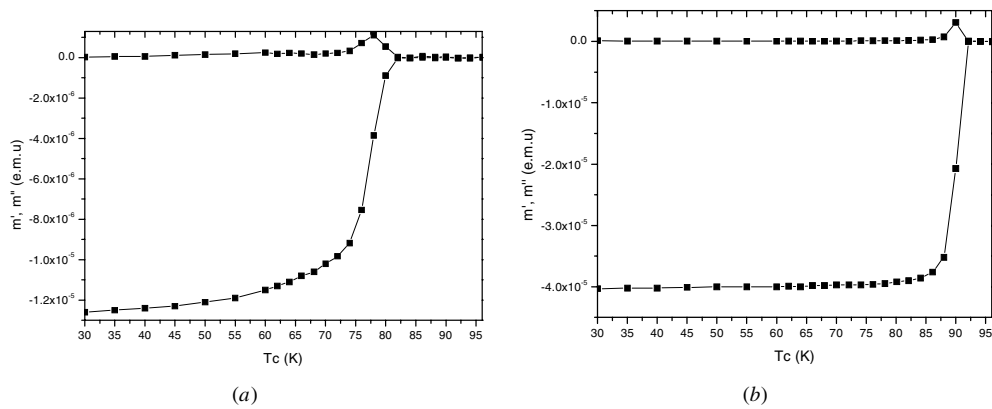
#### 3.1. XRD and susceptibility results

Before discussing the results of our Cu L<sub>3</sub> and O K edge x-ray spectroscopic data it is worthwhile to recall the results of characterization, i.e. magnetic susceptibility and XRD, made on these samples earlier [14]. The magnetic moment of these films falls off very sharply from zero to a negative value of the order of  $10^{(-5)}$  to  $10^{(-6)}$  indicating a well known and typical diamagnetic transition at their  $T_c$  (figures 2(a), 2(b) and 4(a), 4(b)). The value of their  $T_c$  are shown in table 1. Their XRD patterns are shown in figures 1(a), 1(b) and 3(a), 3(b). These films are purely monophasic with some peaks of the substrate LaAlO<sub>3</sub> and SrTiO<sub>3</sub> and exhibiting the  $c$  axis lying perpendicular to the substrate plane.

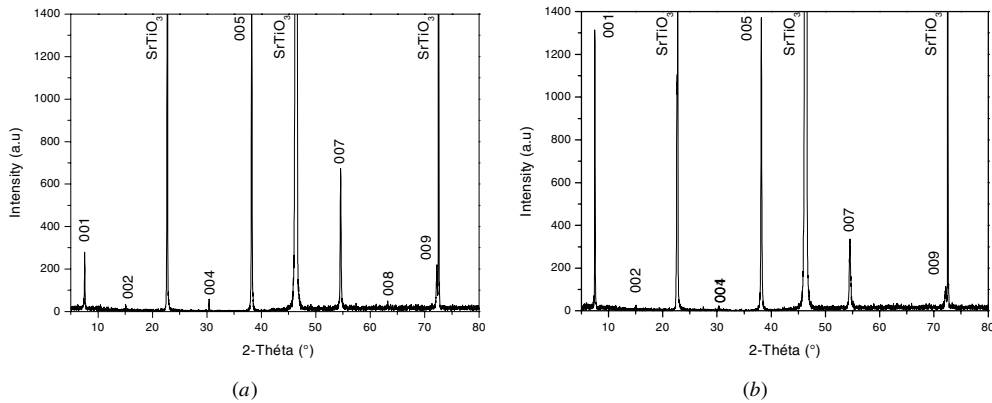
Samples 1 and 2 were deposited under the same conditions on an LaAlO<sub>3</sub> substrate, except sample 1 was quenched to 700 °C followed by the normal cooling parameters [14]. Figure 1(a) presents XRD patterns ( $\theta$ - $2\theta$  mode) for sample 1: the peaks are not well defined



**Figure 1.** XRD patterns of typical NdBCO thin films, sample 1 (84 K) and 2 (92 K) respectively deposited on  $\text{LaAlO}_3(100)$  [14].

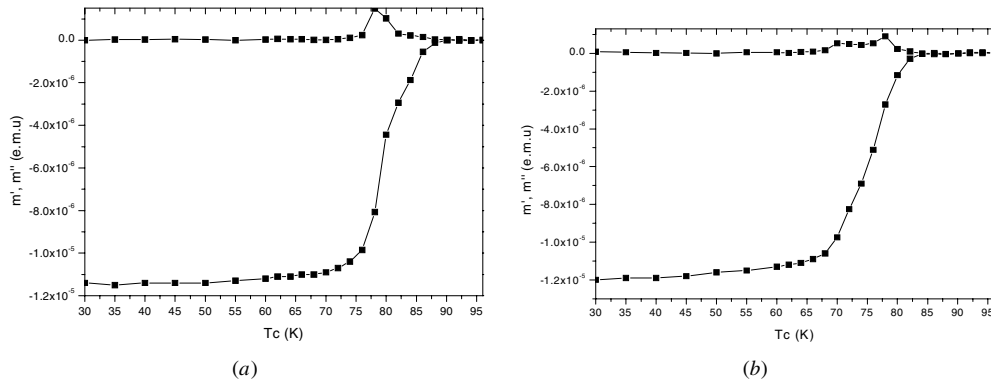


**Figure 2.** Temperature dependence of the ac magnetic susceptibility for NdBCO thin films, sample 1 (84 K) and sample 2 (92 K) respectively [14].



**Figure 3.** XRD patterns of typical NdBCO thin films, (sample 3 (90 K), sample 4 (81 K) respectively deposited on  $\text{SrTiO}_3(100)$  [14].

and may be overlapping. Also the (002) and (004) reflections of NdBCO are not observed, which means that the crystallization is not good. For sample 2, only sharp (00 $l$ ) reflections



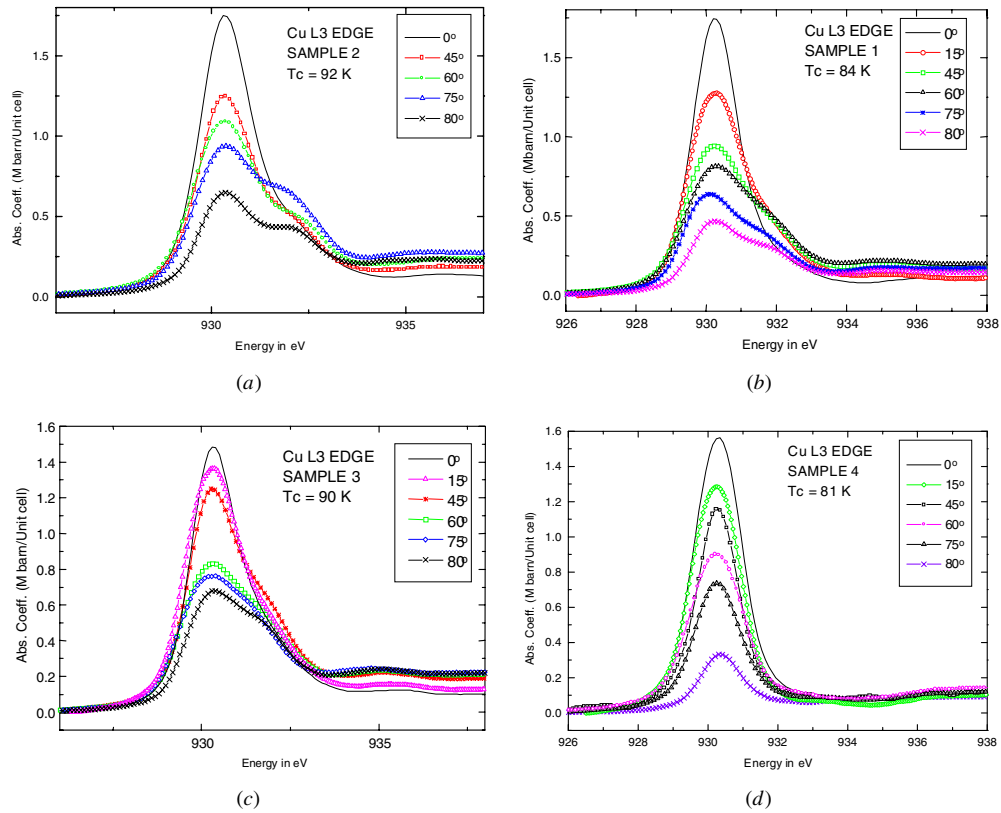
**Figure 4.** Temperature dependence of the ac magnetic susceptibility for NdBCO thin films, sample 3 (90 K) and sample 4 (81 K) respectively [14].

for NdBCO are observed (figure 1(b)). Sample 2 exhibits a narrow superconducting transition ( $\Delta T_c \approx 5$  K) at 92 K and a constant Meissner state at low temperature, which indicates a high superconducting volume fraction (figure 2(b)). In sample 1, however, the width ( $T_c$  of the superconducting transition ( $T_c = 81$  K) is large and the Meissner state is not constant at low temperatures, which can be attributed to the poor crystallization (see figure 2(a)).

In the second series of samples, films were grown on an SrTiO<sub>3</sub> substrate. Sample 3 shows sharp and strong (00 $l$ ) reflections corresponding to the NdBCO structure (figure 3(a)). All of the expected reflections are seen with the appropriate ratio between the intensities of the observed reflections. This indicates a good crystalline quality film, growing with the  $c$  axis perpendicular to the substrate plane.  $T_c$  was found to be 90 K, showing a constant Meissner state at low temperature (figure 4(a)). The XRD patterns for samples 3 and 4 are similar, but the (002) and (004) peaks for sample 4 (figure 3(b)) are weaker compared to those in sample 3 (figure 3(a)). In addition, the (008) reflection for sample 4 is not present, which indicates that sample 4 is not equally well crystallized. This result is supported by the ac magnetic susceptibility measurement (figure 4), indicating a  $T_c$  of 84 K, and the Meissner state is not seen to be constant at low temperatures (figure 4(b)).

### 3.2. Cu L<sub>3</sub> data

The polarization dependent Cu L<sub>3</sub> spectra recorded for the as-prepared films, are shown in figures 5(a)–(d) for the four thin films varying the polarization angle gradually from  $E \parallel ab$  to  $E \parallel c$  orientation. The spectra have been normalised at a point far below the main L<sub>3</sub> peak following the early reports [8, 9, 19], to ensure the true intensity ratio in the different incident angles with respect to the normal to the sample surface. For all these thin films the  $c$  axis lies perpendicular to the plane. The zero degree spectra thus correspond to the case when the E vector is in the  $ab$  plane (or  $E \parallel ab$ ) while the 80 degree spectra nearly correspond to the orientation  $E \parallel c$ . The dipole transition  $2p \rightarrow 3d$  for the Cu L<sub>3</sub> white line probes the density of unoccupied  $3d_{x^2-y^2}$  orbitals for  $E \parallel ab$  and  $3d_{z^2}$  orbitals for  $E \parallel c$ . The intensity of this line, due to the transition  $2p3d^9 \rightarrow 2p3d^{10}$ , is found to severely decrease in intensity pointing to a much lower density of the Cu  $3d_{z^2-r^2}$  state with respect to the Cu  $3d_{x^2-y^2}$  states as observed earlier [8–10, 20]. The shoulder of this Cu L<sub>3</sub> peak represents the itinerant O 2p holes hybridized with Cu 3d and is represented by the transition  $2p \rightarrow 3d^9\bar{L}$ , where  $\bar{L}$  represents a hole in the ligand.



**Figure 5.** The Cu L<sub>3</sub>-edge spectra of as-prepared NdBCO films taken for different orientations of the sample surface and the incident  $E$  vector of the plane polarized synchrotron light beam. The 0° spectra correspond to normal incidence geometry when the  $E$  vector is parallel to the sample surface ( $ab$ -plane) while the 80° spectra represent the case when the  $E$  vector is nearly parallel to the  $c$  axis of the films.

Strangely enough the intensity behaviour of the shoulder, unlike in YBCO (123) or BSCCO (2212) single crystals, instead of decreasing as we go from  $E \parallel ab$  to  $E \parallel c$  orientation increases in the present case. The density of holes ( $n_h$ ) for various orientations estimated quantitatively by the relation,  $n_h = I(3d^9\underline{L})/I(3d^9) + I(3d^9\underline{L})$ , also tends to confirm this behaviour.

Here  $n_h$  is the number of doping holes as a fraction of the total number of holes (doping and covalent) present in that particular orientation and  $\underline{L}$  is a hole in the oxygen ligand orbital. These values are then shown in table 1. The  $3d^9$  and  $3d^9\underline{L}$  contributions were estimated by fittings Gaussians to these Cu L<sub>3</sub> spectra.

There has been a vigorous discussion on the role played by the in-plane and out-of-plane carriers. An important question related to this concerns the role of holes in the apical oxygen. Some theoretical models have been put forth to explain that high temperature superconductivity essentially requires the presence of in-plane orbitals (Cu  $3d_{x^2-y^2}$  and O  $2p_{x,y}$  orbitals) and emphasize that the presence of any out-of-plane orbitals (Cu  $3d_{z^2}$  and O  $2p_z$  orbitals) would be detrimental to the cause of superconductivity in these systems. Some other models essentially require the presence of these out-of-plane orbitals for promoting superconductivity in these cuprates. However, earlier studies have shown that the out-of-plane holes may not be correlated to  $T_c$  in a substantial way [10, 20].

**Table 1.** The density of holes calculated in the NdBCO thin films as a function of the polarization angle from the  $E \parallel ab$  to the  $E \parallel c$  direction.

Sample No.	$T_c$ <sup>a</sup> (K)	0°	15°	45°	60°	75°	80°	90° <sup>b</sup>
NdBCO (2)	92	0.24	0.25	0.30	0.31	0.39	0.40	0.47
NdBCO (3)	90	0.17	0.18 <sup>b</sup>	0.20	0.22	0.35	0.36	0.47
NdBCO (4)	81	0.10	0.11	0.12	0.15	0.17	0.18	0.19
NdBCO (1)	84	0.12	0.22	0.25	0.28	0.38	0.40	0.49

<sup>a</sup>  $T_c$  values have been measured from magnetic susceptibility data.

<sup>b</sup> Values obtained by extrapolation.

Similar behaviour was observed by Srivastava *et al* [10] also in case of some  $c$ -axis oriented superconducting Tl(2212) thin films. However, they do not discuss the significance of this result. Looking at the result from sample 2 and 3 wherein the crystallization and the  $T_c$  are higher and the magnetic moment at lower temperature constant, i.e. the superconducting fraction is higher; contrary to this, films 1 and 4 do not show good crystallization, a lower  $T_c$  and a varying magnetic moment at lower temperature, indicating a relatively lower superconducting fraction. One is therefore tempted to surmise that the higher density of holes in the apical oxygen may be responsible for a higher superconducting fraction by providing a better connectivity amongst different blocks.

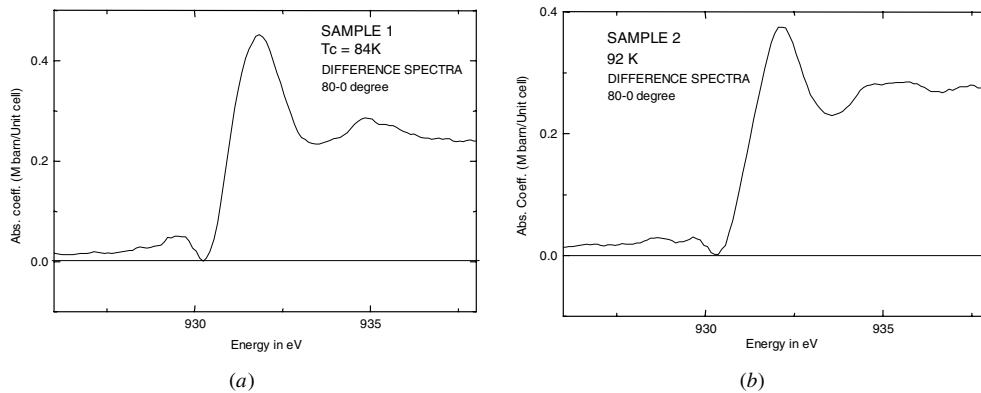
To show this behaviour even more clearly and readily we subtracted the 0 degree spectra from the 80 degree spectra in each case after normalizing the spectra at the peak of the Cu L<sub>3</sub> line. These difference spectra (figures 6(a), (b) shown for two samples only) also bring out the above observation. However, the value of  $T_c$  appears to primarily depend on the density on in-plane holes. These spectra will obviously indicate the density of out-of-plane holes. We can clearly see a substantial intensity of the difference spectra indicating a large number of these holes in all these samples. Therefore this presents a strong challenge to the theoretical models, which are dependent upon the absence of holes with out-of plane symmetry [21, 22]. From the present study we can confidently state that no direct correlation exists between  $T_c$  and out-of-plane covalent and doping hole densities and all the models based on the premise that out-of-plane orbitals, if present, will destroy superconductivity are not dependable.

In the present work we have preferred the bulk sensitive fluorescence yield (FY) mode of detection over the total yield (TY) mode in order to get a reliable estimation of the states. Earlier reports [10] have expressed a fear that some serious complications may arise in the surface sensitive TY technique particularly in case of thin films due to the fact that the surface of the films cannot be cleaned during the experiment as this could cause possible damage to it from sputtering or scraping. Therefore, in the circumstances when the surface of the sample can not be cleaned/cleaved the FY mode is regarded as a better and reliable technique. However, our experience is that the TY technique gives the same results if the surface of the sample is prepared *in situ* by cleaving the sample at a low temperature.

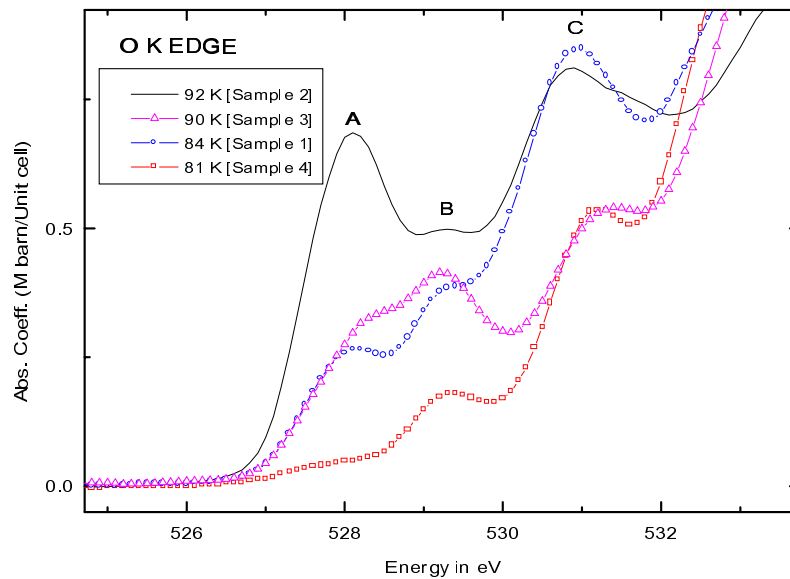
### 3.3. O 1s data

O 1s spectra were also measured in only the  $E \parallel ab$  direction to see the number of itinerant holes in this direction and compare the results with the Cu L<sub>3</sub> data. Figure 7 shows our  $E \parallel ab$  polarized O K-edge spectra recorded in the FY mode for the as-prepared films. In this case the 0 degree spectra will probe the O 2p<sub>x,y</sub> symmetric hole states. The spectra, in general, consist of, amongst others, one important feature—a peak, A, (called a pre-peak) at 528 eV, which corresponds to the transition from the O 1s level to the O 2p impurity states formed in





**Figure 6.** Difference spectra (80 – 0 degree) shown for two only films i.e. sample 1 (84 K) and 2 (92 K) respectively. The huge peak is a clear indication of a large number of out-of-plane holes.

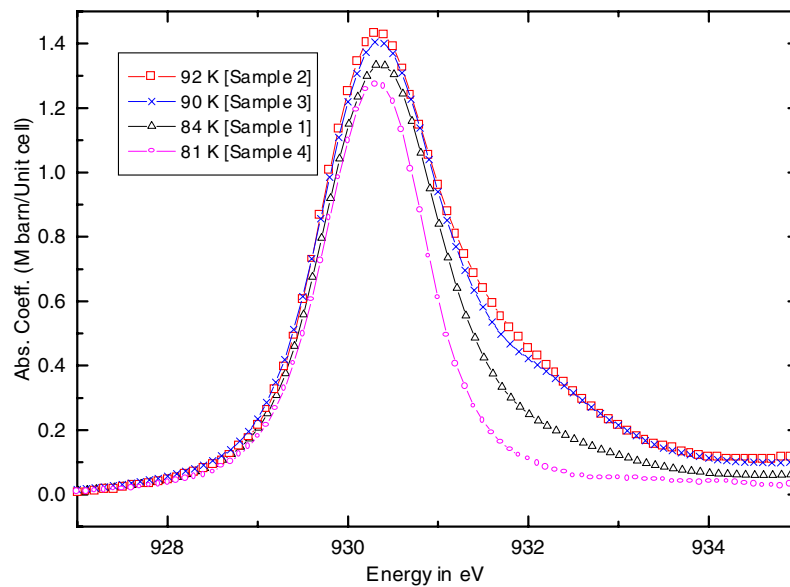


**Figure 7.** The O K-edge spectra taken for the as-prepared NdBCO films for only  $E \parallel ab$  orientation. Peak A indicating the itinerant holes is the most intense for sample 2 (92 K) and very weak for sample 4 (81 K).

the correlation gap as known from the Hubbard model of doped charge transfer insulators [23]. This feature arises due to a doping induced shift of the Fermi level into the valence band, the creation or transfer of holes in the valence band and a transfer of spectral weight from states in the upper Hubbard band to the upper edge of the valence band. The second peak (B) in the spectra arises from transition to the chain ( $O(1)2p_y$ ) and the plane ( $O(3)2p_y$ ) and the third peak (C) appears due to the localized holes. The origin of this peak is composed of states with  $O 2p_{x,y}$  character originating from the upper Hubbard band (UHB) related to  $CuO_2$  and the  $O(2)$  and/or  $O(3)$  atoms [24].

The intensity of the first peak in O K XAS is almost proportional to the amount of hole doping and is known to be a direct measure of the density of the itinerant doping holes [25, 26].

While it appears only as a very weak shoulder in case of the 81 K film, it becomes most pronounced for the 92 K film substantiating our conclusion drawn from our Cu L<sub>3</sub> data that  $T_c$  depends primarily on the density of in-plane holes only. Here we have shown the normalised spectra of Cu L<sub>3</sub> edges at zero degrees ( $E \parallel ab$ ) for all the thin films together for the sake of better comparison with O 1s data (figure 8). We can see that the intensity of the shoulder arising due to  $3d^9\bar{L}$  states (and indicative of itinerant holes) shows the same behaviour as the O 1s spectra for these films, showing a good agreement. The sample with maximum  $T_c$  shows the most intense shoulder while the film of lowest  $T_c$  shows the weakest feature. The high intensity of peak C in case of sample 1 seems to be due to the defects in this non-well-crystallized sample compared with others.



**Figure 8.** The Cu L<sub>3</sub> spectra for  $E \parallel ab$  indicative of itinerant holes, shown for all the films together for comparison with the O K edge spectra. One can notice that the intensity of shoulder  $3d^9\bar{L}$ , indicative of the density of itinerant holes, is a maximum for the sample 2 (92 K) and minimum for the sample 4 (81 K). The inset shows the two Gaussians fitted to the Cu L<sub>3</sub> white line showing the relative  $3d^9$  and  $3d^9\bar{L}$  contributions.

In conclusion, our present study on four NdBCO thin films establishes beyond doubt that  $T_c$  appears to depend primarily on density of in-plane holes and is fairly independent of the out-of-plane holes. On the other hand, the superconducting fraction appears to depend on quality of crystallization and the density of out-of-plane holes, which may themselves be interdependent of each other. The substrate also does not appear to make any difference to either  $T_c$  or the superconducting fraction.

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