# X-ray-diffraction studies of  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$ high-temperature-superconductor thin films

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Single-phase  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films on LaAlO<sub>3</sub>(100) substrates were prepared by molecular-beam deposition and post annealing and they were studied by x-ray-diffraction (XRD) and resistivity measurements. The values of lattice constant  $c<sub>0</sub>$  were determined after correcting for sample displacement errors in the XRD measurements. For an epitaxial  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  film, which showed the onset of superconductivity at 53 K, the lattice constant  $c_0$  was determined as 11.592 $\pm$ 0.018 A. Similar superconducting behavior and same lattice parameter  $c_0$  have been observed for bulk  $Eu_{1,3}Ba_{1,7}Cu_3O_y$  by Iwata et al. [Jpn. J. Appl. Phys. 26, L2049 (1987)]. Bulk  $Dy_1Ba_2Cu_3O_{7-\delta}$  and  $Eu_1Ba_2Cu_3O_{7-\delta}$  have almost the same lattice parameters. Hence the value of x in the  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin film was derived as 0.3 (uncertainty = -0.03, +0.05). The  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film showed complete superconductivity at 89.5 ± 0.5 K (critical transition temperature,  $T_{c,0}$ ). Its lattice constant  $c_0$  was measured as 11.644±0.006 Å. Again, the composition was determined by comparison with lattice parameter  $(c_0)$  and  $T_{c,0}$  of bulk Eu<sub>1.1</sub>Ba<sub>1.9</sub>Cu<sub>3</sub>O<sub>y</sub> as reported by Iwata et al. Since this film is a 90-K superconductor, it is labeled as  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}$  instead of  $Dy_{1,1}Ba_{1,9}Cu_3O_y$  (uncertainty in the value of  $x = -0.03, +0.05$ ).

#### I. INTRODUCTION

There is a great thrust to understand the mechanisms of high-temperature superconductivity in  $RBa_2Cu_3O_{7-\delta}$  $(R = Y, Er, Dy, Gd, Sm, or Eu)$  materials.<sup>1-3</sup> The  $RBa_2Cu_3O_{7-\delta}$  high-temperature superconductors show complete superconductivity at 90 K (for  $\delta \le 0.01$ ). The crystal structure of  $RBa_2Cu_3O_{7-\delta}$  high-temperature superconductors is very interesting. The unit cell consists of three perovskite-type (cubic) cells stacked together along the  $c$  axis.<sup>4-7</sup> There are two Cu-O<sub>2</sub> planar layers and one Cu-0 chain layer in each unit cell which play crucial roles in the high-temperature superconductivity. The 90-K  $RBa_2Cu_3O_{7-\delta}$  superconductor has an orthorhombic crystal structure for which the lattice constants, in case of  $R = Dy$ , are  $a_0 = 3.83$  Å,  $b_0 = 3.88$  Å, and  $c_0 = 11.71 \text{ Å}^4$ . There are two ways to modulate the composition of single-phase  $RBa_2Cu_3O_{7-\delta}$  high-temperature superconductors: (1) The oxygen content can be reduced. In this case, its critical transition temperature  $(T_c)$  drops and the superconductivity is lost as  $\delta$  approaches 1.<sup>3</sup> When the value of  $\delta$  is greater than 0.6, the material adopts a tetragonal crystal structure.<sup>4</sup> (2) Single-phase  $R_{1+x}Ba_{2-x}Cu_3O_y$   $(x=0-0.5)$  materials can be<br>prepared.<sup>4,9,10</sup> When x is greater than zero, some Ba atoms are replaced by smaller  $R$  atoms in the unit cell of  $RBa_2Cu_3O_{7-\delta}$ . Again, this compositional change reduces  $T_c$ . In  $R_{1.5}$ Ba<sub>1.5</sub>Cu<sub>3</sub>O<sub>y</sub>, the superconductivity is completely lost.<sup>9</sup> In fact,  $R_{1.5}Ba_{1.5}Cu_3O_y$  ( $R = La$  and Pr) materials were synthesized and studied before the inven<br>tion of high-T<sub>c</sub> superconductors.<sup>11</sup> tion of high- $T_c$  superconductors.<sup>11</sup>

In this paper, we report on the structural studies of  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  thin films by x-ray diffraction  $(XRD)$ . The values of x in the

single-phase  $Dy_{1+x}Ba_{1-x}Cu_3O_y$  thin films were derived from investigation of transport properties and XRD studies, which are described in the text.

## II. EXPERIMENTAL DETAILS

 $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_r(001)$ thin films were prepared by molecular beam deposition (MBD) and post annealing. The process is described in detail elsewhere.<sup>12</sup> The thin-film deposition was carried out in a miniature ultrahigh-vacuum (UHV) MBD system. Dy,  $BaF_2$ , and Cu were coevaporated from effusion cells in presence of O<sub>2</sub> (oxygen pressure in  $4 \times 10^{-6}$  to  $6 \times 10^{-6}$  Torr range). The films were deposited on LaAlO<sub>3</sub>(100) substrates. The arrival rates (flux) of constituent vapors were determined just before and after thin-film deposition by a movable water-cooled thin-filmthickness monitor (FTM). The fiux of vapors remained constant during deposition (within  $\pm 2\%$ ). The films were post annealed in a particular cycle in wet and dry  $O_2$  to get the oxide.<sup>12,13</sup> The relationships between FTM reading for any constituent vapor and its real flux was determined in separate experiments, but the initial calibrations were crude.<sup>12</sup> Hence, the films deposited with these calibrations were not good. Improvement in calibrations of FTM was necessary to obtain the epitaxial films as described in the next paragraph. The sensitivity of FTM sensor was increased by reading total thickness on the quartz crystal in 200 sec and the thickness value was divided by time to get the (FTM} deposition rate. Note that the deposition rate in  $A$ /sec is related to flux in atoms (or molecules)/ $\text{cm}^2$  sec.

In subsequent MBD experiments, the Dy and  $BaF<sub>2</sub>$  flux were kept constant and the Cu flux was varied. After post annealing, all films were analyzed by XRD using a

 $45$ 



FIG. 1. XRD peak intensity vs Bragg angle for  $2\theta$  in  $6^\circ$ -50° range. (a) XRD spectrum for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film on  $LaAlO<sub>3</sub>(100)$  substrate; (b) XRD spectrum for  $Dy_1$ ,  $Ba_1$ ,  $Cu_3O_v(001)$  thin film on LaAlO<sub>3</sub>(100) substrate. The data for  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  terminates at  $2\theta = 48^\circ$  and, therefore, it does not show the LaAlO<sub>3</sub>(200) peak.  $[(+)$  LaAlO<sub>3</sub>(200) peak;  $(*)$  peaks due to rhombohedral distortion of the substrate or impurities.



FIG. 2. XRD peak intensity vs Bragg angle for  $2\theta$  in 51°-100° range. (a) XRD spectrum for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$ thin film on  $LaAlO<sub>3</sub>(100)$  substrate; (b) XRD spectrum for  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  thin film on LaAlO<sub>3</sub>(100) substrate. [(\*) Peaks due to rhombohedral distortion of the substrate or impurities.]



FIG. 3. Superconducting resistive transition of epitaxial  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film on LaAlO<sub>3</sub>(100) substrate.

Diano diffractometer. A Cu  $K\alpha$  x-ray source (wavelength  $\lambda = 1.542$  Å) was used in the XRD measurements. The resistivity of the films were measured by four-point resistivity measurements or microwave absorption measurements. After deposition of silver pads for resistivity measurements, the films were annealed at 450 °C in dry  $O_2$  for 2 h and slowly furnace cooled. One of the films showed (001) epitaxy without any noticeable contamination and it was labeled as  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$ . Microwave-absorption measurements on this film showed that the film was superconducting. However, the onset of superconductivity was at 53 K and the superconductivity transition width was large. The resistivity data were compared with resistivity versus temperature curves for  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  (x=0-0.5) bulk materials, which has been reported by Iwata et al.<sup>9</sup> From the comparison of transport properties, the  $x$  value in the epitaxial film was estimated to be about 0.3. From these experiments, accurate relationships between FTM reading and flux of constituent vapors were obtained.

 $of$ the improved calibrations Using FTM.  $Dy_1Ba_2Cu_3O_{7-\delta}(001)$  thin films were prepared in subsequent experiments. After careful XRD analysis, which is reported in this paper, it was determined that these films are  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}$ . Figures 1(a) and 2(a) show the XRD spectra for one of the  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$ films. In resistivity measurements, the film showed complete superconductivity at 89.5 $\pm$ 0.5 K (Fig. 3).

# III. DATA ANALYSIS AND RESULTS

The XRD patterns for  $Dy_{1,1}Ba_{1,2}Cu_3O_{7-x}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  thin films are depicted in Figs. 1(a),  $2(a)$ ,  $1(b)$ , and  $2(b)$ . All of the 001 peaks are observable [except for the  $008$  peak in Fig. 2(b)]. In the second XRD scan for  $Dy_{1.3}Ba_{1.7}Cu_3O_y(001)$  [Fig. 2(b)], the x-ray beam intensity was low. The  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$ thicker film also about times is  $1.5$ the For than  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$ film.  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$ , the intensities of 001 peaks

Sample mounting number	<b>XRD</b> data	Standard <b>XRD</b> peak	Measured Bragg angle $2\theta$ (deg)	Calculated Bragg angle $2\theta$ (calc.) (deg)	$-2s/R$	Average $-2s/R$
	Fig. $1(a)$	LaAlO <sub>3</sub> (100)	24.15	23.488	$-0.6772$	
	Fig. $1(a)$	LaAlO <sub>3</sub> (200)	48.66	48.042	$-0.6778$	$-0.6765$
	Fig. $2(a)$	LaAlO <sub>3</sub> (300)	75.80	75.268	$-0.6747$	
$\overline{2}$	Fig. $1(b)$	LaAlO <sub>3</sub> (100)	24.36	23.488	$-0.8923$	$-0.8923$
3	Fig. $2(b)$	LaAlO <sub>3</sub> (300)	75.91	75.268	$-0.8147$	$-0.8147$

TABLE I. Displacement-error correction factor,  $(-2s/R)$ , for x-ray-diffraction data [see Eq. (1)].

compare well with 001 peaks for  $Er_1Ba_2Cu_3O_{7-\delta}(001)$ and  $Nd_1Ba_2Cu_3O_{7-\delta}(001)$  thin films as reported in the and  $Nd_1Ba_2Cu_3O_{7-\delta}(001)$  thin films as reported in the literature.<sup>14,15</sup> The films may not have 100% c-oriented epitaxy but the (100) and (010) peaks, which correspond to  $a$ - and  $b$ -orientations of the film, are undetectable.

To determine the lattice constant  $c_0$  for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  thin films from the XRD data, it was necessary to correct for specimen displacement errors in the XRD measurements. The systematic error correction function for specimen displacement is given by the expression,<sup>16</sup>

$$
\Delta(2\theta) = -2s(\cos\theta/R) , \qquad (1)
$$

where  $s$  is the specimen displacement,  $R$  is the radius of the goniometer circle,  $2\theta$  is the Bragg's angle for difFraction, which is the angle between directions of incident and diffracted x-rays.

In the XRD data for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  thin films, the diffracting atoms are in (001) set of planes. In Figs. <sup>1</sup> and 2, the 001 peak represents first-order diffraction from (001) planes of atoms (order of Bragg reflection,  $n = 1$ ), whereas the peaks labeled as 002, 003, etc., correspond to higherorder diffraction from the (001) planes.

Equation (1) was used to correct for displacement errors. First, the displacement error correction factor,  $(2s/R)$ , was computed using  $\theta$  values for 100, 200, and 300 peaks for the LaAlO<sub>3</sub>(100) substrate  $[a_0=3.79 \text{ Å},$  $\alpha$ =90°4' (Ref. 17)]. The results are shown in Table I. Although the  $LaAlO<sub>3</sub>$  crystal has a slight rhombohedral

distortion at room temperature, which gives some of the peaks marked by an asterisk in Figs. <sup>1</sup> and 2, its structure can be assumed as cubic, since the positions of h00 peaks in XRD spectra for a hypothetical perfectly cubic  $LaAlO<sub>3</sub>$  crystal having same lattice constant will be the same.<sup>18</sup> The values of  $(-2s/R)$  were substituted in Eq. (1) to get the displacement error correction function  $\Delta(2\theta)$ . The measured and corrected values of 2 $\theta$  for 001 peaks are listed in Tables II and III for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$ , respectively. Using the corrected  $2\theta$  values, the lattice constants,  $c_0$ , for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$  thin films were calculated according to the Bragg's law. The results are shown in Tables II and III.

The lattice constant,  $c_0$ , for the  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film is determined as 11.644 $\pm$ 0.006 Å whereas  $c_0$  for the  $Dy_{1,3}Ba_{1,7}Cu_{3}O_{y}(001)$  film is found to be 11.592±0.018 Å.

#### IV. DISCUSSION

The lattice constants for bulk  $Dy_1Ba_2Cu_3O_{7-\delta}$  in its orthorhombic form  $(\delta < 0.01)$  are known to be orthorhomole form  $(0.80, 0.01)$  are known to be<br> $a_0 = 3.83 \pm 0.003$  Å,  $b_0 = 3.885 \pm 0.002$  Å, and  $c_0$  $=11.709\pm0.003$  Å.<sup>4</sup> These values have been derived from precision XRD and neutron-diffraction measurements. Our measured value of 11.644 Å as  $c_0$  for  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film is 0.06 Å smaller than

TABLE II. Calculation of lattice constant  $c_0$  for the Dy<sub>1,1</sub>Ba<sub>1,9</sub>Cu<sub>3</sub>O<sub>7-8</sub>(001) thin film using x-ray-diffraction data [Figs. 1(a) and 2(a)]. The details are described in the text.

<b>XRD</b> peak, hkl	Measured Bragg angle $2\theta$ (deg)	Displacement error correction factor, $-2s/R$	$\Delta(2\theta)$ $(\text{deg})$	Corrected Bragg angle, $2\theta$ (deg)	Lattice constant, $c_0$ (Å)	Mean $c_0$ ( $\check{A}$ ) (standard deviation)
001	8.26	$-0.6765$	$-0.6748$	7.5852	11.656	
002	15.88	$-0.6765$	$-0.6701$	15.2099	11.652	
003	23.57	$-0.6765$	$-0.6623$	22.9077	11.648	
004	31.36	$-0.6765$	$-0.6514$	30.7086	11.647	
005	39.31	$-0.6765$	$-0.6371$	38.6729	11.643	11.644
006	47.45	$-0.6765$	$-0.6194$	46.8306	11.641	$(\pm 0.006)$
007	55.84	$-0.6765$	$-0.5978$	55.2422	11.641	
008	64.57	$-0.6765$	$-0.5720$	63.9980	11.640	
009	73.74	$-0.6765$	$-0.5412$	73.1988	11.638	
0010	83.46	$-0.6765$	$-0.5049$	82.9551	11.641	
0011	94.00	$-0.6765$	$-0.4614$	93.5386	11.640	

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<b>XRD</b> peak, hkl	Measured Bragg angle $2\theta$ (deg)	Displacement error correction factor, $-2s/R$	$\Delta(2\theta)$ $(\text{deg})$	Corrected Bragg angle, $2\theta$ (deg)	Lattice constant, $c_0$ (Å)	Mean $c_0$ ( $\AA$ ) (standard deviation)	
001	8.54	$-0.8923$	$-0.8898$	7.6502	11.557		
002	16.18	$-0.8923$	$-0.8832$	15.2968	11.586		
003	23.90	$-0.8923$	$-0.8730$	23.0270	11.588		
004	31.79	$-0.8923$	$-0.8582$	30.9318	11.565		
005	39.66	$-0.8923$	$-0.8394$	38.8206	11.600	11.592	
006	47.81	$-0.8923$	$-0.8157$	46.9943	11.603	$(\pm 0.018)$	
007	56.19	$-0.8147$	$-0.7187$	55.4713	11.597		
009	74.10	$-0.8147$	$-0.6502$	73.4498	11.604		
0010	83.83	$-0.8147$	$-0.6063$	83.2237	11.610		
0011	94.42	$-0.8147$	$-0.5534$	93.8666	11.609		

TABLE III. Calculation of lattice constant  $c_0$  for the Dy<sub>1.3</sub>Ba<sub>1.7</sub>Cu<sub>3</sub>O<sub>y</sub>(001) thin film using x-ray-diffraction data [Figs. 1(b) and 2(b)]. The details are described in the text.

its bulk value. In case of epitaxial thin films on substrates with small lattice mismatch, there may be some elastic strain in the film if the film is not very thick (say, less than 2000 Å). For c-oriented epitaxial  $Dy_1Ba_2Cu_3O_{7-\delta}$ , its lattice constants  $a_0$  and  $b_0$  are matched with the lattice constants of the substrate (for  $LaAlO<sub>3</sub>$ , its lattice parameter is 3.79 Å). Because of interfacial stresses due to the LaAlO<sub>3</sub>(100) substrate, the lattice parameters  $a_0$  and  $b_0$  of the Dy<sub>1</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub>(001) film can only be compressed. As a result, the lattice constant  $c_0$  of a  $Dy_1Ba_2Cu_3O_{7-\delta}(001)$  thin film can only expand elastically when its unit cell is compressed along the  $x$  and  $y$ axes. Note that the measured  $c_0$  value for the  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film is not larger than its bulk value.

Stresses due to lattice mismatch in the  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films can be described by a two-dimensional case of plane stress.<sup>19</sup> For simplicity, let us assume that the  $Dy_{1+x}Ba_{2-x}Cu_3O_y$  thin film is an isotropic solid in its mechanical properties. In the case of plane stress, stress in the c direction is zero but principal strain in the c direction is nonzero which is described by the relationship

$$
\varepsilon_3 = -\nu(\sigma_a + \sigma_b) \tag{2a}
$$

$$
=-\nu(\sigma_1+\sigma_2),\qquad(2b)
$$

where  $\sigma_a$  and  $\sigma_b$  are stresses along the a and b axes,  $\sigma_1$ and  $\sigma_2$  are principal stresses in the a-b plane, and v is the Poisson's ratio.<sup>19</sup> The strains in a and b directions are  $\varepsilon_a$ and  $\varepsilon_b$ . If the elastic strains in the a and b directions are and  $\epsilon_b$ . It the elastic strains in the *u* and *b* directions are<br>not relieved then  $\epsilon_a = (3.79 - 3.83)/3.83 = -0.01$  and  $\epsilon_b = (3.79 - 3.89)/3.89 = -0.026$ . Since these values of strain are very large and the epitaxial growth temperature is also very high (900°C), the material is bound to undergo plastic deformation. An epitaxial growth process in which the elastic strains due to lattice mismatch are relieved at the film/substrate interface is known as nonpseudomorphic growth. The high annealing temperature (900'C) used in the epitaxial growth of  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films will enhance nonpseudornorphic growth. Hence, the elastic strain will be relieved by plastic deformation at the

 $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)/LaAlO_3(100)$  interface and the lattice mismatch will be accommodated by a network of dislocations at the interface.

To account for residual elastic strains in the films, if present, let us assume that average  $\varepsilon_1 = -0.001$  and  $\varepsilon_2 = -0.002$ . Then  $\varepsilon_3$  is calculated to be 0.0013 (using Poisson's ratio,  $v=0.3$ ). With this estimate, the determined values of  $c_0$  for the  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films will be 0.015 Å larger than  $c_0$  in bulklike  $Dy_{1+x}Ba_{2-x}Cu_3O_y$  materials.

Release of oxygen from orthorhombic  $RBa_2Cu_3O_{7-\delta}$ materials also results in change in lattice constants. Since the sample was kept in air during the XRD measurements, it could lose oxygen. However, it is known that in oxygen deficient  $RBa_2Cu_3O_{7-\delta}$  materials the value of  $c_0$ increases as  $\delta$  approaches 1.<sup>4</sup> Furthermore, for a 90-K  $Dy_1Ba_2Cu_3O_{7-\delta}$  material the value of  $\delta$  must be less than  $0.1$ .<sup>3</sup> Therefore, the small contraction in lattice constant  $c_0$  for the  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film can not be due to loss of oxygen.

Now, we will explain how the lattice constant  $c_0$  for single-phase  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films is related to the value of  $x$  in its composition. Iwata et al. have reported on the lattice constants of orthorhombic (bulk)  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  materials for  $x = 0-0.5$ .<sup>9</sup> In their paper, the values of  $a_0$ ,  $b_0$ , and  $c_0/3$  have been plotted as a function of x for  $Eu_{1+x}Ba_{2-x}Cu_3O_y$ . For  $x = 0$ , their XRD measurements show a  $c_0$  value of 11.70 Å for  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  (i.e.,  $Eu_1Ba_2Cu_3O_7$ ), which is about 0.01 A larger than its bulk value as reported in Ref. 4.

Assuming that the crystal structure and superconducting properties of  $Dy_{1+x}Ba_{2-x}Cu_3O_y$  and  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  are almost identical, our XRD results on lattice parameter  $c_0$  for the epitaxial thin films were compared with  $c_0$  values for  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  $(x=0-0.5)$  as given in Ref. 9. For Eu<sub>1.3</sub>Ba<sub>1.7</sub>Cu<sub>3</sub>O<sub>v</sub> bulk material, the  $c_0$  value is 11.59 $\pm$ 0.01 Å. Hence, the value of x in the  $Dy_{1+x}Ba_{2-x}Cu_3O_y$  thin film, assuming nonpseudomorphic thin film growth, is estimated to be 0.3, since its lattice constant was determined as 11.592 $\pm$ 0.018 Å. Uncertainty in the value of x is  $\pm$ 0.03, which accounts for differences in  $c_0$  for  $Dy_{1+x}Ba_{2-x}Cu_3O_y$  and  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  for same value of x. An additional uncertainty of  $(-0.0, +0.02)$ is added to account for some elastic strains in the film, if present. Hence, the total uncertainty in the value of  $x$  is estimated as  $(-0.03, +0.05)$ .

Now, the  $c_0$  value for the 90-K superconductor thin film is compared with  $c_0$  values for bulk  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  as shown in Ref. 9. It is found that the  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  thin film resembles bulk  $Eu_{1,1}Ba_{1,9}Cu_3O_y$  because their  $c_0$  value are in 11.65  $\pm 0.01$  Å range and, furthermore, the Eu<sub>1,1</sub>Ba<sub>1,9</sub>Cu<sub>3</sub>O<sub>y</sub> material is also a 90-K superconductor. This suggests that the 90-K superconductor thin film is  $\text{Dy}_{1+x} \text{Ba}_{2-x} \text{Cu}_3 \text{O}_y(001)$  in which the value of x is 0.1 as labeled (uncertainty  $=$  -0.03, +0.05). The value of y in the  $Dy_{1,1}Ba_{1,9}Cu_3O_y(001)$  thin film is believed to be  $(7-\delta)$  because a large change in  $\delta$  will reduce  $T_c$  of the material significantly from 90 K, which is not the case. The 90-K superconductivity in the  $Dy_{1,1}Ba_{1,9}Cu_{3}O_{7-\delta}(001)$  thin film is not surprising since many researchers believe that the pure  $RBa_2Cu_3O_7$  hightemperature superconductors have critical transition temperature  $(T_{c,0}^{\dagger})$  above 90 K.<sup>9,20</sup> This does not imply that the 90-K superconductor thin film, which is found to be  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}$ , is a poor material for superconductivity applications because the  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}$  materials show the largest diamagnetic response.

## V. CONCLUSIONS

 $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  and  $Dy_{1,3}Ba_{1,7}Cu_3O_y(001)$ epitaxial thin films on  $LaAlO<sub>3</sub>(100)$  substrates were stud-

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ied by x-ray diffraction. The lattice parameter  $c_0$  for  $Dy_{1,1}Ba_{1,9}Cu_3O_7 - \delta(001)$  thin film was determined as 11.644 $\pm$ 0.006 Å, which is about 0.06 Å shorter than its bulk value. The  $Dy_{1,1}Ba_{1,9}Cu_3O_{7-\delta}(001)$  film shows complete superconductivity at 89.5 $\pm$ 0.5 K (T<sub>c,0</sub>). The  $Dy_1$ ,  $Ba_1$ ,  $Cu_3O_v(001)$  thin film, which shows onset of superconductivity at 53 K, has  $c_0$  value of 11.592 $\pm$ 0.018 A. The estimates of composition of the  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films were made by comparison of critical transition temperature  $(T_c)$  and lattice parameter  $c_0$  of single phase  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$ thin films and  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  bulk materials. Uncertainty in the determined values of  $x$  is estimated to be  $(-0.03, +0.05)$ . The superconducting properties and crystal structure of bulk  $Eu_{1+x}Ba_{2-x}Cu_3O_y$  materials has been reported in Ref. 9. We have shown that x-raydiffraction studies can be used for estimation of composition of single-phase  $Dy_{1+x}Ba_{2-x}Cu_3O_y(001)$  thin films.

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