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ELASTIC PROPERTIES OF Sr SUBSTITUTED $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0, 0.3, 0.6) SUPERCONDUCTORS

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ABSTRACT

Temperature dependent ultrasonic longitudinal and shear velocity measurements in polycrystalline superconducting $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0,0.3,0.6) performed utilizing pulsed echo overlap technique showed step-like anomalies above 200 K in all samples for the longitudinal mode. Sr substitution was observed to soften the elastic anomaly and shifted it from 250 K (*x*=0,0.3*)* to 260 K (*x*=0.6). The results were discussed in terms of ionic radius of Sr and the effect of its substitution on oxygen ordering in the samples. Results of absolute shear and longitudinal velocities, elastic moduli and Debye temperature are also reported.

INTRODUCTION

Ultrasonic studies of superconductors at low temperatures provide useful information on elastic properties of these materials. These studies can provide clues to help better understanding the mechanism of high temperature superconductivity. For RE123 (RE=Rare Earth), there have been numerous ultrasonic studies which showed elastic anomalies around 200 K [1-3 and references therein]. These anomalies are suggested to be related of the oxygen content and are interpreted to be caused by some form of oxygen ordering in the material [1]. However, the reason on why these anomalies occurred is still not fully understood.

On the other hand, the effects of Sr substitutions at the Ba site on superconducting properties of $REBa_2Cu_3O_{7-\delta}$ have been extensively reported [4-8]. Many of these studies found that the Sr substitution at Ba site suppressed T_c. However, so far, to our knowledge, there is no study discussed on why there is nonlinear T_c drop value to the higher Sr substitution. Some suggested it to the planar weight disparity (PWD) phenomenon [12] and some relate it to the hopping interaction between superconducting and normal layer. But, no study has confirmed it yet. Besides, there is still no study which investigates the effect between non linear T_c drop to the elastic anomalies.

This paper reports on the longitudinal and shear wave velocity measurements in superconducting $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0, 0.3, 0.6) and discussed in terms of ionic radius of Sr and the effect of non-linear T_c drop on the elastic anomalies of the samples. The result from electrical resistance (d.c) measurement and X-ray diffraction are also presented.

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EXPERIMENTAL DETAILS

The $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ ($0\leq x\leq 1.0$) samples were prepared from high purity chemical using conventional solid state synthesis by mixing appropriate amounts of Dy_2O_3, $BaCO_3$, $SrCO_3$ and CuO powders with purity ≥99.99%. The materials were ground in an agate mortar then calcined in air at around 900°C for 48 hours with several intermittent grindings and oven cooled. Then the powders were pressed into pellets of 13mm diameter and 2-3 mm thickness under a load of 6-7 tons. The pellets were then sintered at 910°C for 24 hours and slow cooled to room temperature at 24°C/h.

Electrical resistance (d.c) measurements of the samples were carried out using the four point probe technique with the silver paste contacts in conjunction with Janis cryostat model CCS-350ST and the temperature detected by the LakeShore temperature controller model 330 with GaAlAs diode sensor with accuracy of 50 mK. The powder X-ray Diffraction (XRD) analysis by Cu-K_α radiation using Rigaku model D/MAX 2000 PC was used to confirm the sample structure

The ultrasonic velocity measurements were carried out using a Matec 7700 which utilized the pulse echo overlap technique. The quartz transducer was bonded on the polished samples surface using Nonaq stopcock grease. Sound velocity was propagated along the direction of pressing using x-cut (longitudinal) and y-cut (shear) transducer with a carrier frequency of 9 MHz. These measurements were performed in a Janis instrument liquid nitrogen cryostat model VNF 100T and the temperature was changed at a rate of 1K/min during warming and cooling.

RESULTS AND DISCUSSION

The XRD patterns (Figure 1) showed that all of the samples consists of single phased 123 orthorhombic structure with space group Pmmm. Figure 2 showed the results from four point probe electrical resistivity measurements of $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$. We can see that the Sr substitution only slightly affect the metallic normal state behaviour of $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$. Table 1 showed the value of room temperature resistivity, T_c onset, T_c zero and lattice parameters for $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$.

The absolute velocities of longitudinal and shear velocities at 80K, longitudinal modulus, shear modulus, bulk modulus, young modulus, density and Debye temperature are tabulated in Table 2. The polycrystalline ceramics material having two independent elastic stiffness moduli C_L ($=\rho v_l^2$) and μ ($=\rho v_s^2$) where C_L is the longitudinal modulus, μ is the shear modulus, ρ is the density, v_l is the longitudinal velocity and v_s is the shear velocity. The related elastic stiffness moduli, bulk modulus (B) and Young modulus (Y) is determined using the measured ultrasonic wave velocity values. The acoustic Debye temperature (θ_D) was calculated using the standard formula:

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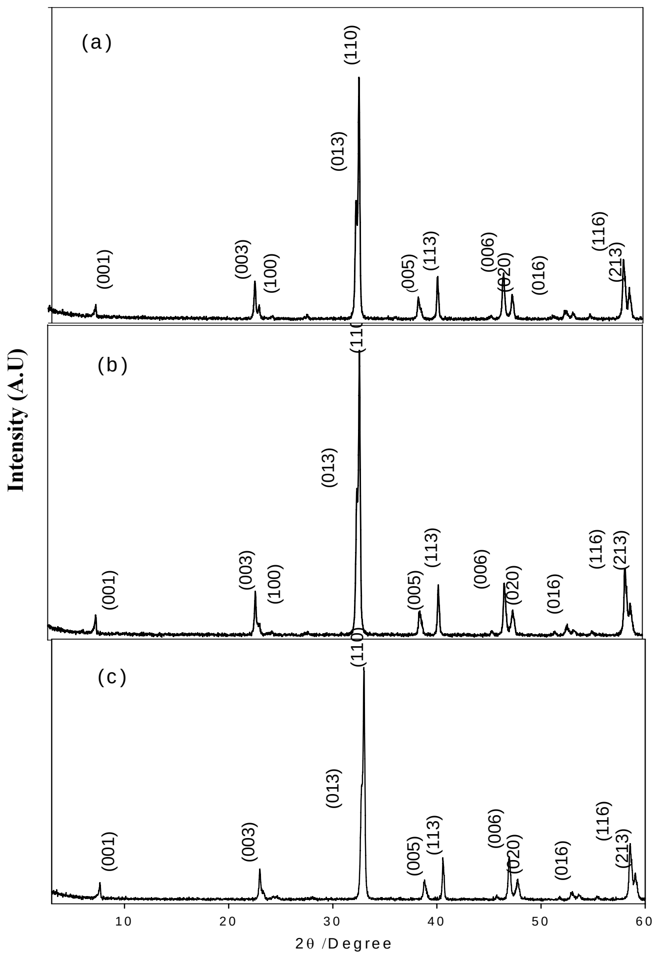
Table 1: $T_{c\ onset}$, $T_{c\ zero}$, room temperature resistivity and lattice parameter for $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0,0.3,0.6)

Sample	$T_{c\ zero}$(K)	$T_{c\ Onset}$ (K)	Resistivity (300K) mΩ cm	Lattice Parameters		
				a (Å)	b(Å)	c (Å)
$DyBa_2Cu_3O_{7-\delta}$	89.7	95.0	5.8	3.870	3.888	11.702
$DyBa_{1.8}Sr_{0.3}Cu_3O_{7-\delta}$	80.1	87.2	7.3	3.839	3.877	11.667
$DyBa_{1.8}Sr_{0.6}Cu_3O_{7-\delta}$	61.1	73.1	4.3	3.820	3.876	11.619



(a)

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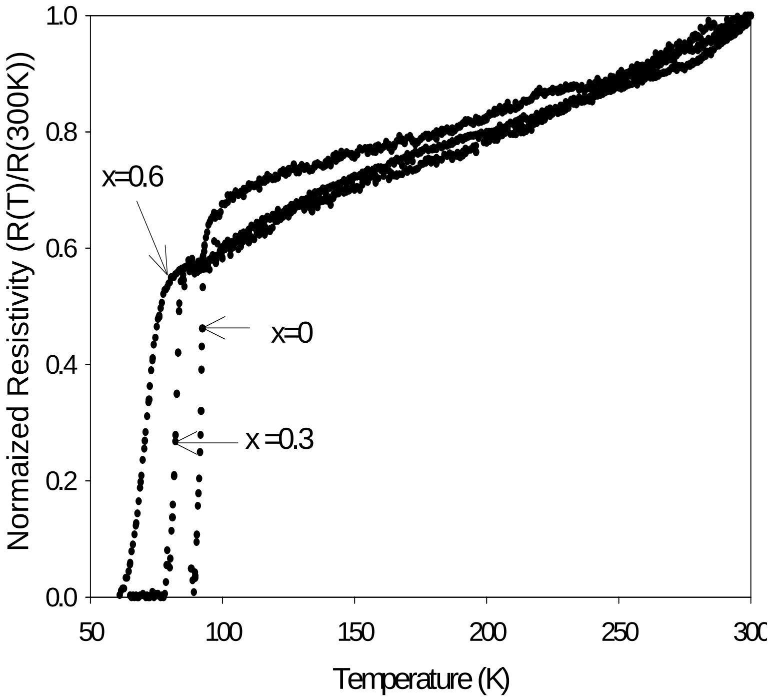

(b)

Figure 1: X-ray Powder diffractograms of superconducting a) $DyBa_2Cu_3O_{7-\delta}$ b) $DyBa_{1.8}Sr_{0.3}Cu_3O_{7-\delta}$ c) $DyBa_{1.8}Sr_{0.6}Cu_3O_{7-\delta}$ Figure 2: Resistance versus temperature curve of $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$, a)x=0, b)x= 0.3, c)x=0.6

$$\theta_D = \left(\frac{h}{k}\right)\left(\frac{3N}{4\pi V}\right)^{1/3} v_m$$

where h is the Planck constant, k is the Boltzman constant, N is the number of mass point, V is the atomic volume and v_m is the mean velocity. The mean velocity is given by

$$\frac{3}{v_m^{\ 3}} = \frac{1}{v_l^{\ 3}} + \frac{2}{v_s^{\ 3}}$$

Table 2: Density, longitudinal velocity (v_l), longitudinal velocity (v_s), longitudinal modulus (C_L), shear modulus (μ), Bulk modulus (B), Young Modulus (Y), and Debye temperature θ_D measured at 80K of a) $DyBa_2Cu_3O_{7-\delta}$ b) $DyBa_{1.8}Sr_{0.3}Cu_3O_{7-\delta}$ c) $DyBa_{1.8}Sr_{0.6}Cu_3O_{7-\delta}$

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Sample	Density (gcm^{-3})	V_t (ms^{-1})	V_s (ms^{-1})	C_L (GPa)	μ (GPa)	B (GPa)	Y (GPa)	θ_{Dmin} (GPa)	θ_{Dmax} (GPa)
$DyBa_2Cu_3O_{7-\delta}$	6.165	4605	2695	130.7	44.8	71.0	111.0	372	373
$DyBa_{1.8}Sr_{0.3}Cu_3O_{7-\delta}$	6.166	5023	2716	155.6	45.5	94.9	117.7	379	380
$DyBa_{1.8}Sr_{0.6}Cu_3O_{7-\delta}$	6.022	5096	2852	156.4	49.0	91.1	124.7	398	399

Substitution of Sr in $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ caused an increase in both longitudinal and shear absolute velocities (v_l and v_s) respectively, and related elastic moduli (Table 2). The Debye temperature θ_D of superconducting $DyBa_2Cu_3O_{7-\delta}$ is 374 K and is comparable to $REBa_2Cu_3O_{7-\delta}$ (RE=Er) superconductors [11]. Substitution of Sr for Ba in $DyBa_{1.7}Sr_{0.3}Cu_3O_{7-\delta}$ and $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ suppressed T_c zero (Figure 2) and increased the θ_D (Table 2) value. The decrease in T_c following an increase in θ_D (with increasing Sr) content is in contrast to the standard BCS theory in the weak coupling limit where $T_c \sim \vartheta_D e^{-1/\lambda}$ [5]. The theory predicts that an increase in θ_D would accompany an increase in T_c and this is not observed in these Sr-substituted material. However, the coupling constant (λ) of the samples may not be constant when Sr is substituted.

Temperature dependence of longitudinal velocities of $DyBa_2Cu_3O_{7-\delta}$, $DyBa_{1.7}Sr_{0.3}Cu_3O_{7-\delta}$ and $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ (Figure 3) showed a change of 1.7%, 1.9% and 1.1% respectively. A change in slope was observed around 240K for $DyBa_2Cu_3O_{7-\delta}$ and $DyBa_{1.7}Sr_{0.3}Cu_3O_7$-δ indicating pronounced lattice stiffening tendency. However, the elastic anomaly for $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ was shifted to 260 K. The higher substitution reduced the change in gradient from 0.0078%/K for $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0), to 0.0043 %/K for $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0.3) and the highest substitution of Sr, $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (x=0.6) the slope gradient suppressed to 0.0023 %/K. For the shear mode (Figure 4), the velocity change were 1.28% and 1.09% for $DyBa_2Cu_3O_{7-\delta}$ and $DyBa_{1.7}Sr_{0.3}Cu_3O_{7-\delta}$ respectively. The smallest percentage different in shear velocity was observed in $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ with 0.75%. No velocity anomaly was observed for this mode.

The increase in longitudinal and shear velocity for $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ (Table 2) may be due to the cell volume change as a result of substitution of the smaller Sr^{2+} (ionic radius: 1.25Å) for Ba^{2+} (ionic radius:1.42 Å). The substitution may also have altered interatomic distances and bonding lengths in $DyBa_2Cu_3O_{7-\delta}$ and this volume change was detected by longitudinal phonon [1].

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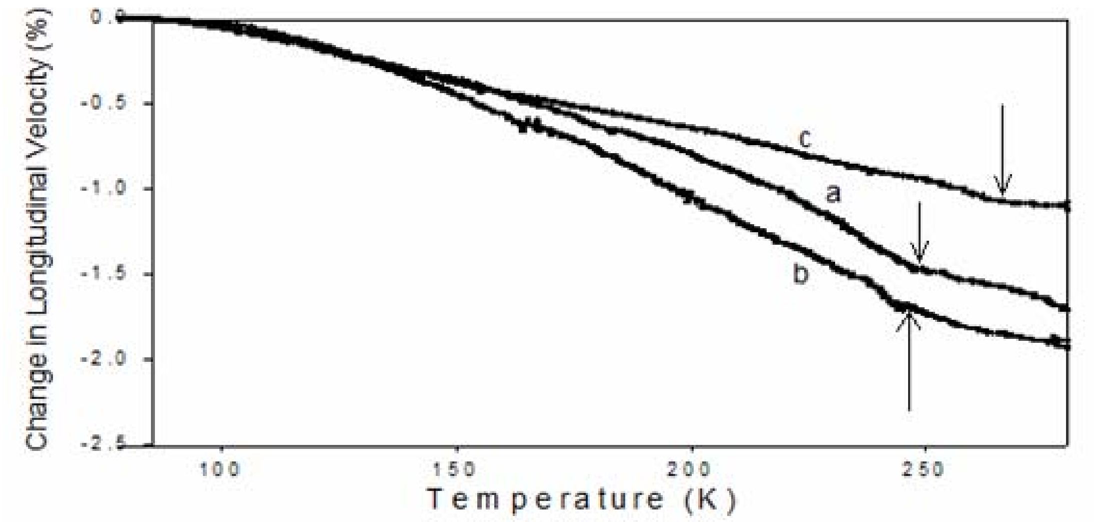

Figure 3: Temperature dependence of longitudinal velocity for a)$DyBa_2Cu_3O_{7-\delta}$ b) $DyBa_{1.7}Sr_{0.3}Cu_3O_{7-\delta}$ c) $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ (The solid arrow indicating the anomaly)

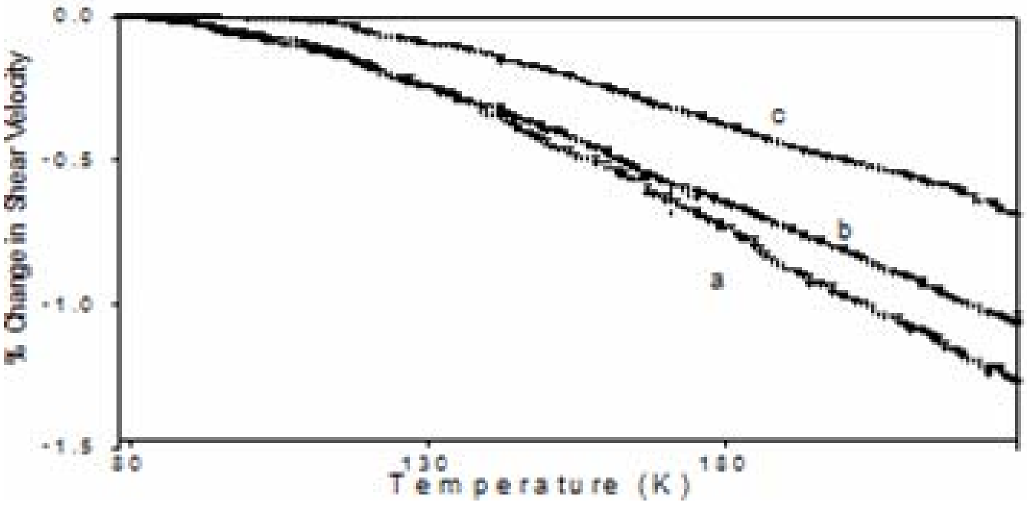

Figure 4: Temperature dependence of shear velocity for a)$DyBa_2Cu_3O_{7-\delta}$ b) $DyBa_{1.7}Sr_{0.3}Cu_3O_{7-\delta}$ c) $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$

From the graph (Figure 3), we found that Sr substitution at Ba site have softened the anomalies. There are many studies reporting that the elastic anomalies were affected by the oxygen content in superconductors [1, 3, 13, 14 and references therein]. However the previous Retvield analysis on samples using similar preparation method had reported [4] that oxygen content was fairly maintained, after Sr was substituted into the Ba site in RE123. So, assuming that O_2 content of the samples are maintained, then possibly, the anomaly is not due to differences in O_2 content. Therefore, we expect that the parameter that governing the suppression of elastic anomalies in our sample was the

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lack of oxygen ordering in the CuO plane rather than the lack of oxygen content. Besides, there were also some reports that proposed the anomalies were influenced by oxygen ordering [3,15]. However, the shifted anomaly for $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ might bring a new clue which indicates the low T_c zero value of superconductor as the T_c zero for $DyBa_{1.4}Sr_{0.6}Cu_3O_{7-\delta}$ is 61K which is much lower than $DyBa_2Cu_3O_{7-\delta}$ and $DyBa_{1.7}Sr_{0.3}Cu_3O_{7-\delta}$.

CONCLUSIONS

The substitution of Sr for Ba site in $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$ decreased $T_{c\ zero}$ but enhanced the elastic properties of $DyBa_{2-x}Sr_xCu_3O_{7-\delta}$. The anomaly was strongly found in Sr-free substituted sample but at higher substitution level at Ba-site, the anomaly was softened and shifted to higher temperature.

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