

## Study of ultrasonic properties of orthorhombic and tetragonal RBaCuO (R = Y, Gd, Dy) compound

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**Abstract** : RBaCuO (R = Y, Gd, Dy) compound in the orthorhombic form is a high temperature superconductor. This compound was prepared in orthorhombic as well as in tetragonal form. Beside resistivity measurements, ultrasonic measurements were made using pulse-echo-overlap method at 5 MHz in the system up to liquid nitrogen temperature. Results are discussed as a function of component R, oxygen content and temperature.

**Keywords** : Ultrasonic properties, high temperature superconductors.

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### 1. Introduction

Since the discovery of high temperature superconductivity in layered cuprates, there have been many reports of measurements of their ultrasonic parameters. The motivation for these experiments has been both to understand the thermodynamics of the superconducting transition, and to search for structural phase transitions [1-4]. Regarding the latter, most people agree so far as the presence of one or more structural transitions are concerned [5,6], although the relationship of these transitions to the superconductivity is not clear.

### 2. Experimental details

In this paper, we report the ultrasonic parameters in RBaCuO (R = Y, Gd or Dy) compounds in non-superconducting tetragonal phase (series-1), in orthorhombic superconducting phase (series-3) and in mixed phase (series-2).

In series-1, the stoichiometric amounts of doubly calcinated mixture of yttrium oxide/gadolinium oxide/dysprosium oxide, barium carbonate and copper(II) oxide were sintered in oxygen atmosphere and then quenched to prevent tetragonal compound to transform into

orthorhombic superconducting compound. The XRD results also showed the presence of tetragonal phase as major constituent. In this case, the resistivity measurements show no superconducting transition till liquid nitrogen temperature.

In series-2, barium carbonate was replaced by barium peroxide. The calcinated powder was directly pressed in the shape of right circular cylinders and sintered in air atmosphere and slow cooled at the rate of 60°C/hour in air. As barium peroxide has excess oxygen which is quite sufficient to oxygenate RBaCuO compound to raise its oxygen content (7-x) above 6.5, but still not sufficient enough to change the whole of material into orthorhombic form. Thus, as a result the final material contained both orthorhombic as well as tetragonal phases. This was also evident from XRD studies that showed mixed composition with orthorhombic as the main phase. In case of yttrium compound, the  $T_c$  (onset) was observed at 90 K but resistance does not become zero even at liquid nitrogen temperature (though resistance drop of about 30 times was observed). For gadolinium compound,  $T_c$  (onset) was observed at 86 K and upto 77 K, resistance drop of 27% was observed. In case of dysprosium compound,  $T_c$

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(onset) was observed at 90 K and resistance drop of about 58% is observed up till liquid nitrogen temperature.

The series-3 was prepared by sintering series-2 samples in oxygen atmosphere, followed by slow cooling in oxygen atmosphere at the rate of 100°C/hour to facilitate oxygen pick up and conversion of tetragonal phase into orthorhombic phase. XRD studies also showed orthorhombic phase as the major phase. In case of yttrium compound, the  $T_c$  (onset) was observed at 92 K and  $T_c(0)$  was observed at 78 K. For gadolinium compound  $T_c$  (onset) was observed at 90 K and  $T_c(0)$  was observed at 77 K, whereas in case of dysprosium compound  $T_c$  (onset) was observed at 90 K and  $T_c(0)$  was observed at 78 K.

The ultrasonic measurements were made at 5 MHz by using pulse echo overlap technique. The two basic parameters measured were longitudinal velocity ( $V_L$ ) and transverse velocity ( $V_S$ ). Longitudinal velocity was measured from 270 K to liquid nitrogen temperature, however transverse velocity could not be measured below 145 K due to coupling problems. Bulk modulus was calculated from measured longitudinal velocity, transverse velocities and density using standard relation [7].

$$K = (V_L^2 - 4V_S^2/3)d.$$

Measured ultrasonic velocities at 270 K for various RBaCuO compounds are listed in the Table 1.

**Table 1.** Ultrasonic velocities for various RBaCuO compounds Longitudinal velocity ( $V_L$ ) m/s at 270 K.

Series	Compound		
	Yttrium	Gadolinium	Dysprosium
Series-1	3848.3	3808.6	3785.1
Series-2	3872.4	3842.9	3992.4
Series-3	4061.6	3984.1	4040.5

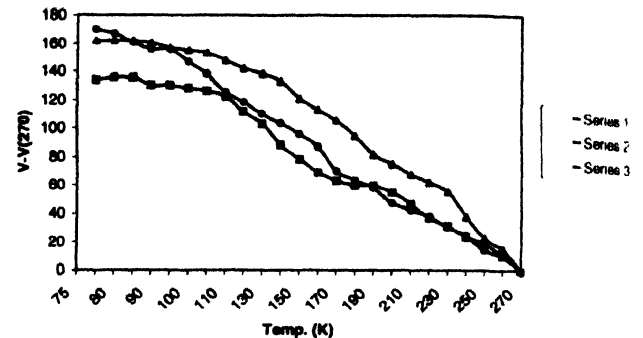
  

Series	Compound		
	Yttrium	Gadolinium	Dysprosium
Series-1	2298.9	2273.2	2360.6
Series-2	2307.5	2285.4	2341.7
Series-3	2417.4	2380.0	2420.5

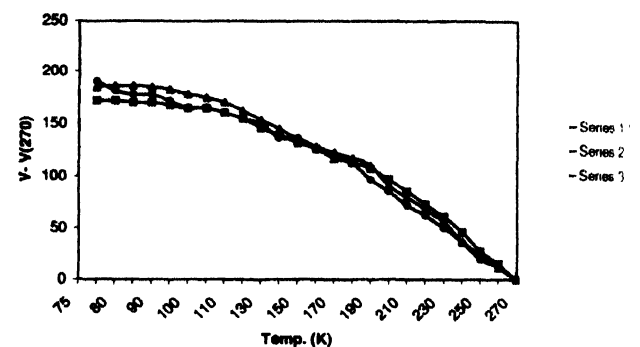
### 3. Results and discussion

In all the series, the ultrasonic velocity showed increase with increase of orthorhombicity. Longitudinal velocity (Figures 1–3) showed continuous increase with decrease of temperature. In general longitudinal velocity measurements show anomalous behaviour in three regions. These were observed in the temperature ranges 80–120 K,

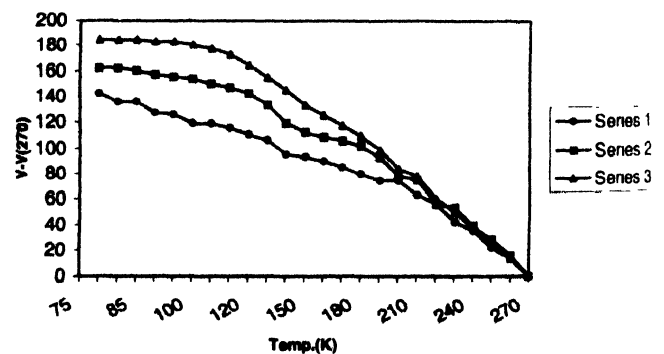
135–190 K and 205–255 K. In series-3, softening of longitudinal velocity was observed near  $T_c$ . The longitudinal velocities have minimum values for the series-1 while maximum values were observed in series-3. The



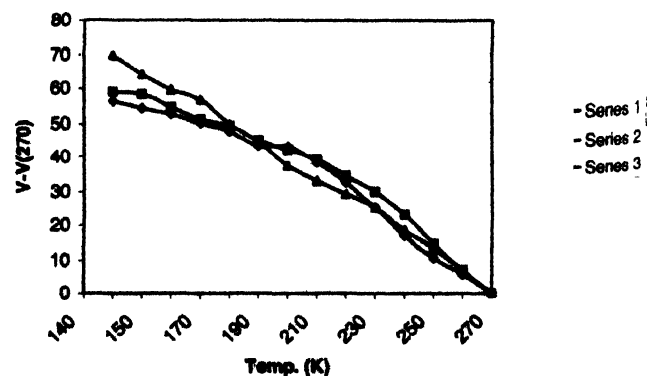
**Figure 1.** Relative longitudinal velocity (m/sec) vs temperature (K) for yttrium compounds.



**Figure 2.** Relative longitudinal velocity (m/sec) vs temperature (K) for gadolinium compounds.



**Figure 3.** Relative longitudinal velocity (m/sec) vs temperature (K) for dysprosium compounds.



**Figure 4.** Relative transverse velocity (m/sec) vs temperature (K) for yttrium compounds.

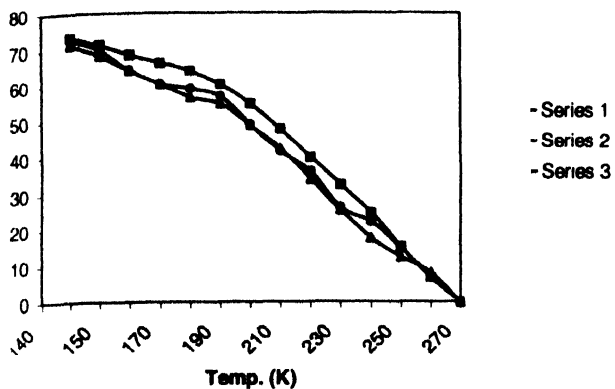


Figure 5. Relative transverse velocity (m/sec) vs temperature (K) for gadolinium compounds.

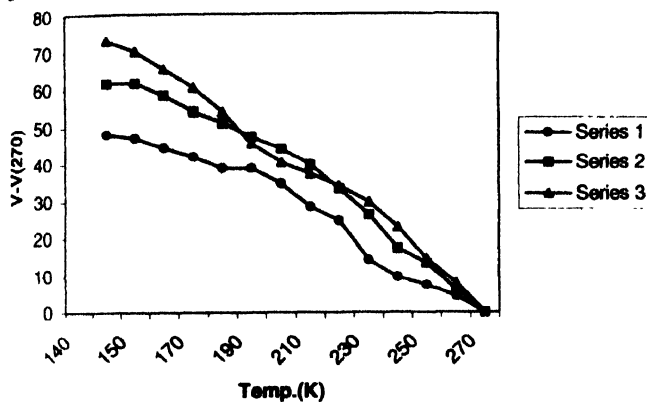


Figure 6. Relative transverse velocity (m/sec) vs temperature (K) for dysprosium compounds.

temperature ranges for the anomalous regions also reduced with the increase of orthorhombicity. These two effects could be due to decreased grain sizes and improved microstructure.

Transverse velocity (Figures 4–6) also showed continuous increase with decrease of temperature in the temperature range studied. For all the samples, besides a few differences, the shape of the curve is almost the same. Transverse velocity also shows increase with increase of orthorhombicity of the sample. These curves in general showed two regions of anomalous behaviour, one in the range 180–190 K and other in the range 210–240 K. For all the samples, the value of bulk modulus (Figures 7–9) also shows anomalous behaviour in these temperature ranges.

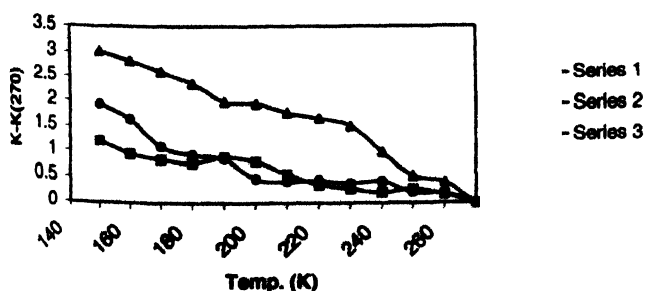


Figure 7. Relative bulk modulus (GPa) vs temperature (K) for yttrium compounds.

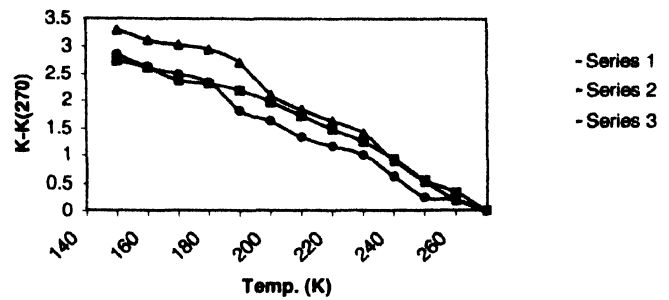


Figure 8. Relative bulk modulus (GPa) vs temperature (K) for gadolinium compounds.

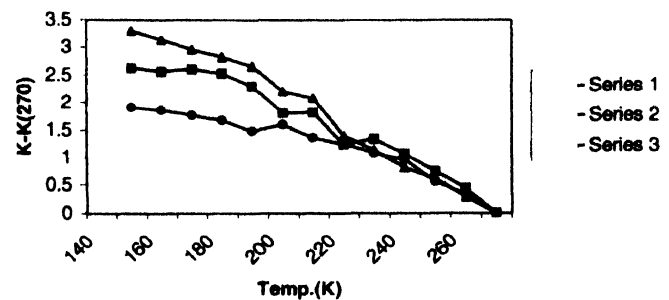


Figure 9. Relative bulk modulus (GPa) vs temperature (K) for dysprosium compounds.

These various maximas or anomalies are believed to be caused by strong lattice anharmonocities or lattice instabilities associated with phase transitions. One (or more) of these peaks may be due to Debye relaxation process. With the variation of oxygen content from six to seven, a distinct increase in the *c*-axis length near the stoichiometry of 6.4 oxygens (per formula unit) was observed [3,8,9]. Since variation of oxygen content results in the change of copper-oxygen coordination number. This change can also contribute to the change of velocity among the various series. Perhaps, these metastable microstructural states are responsible for the observed anomalies in the ultrasonic velocities and bulk modulus. There is likelihood that these microscopic transitions could be masked by other factors like anisotropy, twinning, porosity, grain size, density or technique of material preparation.

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