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Anomalous behaviour of sound velocity in the high T_c superconductor $YBa_2Cu_3O_7$

R SRINIVASAN, V RAMACHANDRAN, A T SESHADRI and G ANANDA RAMADASS

Department of Physics, Indian Institute of Technology, Madras 600 036, India

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Abstract. The longitudinal acoustic wave velocity in $YBa_2Cu_3O_7$ has been measured in the range 300 to 85 K using the ultrasonic pulse superposition technique. The sound velocity shows a steep drop in its value at 260 K and decreases continuously till the superconducting transition temperature. Below T_c the sound velocity increases steeply. A hysteresis is seen when the experiments are performed during a cooling-warming cycle. The results are compared with recent measurements by Ewert *et al* on the samples with the same chemical composition.

Keywords. High temperature superconductivity; anomalous behaviour; sound velocity; $YBa_2Cu_3O_7$.

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1. Introduction

The discovery of superconductivity in strontium-doped lanthanum copper oxide and in yttrium barium copper oxide has been followed by a study of various physical properties of these materials. The velocity of sound in $La_{1.85}Sr_{0.15}Cu_2O_4$ has been studied on sintered pellets by Fossheim *et al* (1987) and Luthi *et al* (1987). Though there is a difference in the values of the longitudinal sound velocity measured by the two groups, presumably arising from the nature of preparation of the pellets and their porosity, the general trend of the temperature variation of the acoustic wave velocity is similar. The sound velocity shows an abrupt drop in value as the pellet was cooled below 250 K. Below 200 K the drop in velocity is gradual. Fossheim *et al* (1987) found a hardening of the acoustic wave velocity below 90 K while such a trend is not seen in the data of Luthi *et al* (1987). Both these authors do not mention whether they looked for any hysteresis in the acoustic velocity when the temperature of the sample was cycled from room temperature to the transition temperature and back. The attenuation of the longitudinal wave was also measured by Fossheim *et al* (1987) and the attenuation was found to rise rapidly below 250 K as the specimen was cooled. There was a broad maximum in the attenuation and a drop in attenuation was seen below 90 K. These authors believe that the softening of the acoustic mode below 250 K indicates a tendency towards structural instability which persists over a wide temperature range. We have studied longitudinal sound velocity in $YBa_2Cu_3O_7$. When we completed our measurements, we received a preprint of a paper by Ewert *et al* (1987) on a similar work. The present paper reports our results and compares them with

those of Ewert *et al* who made measurements on two separate samples (hereafter these samples will be referred to as E_1 and E_2).

2. Experimental details

The compound $YBa_2Cu_3O_7$ was prepared by high temperature solid state reaction of a mixture of high purity Y_2O_3, $BaCO_3$ and CuO in stoichiometric proportions (950 C, 24 h in air, two repeat grindings and heating, and final oxygen treatment at 900 C for 24 h followed by slow cooling to room temperature). X-ray powder diffraction photographs showed that the material was single phase $YBa_2Cu_3O_7$. The transition temperature at which resistance became zero was found to be 91 K. Sintered pellets of 1·2 cm diameter and about 1·8 cm height were prepared under a pressure of 4·2 tonnes/cm^2. The density of the pellets was 63% of the theoretical density.

The longitudinal ultrasonic sound velocity in the pellet was measured at a frequency of 10 MHz using the pulse superposition technique with an ultrasonic interferometer supplied by Messrs Systems Dimensions of Bangalore. The velocity

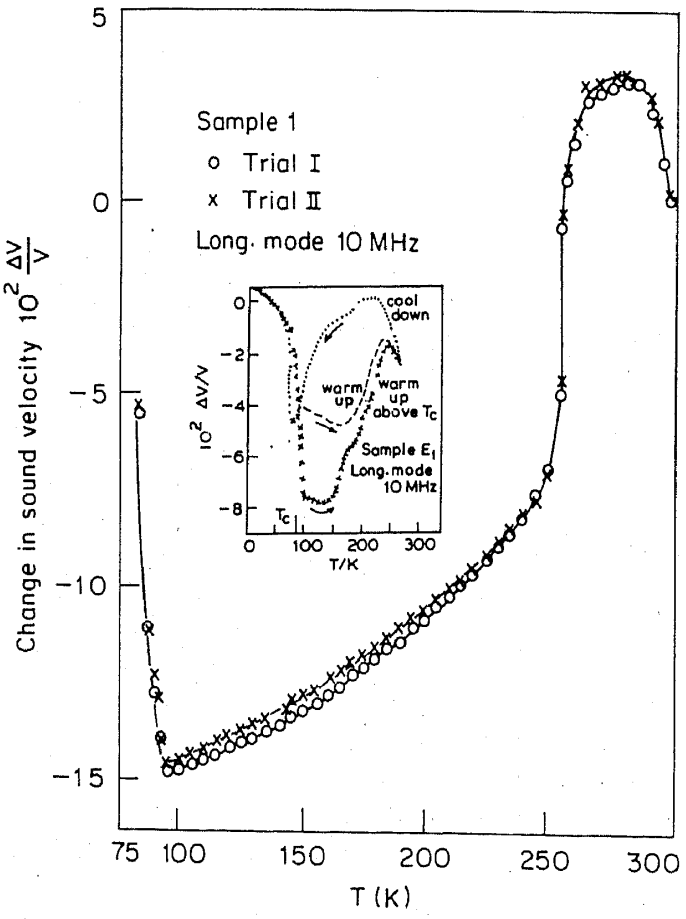

Figure 1(a). Change in sound velocity as a function of temperature for sample (1) of $YBa_2Cu_3O_7$ in I and II trials. The inset shows change in sound velocity observed by Ewert *et al.*

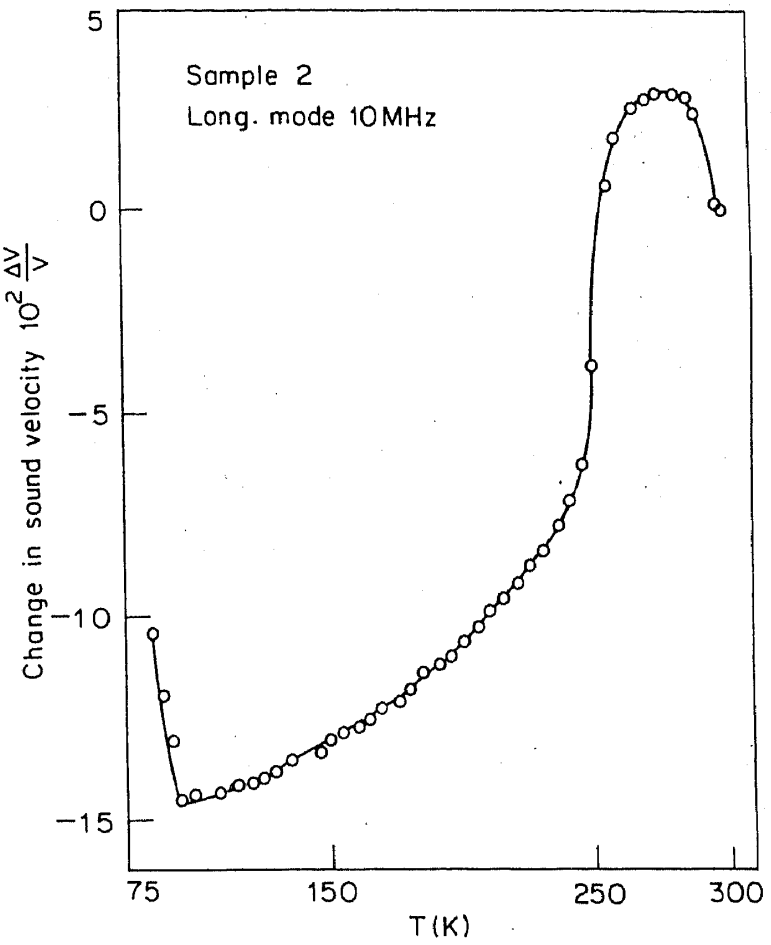

Figure 1(b). Change in sound velocity as a function of temperature for sample (2) of $YBa_2Cu_3O_7$.

was measured in the range of temperatures 85–298 K. The velocity measurement was accurate to 10 m/sec and the temperature measurement to 1 K. Four echoes of the pulse could be clearly seen and the attenuation was determined from the amplitudes of successive echoes.

Measurements were made on two of our pellets. At first the sound velocity and attenuation measurements were made while the sample was cooled from room temperature. On receiving the preprint of Ewert *et al* (1987) sound velocity measurements were made by us on one of our pellets as it was warmed up after cooling it to 85 K, to see whether any hysteresis effects were present. We have observed hysteresis in the case of our sample 1 which is shown in figure 2.

3. Results

The room temperature value of the sound velocity measured on the two samples agreed well and was only 2200 m/sec. This must be compared with the value of 5000 m/sec in a sample having a density of 92% of the theoretical density and 4400 m/sec in a sample having a density 78% of the theoretical density, of Ewert *et al.*

The low value of the velocity in our samples may partially be due to the lower density and higher porosity of the samples studied. It is also possible that at the lower density the grains may be larger and during pelletization they might have taken a preferred orientation so that the contribution to the sound velocity comes mostly from grains in which the direction of propagation is a direction of low acoustic velocity.

Figures 1(a) and (b) show plots of $\Delta V/V_{RT} \times 10^2$, where $\Delta V = V(T) - V_{RT}$ is the change in the value of the velocity from its room temperature value. The data points for two different runs on the same sample as well as runs on the two different pellets are in good agreement with one another. Initially the sound velocity increases as the specimen is cooled. However at 260 K there is a steep drop in the velocity from 2225 m/s at 260 K to 1975 m/s at 220 K. This is followed by a more gradual fall in the velocity to 95 K. The velocity then shows a very steep increase from 1865 m/s at 95 K to 2075 m/s at 85 K. This is the behaviour observed when the specimen is cooled from room temperature.

The precipitous drop in velocity between 260 and 220 K is similar to what has been observed in strontium doped lanthanum copper oxide by Fossheim *et al* (1987). Ewert *et al* (1987) observed a drop in the velocity in their sample E_2 (density 78% of theoretical density) when the specimen was cooled from 250 K and also the steep rise in the velocity below the transition temperature. The inset in figure 1(a) shows the change $\Delta V/V$ (200 K) observed by them in their sample E_1. The shape of their curve is different from the shape of our curve. The magnitude of the change in velocity in our sample is nearly two to three times the value found by them. However the steep rise in the sound velocity below the transition temperature is also seen by them.

Ewert *et al* (1987) had seen a hysteresis effect in their sample E_1 on heating and cooling the sample. To see if a similar hysteresis effect was present in our sample, we cooled our sample to 85 K and measured the velocity as the sample was warmed up. Figure 2 shows the plot of $\Delta V/V$ for both the cooling and warming regimes. We do observe a hysteresis, the velocity on warming being lower than the velocity on cooling. Ewert *et al* (1987) observed a similar behaviour in their sample E_1. However we see the hysteresis only at temperatures around 95 to about 170 K. Beyond 170 K the warming and cooling curves are in agreement. Also the maximum difference in $\Delta V/V_{RT}$ is only 3·5%. On the other hand in sample E_1, Ewert *et al* find the hysteresis all the way up to room temperature and the maximum difference in $\Delta V/V$ between the cooling and warming regimes is about 8%.

Ewert *et al* (1987) measured the sound velocity on a second sample which was made of finer grains and had a density approximately 92% of the theoretical density. In this sample they found a monotonic increase in the sound velocity as the temperature was decreased through the transition. They did not observe the anomalous behaviour found in their sample E_1.

This can be explained if we assume that the sound velocity in a narrow range of directions of propagation around a given crystallographic direction softens quite dramatically as the transition temperature is approached and also hardens rapidly as the material becomes super-conducting. This crystallographic direction may itself be a direction of low velocity of propagation in the normal metal. On the other hand, the sound velocity in other directions in the crystal increases with decreasing temperature as in any normal material. If the pelletization conditions are such that most of the grains are so oriented that the direction of the propagation of the sound makes a small angle with the direction of the soft acoustic mode, then not only the sound velocity but

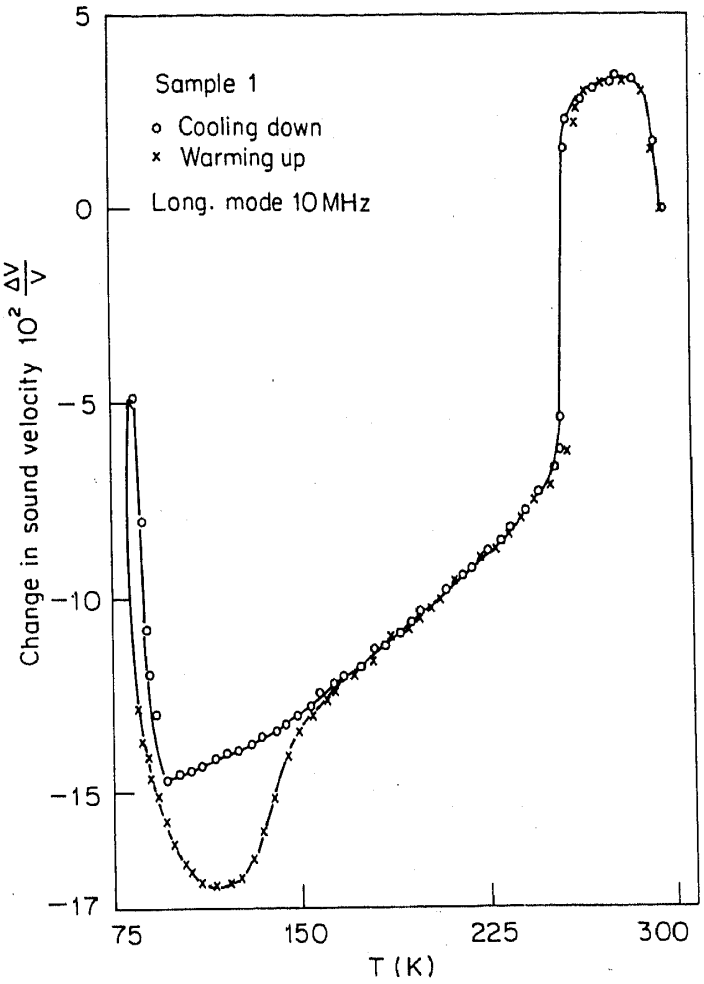

Figure 2. Change in sound velocity of sample (1) as a function of temperature while cooling down and warming up.

also the change in the sound velocity is large in such a pellet. The hysteresis behaviour may be compounded by the porosity of the pellet. On the other hand if the pellet is made of homogeneous grains of small size then a more perfect randomization of crystallographic orientation is achieved in such a pellet. In such a pellet the sound velocity is an average over a large number of directions and so the reduction in the wave velocity of the mode in a narrow range of directions is smoothed out by the normal behaviour in the other directions. We believe that this could be the explanation for the abrupt hardening of the sound velocity seen in our sample and in sample E_1 of Ewert *et al* while no such behaviour is observed in their more homogeneous sample E_2. If this explanation is correct then there is a considerable softening of the acoustic mode in one preferred crystallographic direction. It would be worthwhile to make a theoretical calculation of the anisotropy in the velocities of the longitudinal modes. It is likely that the longitudinal mode with its wave vector perpendicular to the copper oxygen infinite plane has the lowest velocity and softens appreciably as the temperature of the sample is lowered. If so its softening must be at least coupled to the electronic energy arising from the two dimensional character of the electronic energy band.

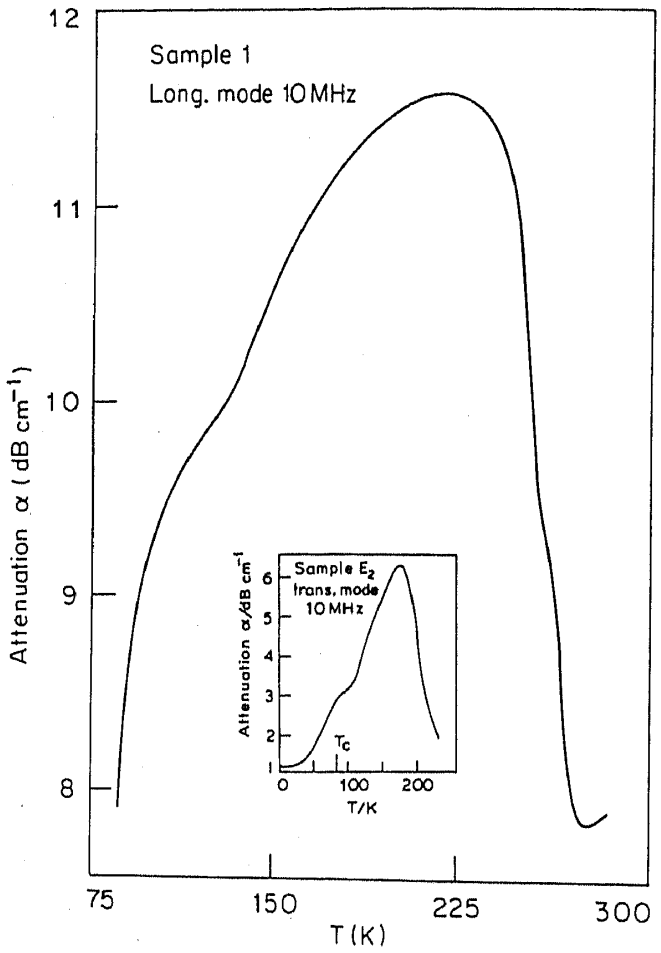

Figure 3. Ultra sound attenuation (α) as a function of temperature for sample (1). The inset shows ultrasound attenuation in transverse mode observed by Ewert *et al.*

Figure 3 gives the attenuation in dB/cm as the material was cooled. The attenuation shows an initial drop as the sample is cooled from room temperature. This is followed by a sharp rise near 270 K. Then it shows a maximum with a tendency for the attenuation to drop as the transition point is approached. It was difficult to estimate the attenuation at the transition temperature as the echoes were not stable. The measurements were done up till 95 K. The shape of the attenuation curve resembles that of Ewert *et al* for transverse mode. The inset in figure 3 shows attenuation curve of Ewert *et al.*

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References

Ewert S, Guo S, Lemmens P, Stellmach F, Wynants J, Art G, Dorothee Bonnenberg, Kliem H, Comberg A and Helga Passing 1987 *Solid State Commun.* (to appear)

Fossheim K, Laegreid T, Sandvold E, Vassenden F, Muller K A and Bednorz J G 1987 *Solid State Commun.* **63** 531

Luthi D, Wolf B, Kim T, Grill W and Renker B 1987 *Jpn. J. Appl. Phys.* Vol. 26 Supplement 26–3.