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Ultrasonic attenuation measurements in sinter-forged $YBa_2Cu_3O_{7-\delta}$

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We report ultrasonic attenuation measurements on sinter-forged $YBa_2Cu_3O_{7-\delta}$ material, which differs from ordinary sintered material in that the crystallites are preferentially oriented to form a uniaxial sample. Three peaks in attenuation, at temperatures of 250, 180, and 70 K, were observed for longitudinal waves propagating perpendicular to the forging axis, which is similar to that reported in ordinary (isotropic) polycrystalline samples. However, for both transverse and longitudinal sound propagated along the forging axis we have a different behavior, with only one peak at 180 K, showing a strong anisotropy. It is suggested that sound waves traveling parallel to and normal to the Cu-O planes may account for the anisotropic effect, and a relaxation mechanism may explain the increase in shear wave attenuation which was seen with decreasing temperature.

Sound attenuation measurements have proved to be very useful in investigating the bulk properties of superconducting materials. Ultrasound investigations of high-T_c materials such as $YBa_2Cu_3O_{7-\delta}$ could provide useful information for understanding the mechanisms that produce superconductivity in these materials, and measurements have been performed on isotropic polycrystalline samples.[1-5]

Until sufficiently large single crystals do become available for ultrasonic measurements, a good compromise is to do the present measurements on sinter-forged $YBa_2Cu_3O_7$ samples, which show a marked degree of anisotropy with respect to the forging axis. These sinter-forged materials differ from ordinary sintered polycrystals. The latter are isotropic and therefore do not offer the possibility of differentiating effects that are orientation dependent and which would provide insight into the polarization dependence of the interactions.

In this paper, we report attenuation measurements in sinter-forged $YBa_2Cu_3O_{7-\delta}$ materials, which differ from ordinary sintered isotropic polycrystals in that the individual crystallites in these samples are preferentially oriented along the forging axis. As determined by x-ray diffraction and optical micrographs, the sinter-forged samples have the axis of their individual crystallites aligned parallel to the forging axis with an rms deviation of about 20°; the average crystallite size is about 11 μm thick along the c axis and 37 μm in diameter.

There are five propagation configurations involving the principal axes of this uniaxial compact: longitudinal waves propagating parallel and perpendicular to the forging axis, transverse waves propagating perpendicular to the forging axis with the polarization parallel or perpendicular to the forging axis, and finally, transverse waves propagating along the forging axis.

The experiments were performed on three sinter-forged samples, whose density was determined (with the Archimedes method) to be 97% of that of single crystals. The preparation process is described in Ref. 6. The samples were cut into rectangular shapes, ground to have opposite sides parallel and then polished to a 3-μm finish. Either 12-MHz X-cut or 14.5-MHz AC-cut overtone-polished quartz transducers were used to produce longitudinal or transverse sound waves. Two matched transducers were epoxy bonded (with Epon Resin 815) to opposing faces of each sample to perform "transmission" measurements, in contrast to the conventional single-ended reflection method, in order to reduce the interference of the signal echoes with the rf feed through. A pulse-echo method was used to measure the attenuation and involved the Matec GA-825, ESA-8000A, and 6600 system to generate and detect the rf pulses.

The signal obtained from the receiving transducer was applied to two independent detectors. The first was the Matec ESA-8000A phase sensitive detector which has a computer controlled data acquisition system directly yielding attenuation changes. The second was a more conventional pulse envelope detector, the Matec 6600, the output of which was boxcar detected (and averaged) and applied to an x-y chart recorder. The resulting attenuation changes were essentially identical for both detectors.

Figure 1(a) shows longitudinal ultrasonic attenuation for 12 MHz with sound propagating perpendicular to the forging axis. Surprisingly, the shape of the curve is very similar to that reported earlier by several groups[2-5] for ordinary isotropic sintered polycrystalline samples. Broad peaks in the attenuation were observed around 250, 180, and 70 K, the latter being below the superconducting transition temperature of 91 K (obtained from the midpoint of resistance transition which is plotted on the same figure). Compared to the results previously reported on isotropic compacts, the position of the peaks is shifted by a few degrees Kelvin. Bhattacharya *et al.*[2] reported peaks around 130 and 65 K in the longitudinal sound attenua-

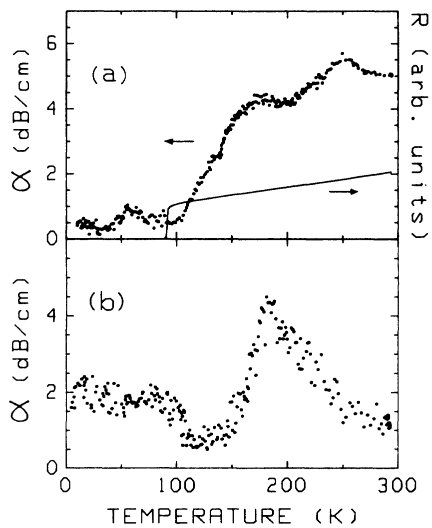

FIG. 1. The ultrasonic attenuation of 12-MHz longitudinal waves propagating (a) perpendicular to the forging axis (the solid line is the resistance) and (b) parallel to the forging axis.

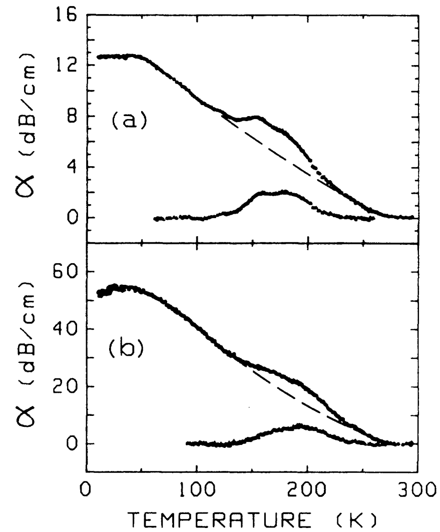

FIG. 2. The ultrasonic attenuation of transverse waves propagating parallel to the forging axis at (a) 14.5 MHz and (b) 41.0 MHz. The dashed lines indicate the interpolated background of (a) and (b) and the lower curves are the resulting attenuation after the background is subtracted, respectively.

tion at 5 MHz; Sun *et al.*[3] reported a peak at about 82 K, at 10 MHz, which shifted to higher temperatures with higher frequencies; Xu *et al.*[4] observed peaks at 250 and 84 K for 15 MHz; and He Yusheng *et al.*[5] reported peaks at 250, 160, and 78 K for 10 MHz. On a different compound, $Y_2Ba_4Cu_6O_{14}$, Horie *et al.*[7] found peaks in attenuation at 245 and 160 K for 10 MHz. From these relatively similar reports, we can conclude that the longitudinal ultrasonic attenuation in the vicinity of 10 MHz has peaks in three different temperature regions: around 250, 160, and 70 K. Variations from sample to sample, due to differences in grain size, porosity and T_c's, as well as a variation in the frequency of the sound waves, caused the position of the peaks to shift somewhat in temperature. Sound velocity measurements[8,9] also showed anomalies in the same three temperature ranges. These attenuation peaks may be caused by strong lattice anharmonicities or lattice instabilities associated with phase transitions. One or more of the peaks may be Debye relaxation maxima. The temperature-dependent relaxation time could be associated with a relaxation between a two-energy-level system, such as oxygen tunneling between two lattice sites, or the thermal excitation of a soft plasmon, an exciton, a spin-density wave or a charge-density wave.

Figure 1(b) shows the longitudinal attenuation versus temperature at 12 MHz for sound traveling parallel to the forging axis. Here, we saw only one pronounced peak at 180 K and a change in slope near T_c.

Figure 2(a) displays the attenuation for transverse waves as a function of temperature at 14.5 MHz for sound propagating along the forging axis. Note the attenuation monotonically increases on cooling, which is in contrast to the tendency shown in Fig. 1(a), where the attenuation coefficient in general decreased as the temperature was lowered. A shoulder in the upper curve in Fig. 2(a) appears at a temperature ~160–170 K. The dashed line represents an interpolation of the background attenuation subtending the shoulder. The lower curve shows the resulting attenuation curve when the background is subtracted. This resultant curve has a maximum at around 180 K, which corresponds to the peak in longitudinal damping in Fig. 1(a).

In Fig. 2(b), we show the transverse attenuation at 41 MHz, again for propagation parallel to the forging axis. The behavior looks quite similar to that shown in Fig. 2(a), however the total attenuation change per centimeter from room temperature to 10 K at 41 MHz is approximately 4.2 times that at 14.5 MHz, which implies a frequency dependence of about $\omega^{3/2}$ (which may be compared to the ω^2 behavior typical of hydrodynamic attenuation mechanisms). Again there is a shoulder in the upper curve and after the background is subtracted, using the same procedure as in Fig. 2(a), a relaxation curve is obtained with a maximum around 195 K, as seen in the lower curve.

There is evidence of three relaxation maxima for longitudinal waves propagating perpendicular to the c axis, while there is evidence for only one relaxation maximum when either longitudinal or transverse waves propagate parallel to the c axis. The values of the temperatures associated with these maxima are given in Table I. It is apparent that the maximum at around 180 K is obtained for both longitudinal and transverse waves while the maxima at 70 and 250 K are only seen with longitudinal waves propagating perpendicular to the c axis.

Longitudinal waves traveling parallel to the c axis will produce stresses which change the separation between the Cu-O planes in this perovskite superconductor. Transverse waves will displace these planes in a shearing motion. Both of these distortions appear to couple to the relaxation mechanism which produces the 180 K maxima.

TABLE I. Ultrasonic attenuation peak position in temperature vs type of sound waves and their frequencies.

Sound propagation	Attenuation peak position (K)		
12 MHz longitudinal, $q \perp c$	70	180	250
12 MHz longitudinal, $q \parallel c$	...	180	...
14.5 MHz transverse, $q \parallel c$	...	180	...
41.0 MHz transverse, $q \parallel c$	...	195	...

Longitudinal waves propagating perpendicular to the c axis will produce stresses within the Cu-O planes. These appear to be the distortion required to couple to the relaxation mechanisms which produce the maxima at 70 and 250 K. Therefore, we may assume that these maxima are associated with interactions originating in the Cu-O planes. The maximum at 180 K is also produced by these distortions. This could imply that the relaxation process that produces this maximum is an isotropic effect or it could be due to the fact that there is only 80% alignment of the c axis of crystallites along the forging axis.

Sun *et al.*[3] have reported frequency dependent measurements on the maxima that appears around T_c in a sintered $YBa_2Cu_3O_{7-\delta}$ sample. They find an activation energy of about 400 K for the excitations that couple to the sound waves. Our present results would indicate that these excitations may be associated with the two-dimensional Cu-O planes.

Figures 2(a) and 2(b) indicate that the maximum shifts from 180 to 195 K when the frequency of the transverse waves is changed from 14.5 to 41 MHz. These results are consistent with a relaxation model wherein the attenuation goes as $\alpha(\omega)/\alpha(\omega)_{max} = 2\omega\tau/(1+\omega^2\tau^2)$. The maximum occurs at $\omega\tau = 1$ and shifts to lower values of τ as ω is increased. Therefore, we may make the reasonable deduction that τ decreases with increasing temperature for the mechanism that is involved in this relaxation process.

It is perplexing that the overall attenuation for transverse waves propagating parallel to the forging axis increases as the temperature is reduced. It may be that this indicates an interaction with the macroscopic crystallites themselves, which experience a shearing in the matrix during the passage of a transverse wave parallel to the forging axis. If the crystallites do not move in phase with the sound wave, a relaxation maximum could result and the fact that the attenuation increases with decreasing temperature is just an indication that, although $\omega\tau$ increases with decreasing temperature, it is still smaller than one for this temperature range at 14.5 MHz. However, there is an indication of a maximum in the 41 MHz data at around 25 K. So it is possible that $\omega\tau = 1$ for this frequency at 25 K yielding a $\tau = 4\times10^{-9}$ sec.

The above model would suggest that measurements with shear waves propagating perpendicular to the forging axis would exhibit different characteristics, depending on the polarization direction of the waves. If the waves are polarized perpendicular to the forging axis then it might be expected that the attenuation would behave similarly to the data shown in Fig. 2(a). If the polarization were parallel to the forging axis then the attenuation would be similar to the data shown in Fig. 1(a), since the Cu-O planes would be sheared by the transverse waves; and, the shearing motion on the area of the perimeter of the crystallites should produce less losses than it does on the faces. Measurements are in progress to determine the temperature dependence of the attenuation for these two polarizations.

In summary, attenuation data have been obtained on sinter-forged $YBa_2Cu_3O_{7-\delta}$, having a c-axis alignment of the crystallites within about 20° of the forging axis. Three maxima, at 70, 180, and 250 K, are obtained with longitudinal waves traveling perpendicular to the forging axis while only one maximum, at 180 K, is obtained with either longitudinal or transverse waves traveling parallel to the forging axis. A model is presented that suggests that the 70 and 250 K maxima are associated with distortions of the Cu-O plane while the 180 K maximum may be associated with volume distortions. The attenuation of transverse waves increases as the temperature is lowered, in contrast to the behavior of longitudinal waves. It is suggested that this increase in attenuation is produced by a relaxation mechanism involving a shearing of the crystallites in the sinter-forged sample by the shear motion, which is predominantly parallel to the diameter of the crystallites.

More results have been obtained on the attenuation peak, below T_c, that had been reported earlier by Xu *et al.*[4] Recent results reported by Sun *et al.*[3] on $YBa_2Cu_{43}O_7$ on sintered samples as a function of frequency show that the peak around T_c moves to higher temperatures as the frequency is increased. This leads us to believe that this particular peak is a relaxation peak.

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