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Ultrasonic Study of $YBa_2Cu_3O_{7-x}$ Superconductors

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Abstract. We investigated the mechanical properties of the sintered $YBa_2Cu_3O_{7-x}$ superconductors from the view point of the porosity and elastic anomalies. In the highly textured sample (5.93 g/cm³, porosity of 7%), the Debye temperature was 440 K determined from the longitudinal and transverse velocities ($V_1 = 4.9$ km/s, and $V_t = 3.2$ km/s), which agreed very well with those measured by the specific heat and Mossbauer experiments ever reported. The background attenuation due to the grain-boundary scattering was 7 dB/cm at 9.7 MHz - longitudinal ultrasonic mode. On the other hand, in the porous sample (4.7 g/cm³, porosity of 25%), the Debye temperature was 320 K determined from the velocities of $V_1 = 3.5$ km/s and $V_t = 2.3$ km/s, and the background attenuation was 10 dB/cm at 9.7 MHz - longitudinal mode. Some kinds of elastic anomalies were observed. In the sample (x = 0.53, T_c the step-like attenuation was detected just above T_c (2 dB/cm), which was the ultrasonic absorption due to the lattice instability, and essentially related to the superconducting transition. The relaxation-type sharp peak was observed (9 dB/cm at 62 K) in the sample of YBa₂Cu₃O_{7-x} ($x \approx 0$, $T_c \approx 90$ K), with a narrow temperature width of 15 K. It was due to the ultrasonic absorption by jump motion of oxygen atoms between two stable sites.

1. INTRODUCTION

Ultrasonic method, a powerfull tool for studies of the metal superconductors, has been also applied to the the high- T_c cuprate superconductors in order to clarify the superconductivity mechanism through the elastic properties. Levy reviewed the many studies of ultrasonic measurements of the cuprate superconductors [1]. Wu-Ting and Fossheim surveid on the elastic anomalies - lattice instabilities, and discussed their mechanisms [2]. Dominec et al. summarized the elastic constants of Bi-Sr-Ca-Cu-O (BSCCO), La-Sr-Cu-O (LSCO), and Y-Ba-Cu-O (YBCO) superconductors [3]. More than two hundred papers have been published on YBCO related materials, however, the relation between elastic anomalies and superconductivity is not clarified enough.

In this study we made the experiment of ultrasonic propagation in the sintered $YBa_2Cu_3O_{7-x}$ superconductors in order to investigate the elastic anomalies caused by the various lattice-instability mechanisms. We report the ultrasonic absorption which is essentially related to the superconducting transition, and discussed the high temperature superconductivity mechanism.

2. EXPERIMENTAL

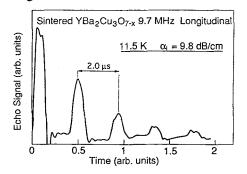
Two YBCO samples with different critical temperatures (T_c) were prepared. The specifications are summarized in Table I. The porosity of the 90-K YBCO ($T_c = 90$ K) was as high as 25 %. The highly textured 60-K YBCO ($T_c = 60$ K) was low in the porosity and oxygen content. The samples were shaped into the disks of 10 mm in diameter, and the two parallel surfaces were polished smoothly. The ultrasonic measurement was a standard pulse echo method. Piezoelectric quartz plates were used for generation and detection of the ultrasonic waves. The quartz plate was fastened to the YBCO sample with silicone grease. The YBCO sample was attached to a Cu stem and set into the cryostat. The temperature was stabilized within 0.2 K during the measurement. Ultrasonic attenuation coefficient was determined from the decay of ultrasonic pulse energy during the propagation. The attenuation spectrum was plotted as a function of temperature.

x		density (g/cm ³)	porosity (%)	thickness (cm)
≈ 0	(T _c ≈ 90 K)	4.7	25	0.35
0.35	$(T_c = 60 \text{ K})$	5.93	7	0.4

 Table I
 Specifications of the sintered YBa₂Cu₃O_{7-x} superconductors.

3. RESULTS

Figure 1 shows a typical echo train in the 90-K YBCO at longitudinal-9.7 MHz and 11.5 K. The attenuation was 9.8 dB/cm, most of which was ultrasonic energy loss due to the grain-boundary scattering of the sound wave. The longitudinal and transverse velocities were $V_1 = 3.5$ km/s and $V_t = 2.3$ km/s. These were lower than those in the single crystal YBCO because of the porosity. The temperature dependence of the attenuation is plotted in Fig. 2. The background attenuation of 10 dB/cm was almost constant in the measurement temperatures. Three attenuation peaks were observed. The attenuation change $\Delta \alpha_i$ in the second peak at 62 K was as large as 9 dB/cm, and also large attenuation in the third peak at 180 K. The first peak at 30 K, however, was less than 1 dB/cm. In this spectrum, no remakable attenuation change was observed at 90 K.



Sintered YBa₂Cu₃O_{7-x} 9.7 MHz Longitudinal

Figure 1: Ultrasonic pulse echo train in the 90-K YBCO.

Figure 2: Temperature dependence of the ultrasonic attenuation in the 90-K YBCO.

Figure 3 shows the echo train in the 60-K YBCO at 14.0 K. The longitudinal and transverse sound velocities were $V_1 = 4.9$ km/s and $V_t = 3.2$ km/s, which were very close to those in the single crystal. Small ripples appearing in the echo pattern was due to the sound wave reflection from microcracks introduced characteristically into the highly textured samples, however, they did not affect the determination of the ultrasonic parameters. The temperature dependence of the attenuation around T_c is plotted in Fig. 4. The background attenuation was less than 7 dB/cm. The step-like attenuation change $\Delta \alpha_l = 2$ dB/cm was observed at T_c . A small respose was observed below T_c .

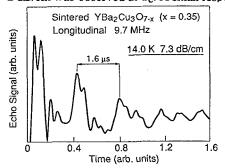


Figure 3: Ultrasonic pulse echo train in the 60-K YBCO.

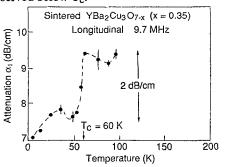


Figure 4: Temperature dependence of the ultrasonic attenuation in the 60-K YBCO around T_c.

4. **DISCUSSION**

We evaluate the porosity dependence of the Debye temperature Θ_D expressed by the equation,

$$\Theta_{\rm D} = ({\rm h/k})(9{\rm N}/4\pi{\rm V})^{1/3}/[1/{\rm V_l}^3 + 2/{\rm V_t}^3]^{1/3} \tag{1}$$

h and k are the Plank's and Boltzmann's constants, respectively. The number of atoms N in the YBCO unit cell is 13, and the unit cell volume V is 174×10^{-24} cm³. From the longitudinal and transverse velocities (V₁, V_t), Θ_D is determined as 320 K for the 90-K YBCO and 440 K for the 60-K YBCO. The latter is close to the value of the single crystal. These values of Θ_D assure the validity of the ultrasonic experiment and the physical characteristics of the YBCO samples. Here, we must remark that Θ_D is determined by the velocities dependent on the porosity, and is independent of the oxygen content.

The change of the ultrasonic attenuation near 60 K is drastic both in the 90-K and 60-K YBCO. This peak looks as like as that observed by Cannelli and Cantelli in the internal fiction experiment on the outgassed YBa₂Cu₃O₆ [4]. They identified it with the mechanical energy absorption by oxygen-atom jump motion between two stable sites in the Cu-O plane. However, since our sample YBa₂Cu₃O_{7-x} ($x \approx 0$) is of highest oxygen stoichiometry, it would not be appropriate to identify our 62-K peak simply with that of oxygen jump motion.

On the other hand, the change $\Delta \alpha_1 = 2$ dB/cm in the 60-K YBCO is distinctly different from the relaxation peak. The attenuation shows a rapid decay just below T_c and a plateau above T_c, which suggests the condensation associated with the superconducting transition. We analyze this using the model of strong correlation between carriers and local lattice distortion [5]. The increase in the ultrasonic attenuation is,

$$\Delta \alpha_{\rm l} (\rm dB/cm) = 8.686 \ x \ A\omega/(2V_{\rm l}), \tag{2}$$

in the strong correlation limit. A is the relaxation strength,

$$A = 4n_{\rm H}m^*V_{\rm F}^2/(15\rho V_{\rm I}^2).$$
(3)

We calculate the attenuation using the parameters as follows. The carrier (hole) concentration n_H is 1×10^{21} cm⁻³ typically in the high-T_c superconductors. The effective mass m* is 7m_e determined from the de Haas van Alphen oscillation where m_e is the free-electron mass [6], and the Fermi velocity V_F is 7.5 x 10⁷ cm/s from the Landau damping in Raman experiment [7]. The mass density ρ and longitudinal sound velocity V_I are 5.93 g/cm³ and 4.9 x 10⁵ cm/s in our 60-K YBCO. The attenuation $\Delta \alpha_1$ (calc.) = 3.6 dB/cm is well corresponding to the measurement $\Delta \alpha_1$ (meas.) = 2 dB/cm, even considering the arbitrariness in the carrier concentration.

The attenuation change at T_c was not observed in the 90-K YBCO. This would be because the high background attenuation disturbed the detection, or, it was hidden under the skirt the large relaxation peak. We could refer an interesting report of the ultrasonic measurement [8]. Sun et al. extracted a step-like attenuation at T_c in the 90-K YBCO of which spectral pattern was the same as in Fig. 4.

We observed the abrupt attenuation change also in the sintered Bi₂Sr₂CaCu₄O₈ (Bi-2212, BSCCO) at T_c (= 84 K) [9]. The value of $\Delta \alpha_1$ was 0.9 dB/cm at 9.7 MHz, the same extent with that of the 60-K YBCO.

This $\Delta \alpha_1$ is coincident with the internal friction measured by Huang et al. [5]. The lattice softening in the BSCCO was also observed by resonant neutron scattering and Raman scattering measurements [10,11]. Missori et al. observed the oxygen atom displacement in the BSCCO in the EXAFS experiment [12]. These experimental facts are consistent with our work, and would validate the discussion of the local lattice distortion associated with the superconductivity.

5. CONCLUSIONS

The elastic properties of the sintered $YBa_2Cu_3O_{7-x}$ high- T_c superconductors was investigated by ultrasonic pulse echo measurements. The porosity-dependent sound velocity and the various ultrasonic absorption were clarified. The relaxation-type large absorption was due to the jumping motion of oxygen atoms. The step-like absorption was directly related to the superconducting transition which was discussed by the model of strong correlation between carriers and local lattice distortion.

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