Journal of Alloys and Compounds, 211/212 (1994) 300–304
JALCOM 121

The effect of neutron and γ irradiation on YBCO and B(Pb)SCCO high-T_C superconductors

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Abstract

The effect of neutron irradiation on YBCO high-T_C superconductor ($T_C = 93$ K) and the effect of γ irradiation on B(Pb)SCCO high-T_C superconductor ($T_C = 87$ K) were studied. Each sample was given successive irradiation for 5, 10, 15, 20 and 30 h. The mechanical loss spectra were recorded between liquid nitrogen and room temperature. In YBCO, the height of the four relaxation peaks observed increases as the neutron dose is increased. The peak heights increase by nearly a factor of two (when comparing neutron irradiation for 30 h with the non-irradiated sample). In B(Pb)SCCO, the peak heights increase by a factor of 1.5 (for 30 h γ irradiation). The peak temperatures in both high-T_C superconductors show almost no change as a function of the neutron or gamma irradiation. In YBCO, the peaks occur at 113, 123, 147 and 243 K, whereas in B(Pb)SCCO, the peaks occur at 87, 112, 128, 170 and 230 K, when measurement is carried out at 10 MHz using the pulse-echo technique (single-ended).

1. Introduction

Mechanical loss experiments (internal friction) are very sensitive to the structural defects of compounds and their superconductivity. Ultrasonic attenuation also provides information on superconductors, *e.g.* the change in the longitudinal and shear sound velocity (Δv_l and Δv_s respectively) in the vicinity of the critical temperature T_C, the longitudinal L and shear G moduli, the bulk modulus B and Young's modulus E. These are related as follows [1–3]: $L = \rho v_l^2$; $G = \rho v_l^2$; $E = G(3L - 4G)/(L - G)$;

$$\Delta B/B = (\Delta v_l/v_l)[2/\{1 - (\tfrac{4}{3})(v_s^2/v_l^2)\}]$$

where ρ is the density of the superconductor. Furthermore, from the measurement of the mechanical spectra at different frequencies f, we can calculate with good approximation, the bound energy of small bipolarons $'\Delta$ (which is believed to be the mechanism responsible for the relaxation peaks in the vicinity of the critical temperature T_C [4, 5]) using the following relation [6]

$$'\Delta = \left\{\frac{-8W}{(\ln f/f_0)T_m}\right\} T_C \qquad (1)$$

where W is the activation enthalpy for the relaxation process and f_0 is the attempt frequency. The effect of the grain size on T_C [7, 8] and on the hysteresis behaviour of both velocities (v_l and v_s) [9] was detected by ultrasonic attenuation measurement.

In this paper, we study the effect of neutron and γ irradiation on two high-T_C superconductor compounds, *i.e.* $Y_1Ba_2Cu_3O_{7-x}$ and $Bi_{0.8}Pb_{0.2}SrCaCu_{1.5}O_y$, by ultrasonic attenuation measurement at 10 MHz using the pulse-echo technique (single-ended).

2. Experimental procedures

The $Y_1Ba_2Cu_3O_7$ (YBCO) samples used in this study were prepared by the standard solid state reaction in the form of pressed ceramic pellets. The critical temperature was found to be 90 K. $Bi_{0.8}Pb_{0.2}SrCaCu_{1.5}O_y$ (B(Pb)SCCO) was found to have $T_c = 87$ K. A detailed explanation of the preparation of YBCO and B(Pb)SCCO and the technique used, *i.e.* ultrasonic attenuation (at 10 MHz), is given in refs. 10 and 11 respectively.

The YBCO high-temperature compound was given successive neutron doses (expressed in units neutron–velocity–time, nvt) at room temperature, *i.e.* 1.8×10^{10} nvt (5 h exposure), 3.6×10^{10} nvt (10 h exposure), 5.4×10^{10} nvt (15 h exposure), 7.2×10^{10} nvt (20 h exposure) and 10.8×10^{10} nvt (30 h exposure). The B(Pb)SCCO high-temperature superconductor was irradiated by γ rays (expressed in units gamma particles–velocity–time, γvt) giving doses of 4.28×10^8 γvt (10 h exposure), 6.45×10^8 γvt (15 h exposure), 8.6×10^8 γvt (20 h exposure) and 12.85×10^8 γvt (30 h exposure).

SSDI 0925-8388(94)00121-R

Detailed information on the irradiation procedures and the ultrasonic attenuation measurement at 10 MHz from liquid nitrogen (77 K) to room temperature (300 K) is given elsewhere [12].

For the frequency used in this work, the damping was calculated using the following relation

$$Q^{-1}=(0.036\alpha C)/f$$

where α is the ultrasonic attenuation (dB cm^{-1}), C is the speed of sound (cm s^{-1}) and f is the frequency of measurement (Hz). In order to find the strength of the relaxation peaks, the background friction was subtracted using the method devised by Alnaser [13].

3. Results and discussion

3.1. Variation of the relaxation peak heights Q_m^{-1} and their temperatures T_m with neutron dose D_n in YBCO

Figure 1 shows the variation of the ultrasonic attenuation with temperature at successive neutron doses from zero to 10.8×10^{10} nvt. Four peaks were observed in this mechanical spectrum, *i.e.* P_1, P_2, P_3 and P_4. As the neutron dose D_n increases, Q_m^{-1} increases accordingly as explained by the following correlations

For P_1: $Q_m^{-1}=4.231\times10^{-4}+1.81\times10^{-15}D_n$, $r=0.6511$

For P_2: $Q_m^{-1}=3.801\times10^{-4}+2.532\times10^{-15}D_n$, $r=0.9664$

For P_3: $Q_m^{-1}=9.173\times10^{-4}+9.675\times10^{-15}D_n$, $r=0.8986$

For P_4: $Q_m^{-1}=12.98\times10^{-4}+10.63\times10^{-15}D_n$, $r=0.8889$

For $P_{average}$: $Q_m^{-1}=7.456\times10^{-4}+6.163\times10^{-15}D_n$, $r=0.9049$ (2)

The above relations indicate that P_2 peak responses vary systematically with neutron irradiation ($r=0.9664$), while P_1 does not ($r=0.6511$).

T_m variation with neutron irradiation is not systematic, *i.e.* there is no relation between T_m and D_n.

3.2. Variation of the relaxation peak heights and their temperature with γ dose D_γ in B(Pb)SCCO

Figure 2 shows the variation of the ultrasonic attenuation, measured at 10 MHz, with temperature at successive γ ray doses from zero to 12.85×10^8 γvt. Five relaxation peaks (P_1, P_2, P_3, P_4 and P_5) were observed. The peak heights Q_m^{-1} were found to increase as the gamma dose D_γ increases (but not as systematic as in YBCO). This can be understood from the following equations

For P_1: $Q_m^{-1}=6.129\times10^{-4}+4.933\times10^{-13}D_\gamma$, $r=0.8738$

For P_2: $Q_m^{-1}=8.266\times10^{-4}+3.658\times10^{-13}D_\gamma$, $r=0.8688$

For P_3: $Q_m^{-1}=1.079\times10^{-3}+3.738\times10^{-13}D_\gamma$, $r=0.8743$

For P_4: $Q_m^{-1}=1.189\times10^{-3}+6.070\times10^{-13}D_\gamma$, $r=0.9341$

For P_5: $Q_m^{-1}=1.567\times10^{-3}+8.286\times10^{-13}D_\gamma$, $r=0.8641$

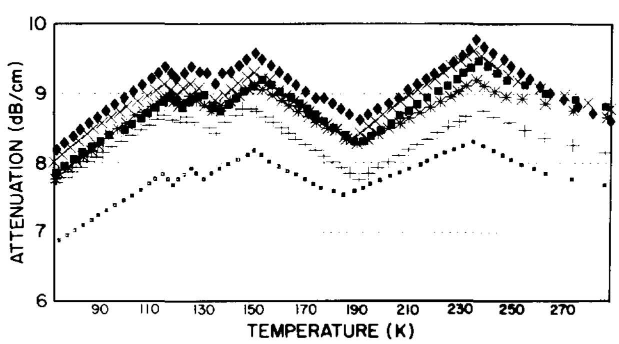

Fig. 1. The variation of the ultrasonic attenuation at 10 MHz with temperature in YBCO high-T_C superconductor given successive neutron doses; •, not irradiated; +, 5 h; *, 10 h; ■, 15 h; ×, 20 h; ◇, 30 h.

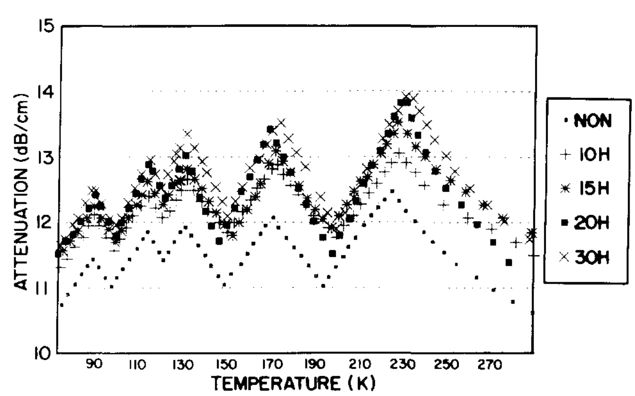

Fig. 2. The variation of the ultrasonic attenuation at 10 MHz with temperature in B(Pb)SCCO high-T_C superconductor given successive γ doses: •, not irradiated; +, 10 h; *, 15 h; ■, 20 h; ×30 h.

For $P_{average}$: $Q_m^{-1}=8.171\times10^{-4}+4.123\times10^{-13}D_\gamma$,

$$r=0.9090 \quad (3)$$

The above relations indicate that P_4 peak responses vary systematically with γ irradiation dose ($r=0.9341$), while P_2 does not.

The variation of T_m with D_γ shows no significant relation, similar to that found in YBCO.

3.3. Variation of the longitudinal sound velocity v_l in YBCO with D_n

Figure 3 shows the variation of the velocity as a function of neutron dose. The sound velocity at each peak increases on average by nearly 6% as the neutron dose is increased to 10.8×10^{10} nvt. Small changes in the slope are observed in the region where the relaxation peaks occur. The figure also shows that the increase in velocity with irradiation is significant up to 7.2×10^{10} nvt, after which the increase in v_l becomes very small (*i.e.* indication of saturation). The room-temperature longitudinal sound velocity before irradiation was 4.893 km s^{-1}. This value is very slightly higher than that measured by Srinivasan *et al.* [14] (*i.e.* 4.1%) and that measured by Lemmens *et al.* [15]. Since a knowledge of the shear velocity of sound is important (in order to calculate the mechanical properties), we can assume that $v_s/v_l\approx0.6$ [3]. However, the mean speed of sound v_m can be calculated using the following relation [16]

$$v_m=1/\{(\tfrac{1}{3})(2/v_s^3+1/v_l^3)\}^{1/3} \quad (4)$$

This means that v_s in our sample is 2.93 km s^{-1} and v_m is nearly 3.221 km s^{-1}. Since the density of our YBCO superconductor is nearly 6 g cm^{-3}, $L=143.6$ GPa, $G\approx51.7$ GPa and $E\approx126$ GPa. These values near T_C (90 K) ($v_l=4.870$ km s^{-1}, $v_s=2.922$ km s^{-1}) become as follows: $L=142.3$ GPa, $G\approx51.2$ GPa and $E\approx124.8$ GPa. When the superconductor is given a neutron dose of 10.8×10^{10} nvt, these mechanical parameters near T_C become equal to $v_l=5.196$ km s^{-1}, $v_s\approx3.118$ km s^{-1}, $L=162$ GPa, $G\approx58.3$ GPa and $E\approx142$ GPa, *i.e.* the mechanical properties increase as the neutron dose increases.

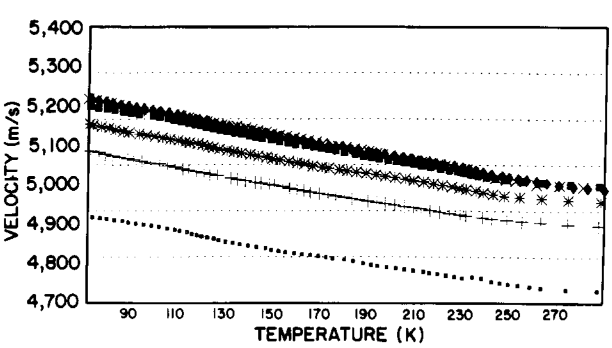

Fig. 3. The variation of the longitudinal velocity of sound with temperature in YBCO high-T_C superconductor given successive neutron doses: symbols as in Fig. 1.

To calculate $\Delta B/B$ we plotted $\Delta v_l/v_l$ *vs.* T as shown in Fig. 4. We have taken the average peak temperature of P_1, P_2 and P_3, which is believed to be a complex peak and was attributed to thermally activated atomic jumps (probably over distances appreciably less than the interatomic distances [17]). The results show that $\Delta v_l/v_l\approx500$ ppm for the non-irradiated sample and $\Delta v_l/v_l\approx450$ ppm, for the strongly irradiated sample, which means that $\Delta B/B$ for the non-irradiated sample of this superconducting compound is much larger than that reported by Dominec and Plechacek [3]. It should be noted that the error for $\Delta v_l/v_l$ varies from 40% to 60%.

The values of Δv_l in this work lie within the values reported in the review paper by Dominec [1], *i.e.* +4500 ppm to −1600 ppm.

3.4. Variation of the longitudinal sound velocity in B(Pb)SCCO with D_γ

Figure 5 shows the variation of v_l with temperature. We were unable to measure the velocity of sound in B(Pb)SCCO as a function of γ irradiation for technical

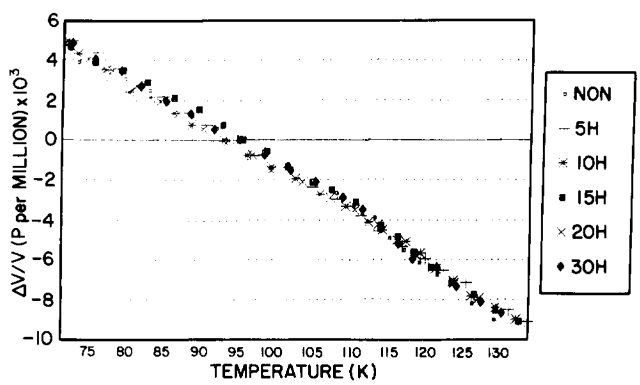

Fig. 4. Plot of $\Delta v_l/v_l$ as a function of temperature for the complex relaxation peak in YBCO high-T_C superconductor given successive neutron doses.

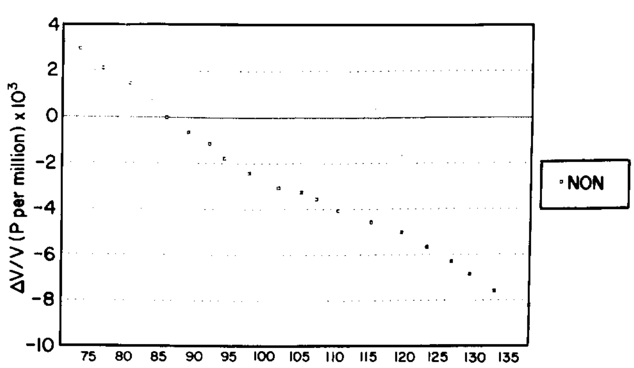

Fig. 5. The variation of the longitudinal velocity of sound with temperature in B(Pb)SCCO high-T_C superconductor.

reasons. However, it is expected that the velocity of sound will also increase as the γ irradiation increases. The mechanical parameters for B(Pb)SCCO near T_C (87 K) are as follows: $L=54.3$ GPa ($\rho=5.146$ g cm^{-3}, $v_l=3.248$ km s^{-1}), $G\approx30.5$ GPa (note that v_s/v_l in B(Pb)SCCO is 0.75 [3]) and $E\approx52.4$ GPa. Figure 6 shows the variation of $\Delta v_l/v_l$ in B(Pb)SCCO, from which the value of $\Delta B/B$ is calculated to be 470, close to that reported in ref. 3.

3.5. *Variation of the background ultrasonic attenuation in high-temperature YBCO and B(Pb)SCCO*

In an attempt to study the relation between the variation of the background attenuation α_{BG} (dB cm^{-1}) and the maximum recorded attenuation α_m with neutron and γ irradiation, we found the following statistical correlations. For YBCO relaxation peaks

$$D_n=-5.738\times10^{11}+6.93\times10^{10}\alpha_m,\quad r=0.9234$$

$$D_n=-5.98\times10^{11}+8.059\times10^{9}\alpha_{BG},\quad r=0.8649 \qquad (5)$$

$$D_n=-3.7957\times10^{11}+2.484\times10^{11}\alpha_m -2.246\times10^{11}\alpha_{BG},\quad r=0.9854$$

For B(Pb)SCCO relaxation peaks

$$D_\gamma=-1.086\times10^{10}+9.010\times10^{8}\alpha_m,\quad r=0.9532$$

$$D_\gamma=-1.066\times10^{10}+9.933\times10^{8}\alpha_{BG},\quad r=0.8400 \qquad (6)$$

$$D_\gamma=-1.052\times10^{10}+1.015\times10^{9}\alpha_m -1.5824\times10^{8}\alpha_{BG},\quad r=0.9548$$

The above relations clearly indicate that there is a strong correlation between the irradiation (either neutrons or γ) and the recorded attenuation (background and maximum) for the relaxation peaks in the high-T_C superconductor compounds. We found from our experiments that the background relaxation peak increases by almost 15% as the neutron dose increases from zero to 10.8×10^{10} nvt in YBCO, while it increases by 9% (on average) as the γ ray dose increases from zero to 12.85×10^{8} γvt in B(Pb)SCCO. This shows that neutron irradiation causes more disorder in the superconductor than γ irradiation. This increase in the ultrasonic attenuation is attributed to the scattering of the sound wave in the disordered (inhomogeneous) high-T_C compounds.

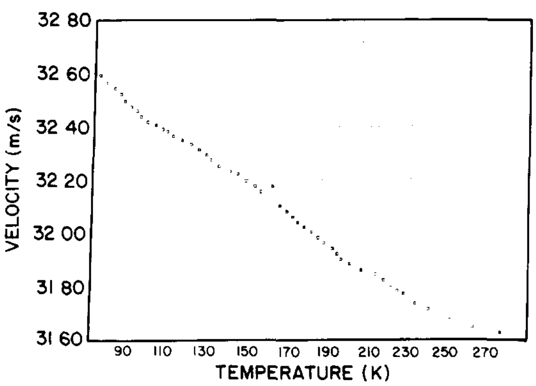

Fig. 6. The variation of $\Delta v_l/v_l$ as a function of temperature for the complex relaxation peak in B(Pb)SCCO high-T_C superconductor.

3.6. *Interpretation of the relaxation peaks in YBCO and B(Pb)SCCO*

P_1, P_2 and P_3 in YBCO are believed to be complex peaks, which may be attributed to thermally activated atomic jumps (probably over distances appreciably less than the interatomic distance [17]). The broad peak P_4 is probably caused by lattice readjustment or instabilities due to ordering of the oxygen atoms and the high T_C. This interpretation is based on the calculated values of the activation energy of the relaxation process for each peak, reported earlier [6, 7]. For B(Pb)SCCO, P_4 and P_5 may be components of one peak and may be due to the thermally activated movement of boundaries by interaction with defects (oxygen vacancies) [18]; P_1, P_2 and P_3 are caused by processes that involve holes (or electrons) since they have low activation enthalpy (0.13–0.18 eV).

4. Conclusions

Neutron and γ irradiation of YBCO and B(Pb)SCCO causes the relaxation peaks to increase, indicating that the components become more disordered, *i.e.* similar to disordered crystals (such as neutron- and electron-irradiated quartz crystals [19] which support the tunnelling model [20]). The similarity between YBCO/B(Pb)SCCO and amorphous SiO_2/neutron-irradiated quartz [21] is confirmed. Since the peak height in the neutron-irradiated superconductor is increased by an average of 120% (as the irradiation increases from zero to 10.8×10^{10} nvt) and γ irradiation causes the relaxation peaks to increase by an average of 50.5% (as the γ dose increases from zero to 12.85×10^{8} γvt), then it can be concluded that neutron irradiation causes more disordering than gamma irradiation. According to Laerman and Vanelstraete [22] tunnelling states are probably situated at the domain boundaries of twin domains induced by neutron irradiation. The relaxation peak temperatures are not affected by neutron or γ irradiation. Finally, the background of the relaxation peaks increases as the neutron and γ dose increases.

Acknowledgments

The authors wish to thank the researcher at the High-temperature Superconductivity Laboratory, Physics Department, University of Bahrain for preparing and testing the two superconductor compounds used in this study. They also wish to thank Professor C.A.L. Becker for reading the manuscript and Mr. M.J. Al-Othman for assisting in the calculations. Finally, sincere thanks are offered to Mrs. Judy Noronha for typing the manuscript.

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