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# The effect of strain on the low-temperature internal friction of $\text{Y}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{7-\delta}$

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## Abstract

The internal friction of partially Sr-substituted  $\text{Y}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{7-\delta}$  (Y123) ceramics was measured by the vibrating-reed method from liquid nitrogen temperature to room temperature at kilohertz frequencies. The intensity of the internal friction peak, which appears around 220 K, decreases upon Sr doping while the peak position shows no systematic change. Our results do not support the previous oxygen order–disorder explanation; we present another approach relating this peak to the crossover in charge-carrier dynamics, and its evolution with Sr doping is discussed in terms of release of internal strain and electron–phonon coupling.

## 1. Introduction

A great number of experiments have been devoted to the investigation of normal-state anomalous behaviour of high-temperature superconductors (HTSC). Among the techniques used, mechanical energy dissipation measurement is a good probe for detecting subtle changes in microstructures and relative dynamic processes. The mechanical energy dissipation in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Y123) below room temperature has been extensively studied—from the low-frequency torsion pendulum [1] to internal friction in the audio-frequency range [2–6] and the megahertz ultrasonic attenuation [7, 8]—together with the corresponding behaviour of the modulus or velocity [9–11]. It is well known that there is a mechanical energy dissipation peak around 220 K, and this peak disappears when a great amount of the oxygen is depleted from the  $\text{CuO}_x$  chain [3–5]. Although many groups have suggested that the 220 K attenuation peak relates to an oxygen order–disorder transition, there is still much debate as to the origin of this internal friction peak.

Recently Anderson *et al* [12] studied the ultrasonic attenuation of melt-textured Y123, and proposed that the origin of the acoustic attenuation peak at 220 K might be a phase transition connected with the formation of a zigzag configuration in the  $\text{CuO}_x$  chain. In view of the similarity in structure of Y123 and  $\text{YBa}_2\text{Cu}_4\text{O}_8$  (Y124), Wu [13] studied the longitudinal and transverse ultrasonic waves in Y124 and related the ultrasonic anomalies to

the interactions between ultrasonic waves and long-wavelength spin excitations. A helium-ion channelling experiment also gave hints as regards a possible phase transition relating to the dynamic behaviour of Y123 in the same temperature range [14]. Meanwhile, there have been many other experiments such as specific heat [15], high-resolution thermal expansion [16, 17], nuclear quadrupole resonance [18], and Raman spectroscopy [19, 20] investigations, which also show anomalous behaviour in this temperature range. Thus it is of great importance to obtain a clear understanding of the anomaly in internal friction at 220 K in order to develop some key insights into the normal state of HTSC.

In order to study the relationship between the oxygen order–disorder process and the 220 K internal friction peak, we have measured the temperature dependence of the internal friction in Y123 with the Ba replaced with Sr. Sr substitution for Ba is expected to suppress the oxygen mobility due to the contraction of the unit cell and consequent compression of the Cu(1)-type sites [21]. If the internal friction peak was really due to an oxygen order–disorder transition, the peak position would move towards higher temperature and the intensity would decrease rapidly on Sr doping, which is contrary to our experimental findings. Thus it seems that the oxygen ordering process could not be solely responsible for the 220 K peak.

## 2. Experimental procedure

Ceramic samples of  $Y(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{7-\delta}$  with  $x = 0, 0.1, 0.2, 0.4,$  and  $0.6$  were prepared by standard solid-state reaction methods. The starting materials, high-purity  $\text{Y}_2\text{O}_3$ ,  $\text{BaCO}_3$ ,  $\text{SrCO}_3$ , and  $\text{CuO}$  powders, were mixed at appropriate ratios and calcined at  $930^\circ\text{C}$  for three days in air with several intermediate grindings. The powders were pressed into rectangular bars before the final calcination, and then oxygenated at  $500^\circ\text{C}$  for 24 hours in flowing oxygen gas.

All samples were found to be single phased by means of powder x-ray diffraction using  $\text{Cu K}\alpha$  radiation. The superconducting transition temperatures determined by four-probe dc resistivity measurement for  $Y(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{7-\delta}$  with  $x = 0, 0.1, 0.2, 0.4,$  and  $0.6$  are 91, 85, 83, 80, and 78 K respectively. The transition widths are 1, 3, 3, 4, and 4.5 K.

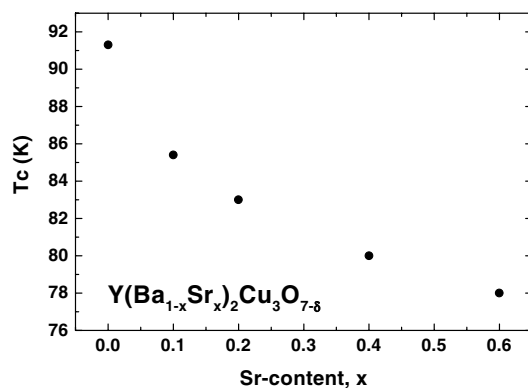
The mechanical energy dissipation,  $Q^{-1}$ , was measured for rectangular bars ( $44 \times 6 \times 0.6 \text{ mm}^3$ ) electrostatically driven in a flexural vibration mode while heating (at rate of  $0.4 \text{ K min}^{-1}$ ) from liquid nitrogen temperature to room temperature in the frequency range 1.5–1.9 kHz.

## 3. Results

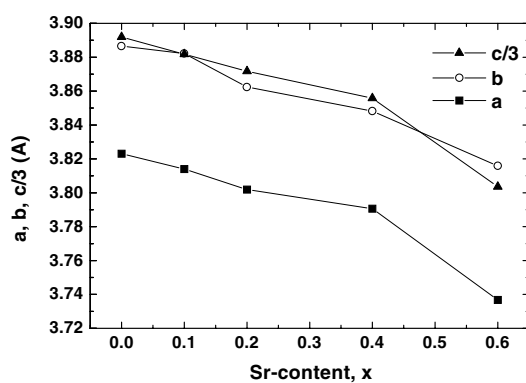
The superconducting transition temperatures are shown for each sample in figure 1, from which we can see that  $T_c$  drops rather slowly compared with the case for Cu site substitution [22]. This provides further evidence for the crucial role of two-dimensional  $\text{CuO}_2$  sheets in high-temperature superconductivity.

The three-dimensional lattice parameters obtained by a least-squares fitting to the x-ray diffraction profiles are shown in figure 2 in relation to different Sr contents. The data indicate that all lattice parameters,  $a$ ,  $b$ , and  $c$ , decrease with increasing Sr concentration. It is expected that such contraction of the unit cell caused by Sr doping would suppress the oxygen mobility, as we will discuss in the next section.

Figure 3 shows the internal friction spectra of  $Y(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{7-\delta}$  with  $x = 0, 0.1, 0.2, 0.4,$  and  $0.6$  below room temperature. In the pure Y123, there are two thermally activated relaxation peaks (labelled P1 and P2) located at 100 and 110 K under our measuring frequency, and another internal friction peak around 220 K (labelled P3), whose position is almost



**Figure 1.** The superconducting transition temperatures for different Sr concentrations in  $Y(Ba_{1-x}Sr_x)_2Cu_3O_{7-\delta}$ .



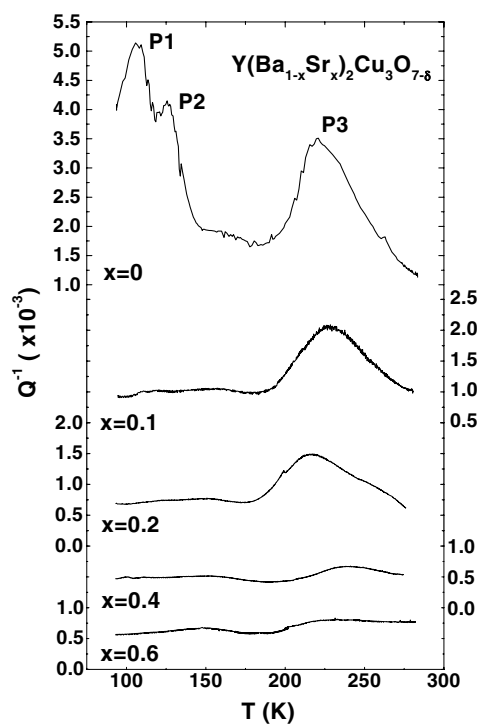
**Figure 2.** Lattice parameters ( $a$ ,  $b$ ,  $c/3$ ) for different Sr concentrations in  $Y(Ba_{1-x}Sr_x)_2Cu_3O_{7-\delta}$ .

insensitive to the vibration frequency as already reported in references [4] and [5]. From figure 3 we can see that the two relaxation peaks (P1 and P2) disappear upon 10% Sr doping while the intensity of P3 decreases much more slowly, as further illustrated in figure 4, and there is no systematic change of the peak position for P3 with increase of the Sr substitution.

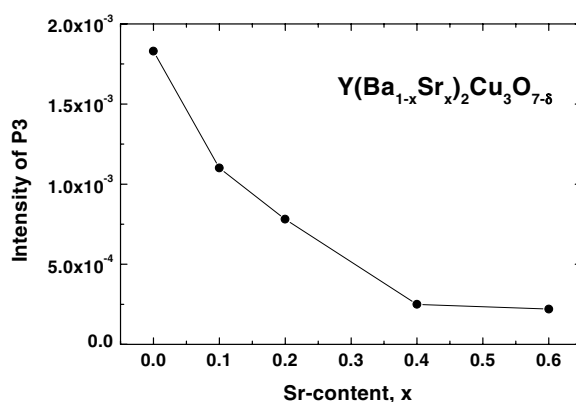
#### 4. Discussion

According to Cannelli *et al* [4], P2 originates from the jumping of chain oxygen atoms O(1) between off-centre positions in a zigzag-chain configuration. Our previous internal friction study on Zn-substituted Y123 [22] seems to conform with this proposal, where P2 remains unchanged on Zn substitution, since  $Zn^{2+}$  ions mainly locate at the Cu(2) sites in the  $CuO_2$  plane and have little effect on chain oxygen atoms. As for P1, possible relaxation species include movement of lattice defects like dislocations [23] and twin boundaries [24], electron or hole (polaron) relaxation [23, 25], and hopping of off-centre atoms [4, 23]. According to reference [22], the jumping of off-centre apical oxygen O(2) in a double-well potential seems more likely.

In this work, the substitution of Sr for Ba in Y123 results in a reduction in the lattice parameters, and such an effect is equivalent to a pressure loading of approximately 10 GPa/ $x$ ,



**Figure 3.** Internal friction spectra of  $Y(Ba_{1-x}Sr_x)_2Cu_3O_{7-\delta}$  with  $x = 0, 0.1, 0.2, 0.4,$  and  $0.6$  in the temperature range 85–300 K.



**Figure 4.** The intensities of P3 versus Sr concentration  $x$  in  $Y(Ba_{1-x}Sr_x)_2Cu_3O_{7-\delta}$ . The intensities are derived by subtracting the baseline from each spectrum.

where  $x$  is the Sr concentration [26, 27]. This induces a chemical instability in  $YSr_2Cu_3O_{7-\delta}$  which makes it difficult for the oxygen atoms to get into or out of the lattice, as it does in the Ba counterpart [28]. The stabilization of  $YSr_2Cu_3O_{7-\delta}$  under ambient conditions can be achieved only when part of the Cu is replaced by an element with higher oxygen affinity such as Ti, Fe, Co, Al, and Ga [29]. In the latter case, as well as for specimens prepared under high oxygen pressure, more excess oxygen enters the lattice and the oxygen content is reported to be higher

than 7.0 [30]; i.e., the real chemical formula is  $\text{YSr}_2\text{Cu}_3\text{O}_{7+\delta}$ . This fact suggests that the  $\text{CuO}_x$  chain layers play the role of supporting the lattice, like the BaO layers in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Here, oxygen atoms in the chain layers become more strongly bound with Sr substitution, which is consistent with the Raman results where the phonon frequencies in  $\text{YSr}_2\text{Cu}_3\text{O}_{7+\delta}$  were reported to be larger than those in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  for most Raman modes [30]. Thus we can say that the oxygen mobility in Sr-substituted Y123 is suppressed. Hence the fact that two relaxation peaks (P1, P2) rapidly diminish upon Sr substitution can be understood as being a result of the freezing of oxygen atoms.

As regards the origin of P3, there has been little agreement up to now. At the early stages of Y123 mechanical study, much attention was paid to the thermal hysteresis of elastic moduli below 220 K. However, the absence of such hysteresis in high-density samples shows that the hysteresis was not an intrinsic property of Y123; nor was it closely correlated with P3 [9]. The elastic modulus is sensitive to the grain boundaries, but P3 is insensitive to them, as evidenced by the absence of the grain boundary found in the ultrasonic attenuation study of single-crystal Y123 [12]. Moreover, the possibility that P3 originates from twin boundary relaxation is also excluded by the early study [31]. Under different measuring frequencies, the position of P3 does not show any clear change [4, 5], and thus the possibility of a thermally activated mechanism can be excluded. Further experiments gave evidence that on depleting a great amount of the oxygen from the  $\text{CuO}_x$ , P3 diminished at the same rate as P2 [4, 5]. It is thus proposed that P3 is related to the oxygen ordering in the  $\text{CuO}_x$  chain [4], possibly corresponding to the formation of oxygen vacancy clusters or the ordering of oxygen in some zigzag fashion. That is to say, the oxygen movements between different crystalline sites or the off-centre sites cause the extra attenuation. But the oxygen ordering process, which is related to a large-scale movement, was reported to take place above room temperature [17, 32, 33]. Moreover this ordering process would be expected to be more prominent at lower oxygen content, i.e. in the underdoped range. This seems to be contradicted by the fact that P3 decreases upon oxygen depletion. Also, it is well accepted that the absence of a 60 K plateau in the phase diagram of  $\text{NdBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is caused by the absence of an oxygen ordering process. However, P3 was in fact observed at 250 K in  $\text{NdBa}_2\text{Cu}_3\text{O}_{7-\delta}$  by Mizubayashi *et al* [34]. Furthermore, the structure of Y124 is similar to that of Y123 except that the single  $\text{CuO}_x$  chain in Y123 is replaced by double CuO chains in Y124 with each chain mutually shifted by half of the lattice parameter along the *b*-axis. Thus compared with Y123, the great advantage of Y124 is that the oxygen sublattice is quite stable below 850 °C and there are neither oxygen vacancies nor twin structures in this system. A large anomaly in the attenuation in addition to a simultaneous slight change in slope of the velocity near 260 K were reported for Y124 [13], which resemble in many ways those commonly found near 220 K for Y123. Thus it becomes questionable to attribute P3 to any oxygen ordering on a large scale, or in a zigzag fashion, as there is no evidence showing that the CuO chains in Y124 have a zigzag configuration.

Our internal friction study of Sr-substituted Y123 does not support such an explanation either—since the decrease in oxygen mobility resulting from Sr substitution will shift the peak position towards higher temperature and diminish the peak rapidly, which is not what is observed in our results. Considering the fact that upon depleting a great amount of the oxygen from the  $\text{CuO}_x$  chain, both P3 and P2 decrease at the same rate, it seems appealing to attribute the origin of P3 to the formation of a zigzag-chain configuration at low temperature. But in our results, the out-of-phase variation of P3 and P2 with Sr substitution casts doubt on this explanation. One must also bear in mind that upon oxygen depletion not only is the configuration of the oxygen sublattice modified, but also the behaviour of the charge carriers changes. So, in view of all the facts mentioned above, it is more reasonable to connect P3 with the dynamic behaviour of charge carriers.

In the Y124 system, P3 seems to locate around the temperature where the pseudogap opens, as determined by the temperature dependence of the spin component of the Cu(2) Knight shift and spin–lattice relaxation time, which reflect the spin dynamics near  $q \simeq 0$  and  $q \simeq (\pi, \pi)$  respectively. Wu [13] suggested that it is the coupling between ultrasonic waves and long-wavelength ( $q \simeq 0$ ) spin excitation that is responsible for the decrease of attenuation below 260 K. A problem will emerge when we apply this assumption to Y123, for the magnetic correlation enhances with lowering of oxygen content while the intensity of P3 decreases [4, 18], and the peak temperature appears too high for the opening of a pseudogap in near-optimally doped Y123.

It is well known that the interaction between charge carriers and the lattice is of key importance for the superconductivity in conventional superconductors. And for high-temperature superconductors, evidence is accumulating of the importance of the interaction between spin, charge, and lattice in the development of high- $T_c$  superconductivity. Most recent research indicates that large polarons, i.e. free carriers, and small polarons (or bipolarons) coexist in cuprates [35, 36]. Zhang *et al* have studied the thermoelectric power and Hall effect of high- $T_c$  superconductors within this model and provided evidence for the dissociation of the local polarons or bipolarons into free carriers with increasing temperature [37]. The change in slope of the nuclear quadrupole resonance frequency  ${}^{63}\nu_Q$  versus  $T$  around this temperature in Y123 also confirms that there is a crossover in the electric behaviour [18]. In Y123, it is suggested that the free carriers dominate above 220 K [37]. Thus we believe that the mechanical energy dissipation peak P3 may relate to such a crossover. Formation of polarons or bipolarons involves local lattice distortion similar to that of Q3-type Jahn–Teller polarons found in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  [38]. The occurrence of such distortion could contribute to the mechanical energy dissipation.

The BaO layer of optimally doped Y123 is highly strained, as indicated by the calculated bond valence sums obtained for the Ba cation [39]. The strain is, in one way, released by the charge transfer from the chain to the plane. In another way, local lattice distortion correlated with the formation of polarons or bipolarons may also help to release the homogeneous strain in Y123. When Ba was partially replaced by Sr, the (Ba, Sr)–O layer became less strained [27] and the requirement for formation of polarons (bipolarons) was relaxed, and hence the intensity of the internal friction peak P3 decreased with Sr substitution.

## 5. Conclusions

We have measured the temperature dependence of the internal friction in a series of Y123 compounds with Sr partially substituted for Ba. The rapid reduction of two low-temperature relaxation peaks (P1 and P2) shows that it is plausible to relate both of them to the oxygen off-centre movement. The intensity of P3 at about 220 K decreases slowly and there is no systematic change in peak position with increase in Sr concentration. These results do not favour the explanation of the oxygen order–disorder process in the  $\text{CuO}_x$  chain, and an alternative proposal regarding P3 is provided on the basis of the coexistence of small polarons (or bipolarons) and free carriers, in which P3 corresponds to a dissociation of local polarons or bipolarons into free carriers with increasing temperature.

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