Elastic Anomalies in Single-Phase $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$

Q.M. Zhang, H.M. Shao, Y.N. Huang, Q. Chen, H.M. Shen and Y.N. Wang

National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China

Abstract: X-ray diffraction measurements, vibrating-reed technique and tensile tests have been employed to investigate the elastic properties of single-phase $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$. The discontinuities of both Young's sound velocity and linear thermal expansion coefficients for a and c directions have been observed near T_c (133K). Internal friction peaks are found in other two temperature ranges of 150K and 250K. Ferroelastic loops are also observed around 150K. A thermodynamics analysis of the experimental data is presented. The discontinuity in Young's sound velocity at T_c and Gruneisen parameter γ are estimated and in agreement with the experiments.

1. INTRODUCTION

Since the discovery of high-temperature superconductivity in lanthanum-barium cuprates by Bednorz and Muller [1], a lot of measurements of elastic properties have been performed on this class of materials such as La-, Y-, Bi- and Tl- system [2-6], particularly focused on YBCO. Recently, a new family of high-temperature superconductors containing Hg with T_c up to 134K has been discovered [7]. To our knowledge, few elastic studies have been carried out on the Hg cuprates. In this paper, the measurements of elastic properties for single-phase Hg-1223 have been made by using X-ray diffraction, vibrating-reed technique and tensile tests. Some anomalies have been observed near 133K(T_c), 150K and 250K. The results are comparable to those obtained in other cuprates. A relevant thermodynamics analysis on the experimental data is presented.

2. SAMPLES and EXPERIMENTS

The samples were prepared by solid state reaction and short time annealing technique using the mixtures of metal oxide powder HgO , BaO , CaO and CuO with start composition $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$. A detailed procedure can be found elsewhere [8]. Energy dispersive X-ray spectroscopic analysis data of several flat grains of the Hg-1223 sample shows that the cation stoichiometry is in agreement with the proposed chemical formulae. From the electron-microscopy investigations, the volume fraction of pure Hg-1223 was estimated to be at least 80-90%. T_c is 133K as determined by ac susceptibility and dc resistivity measurements, shown in the inset of Fig. 1(a). The X-ray diffraction patterns also indicate a nearly single phase corresponding to the tetragonal structure of space group of $P4/\text{mmm}$ with lattice parameters $a=3.85$ angstroms and $c=15.86$ angstroms.

X-ray diffraction measurements for powder samples were taken on a Rigaku rotating anode X-ray diffractometer D_{max}/RA with monochromatized $\text{Cu K}\alpha$ radiation, and the step width and sampling time are 0.02 degree and 2s/step, respectively.

The method of vibrating-reed with a very low strain amplitude ($<10^{-6}$), which can minimize the influence of some kind of amplitude-dependent peaks, was chosen to measure the internal friction of samples.

Fig. 1 Temperature dependence of (a) Young's modulus sound velocity and (b) internal friction with a resonant frequency $f = 490\text{Hz}$.

Fig. 2 Temperature dependence of lattice parameters a and c . The discontinuities in linear thermal expansion along a and c occur at T_c and 250K .

Tensile tests were performed on Mi-44 micromachine from room temperature to 80K and the curves of stress vs. strain were recorded during cooling process.

3. RESULTS and DISCUSSION

The temperature dependence of the Young's sound velocity is shown in Fig.1(a). It is clear that around T_c there occurs a smeared discontinuity of 110 ppm in sound velocity and 30 ppm in its temperature derivative, as shown in the inset of Fig.1(a). In fact, the anomalous changes near T_c have been noted previously in most studies of the elastic properties of the high- T_c superconducting ceramics and single crystals [2-6, 13]. The internal friction below T_c drops obviously at the turning point of T_c , as can be seen in Fig. 1(b). It is suggested that the drop is associated with the superconducting condensation and the smearing of the superconducting gap structure [9]. In Fig. 2(a) and 2(b), lattice parameters a and c show kinks at T_c . It can be obtained from the data that the discontinuity at T_c in linear thermal expansion $\Delta\alpha$ is $1.5 \times 10^{-5}\text{ K}^{-1}$ for a and $2.0 \times 10^{-5}\text{ K}^{-1}$ for c . The values are larger than those obtained in other high- T_c cuprates [10]. This agrees with the conclusion drawn from pressure studies, which indicate that the Hg-superconductors are highly compressible compared with other high-temperature superconductors [11].

In Fig. 1(b), a weak internal friction peak is observed around 150K . The anomaly, as well as that at 250K , has been reported in other high- T_c cuprates by different groups [2-6, 13]. But the exact temperature varies substantially, ranging from 108K in some YBCO single-crystals to about 160K , depending possibly on the oxygen stoichiometry [12]. In our internal friction experiments, the temperatures 150 and 250K corresponding to the two internal friction peaks have no observable shift when the resonant frequency varies from 200 to 1000 Hz . Thus, we tend to attribute them to phase transitions although their origins are not clear. It is interesting to note that ferroelastic loops are found around 150K by tensile tests, shown in Fig. 3. Similar results are obtained in other cuprates[13]. Therefore, the elastic anomalies at tens of Kelvins above T_c should be viewed as a intrinsic feature for high- T_c cuprates. In Fig. 2(a), a discontinuity in lattice parameter a is observed near 250K . The discontinuity is estimated to be no more than 4×10^{-4} under the limit of the resolution of X-ray diffraction. It confirms that a first-order phase transition occurs at this temperature because Δa is proportional to the entropy change ΔS (see below).

Fig. 3 Curves of stress (σ) vs. strain (ϵ) around 150K. Ferroelastic loops appear at this temperature.

Now we present a thermodynamics analysis for our results. In the vibrating-reed method, the effective Young's modulus of an isotropic sample is given by

$$Y = 3GB/(B+G/3) \quad (1)$$

where B and G are some effective bulk and shear moduli of the effective isotropic medium. A thermodynamics analysis yields the following anomalies in bulk and shear moduli and the strain at the transition temperature [14]:

$$\frac{\Delta B}{B} = -\frac{\Delta C_p}{T_c} B \left(\frac{\partial T_c}{\partial P} \right)^2, \quad (2a)$$

$$\frac{\Delta G}{G} = T \Delta S G \left(\frac{1}{T_c} \frac{\partial^2 T_c}{\partial \sigma_s^2} \right), \quad (2b)$$

$$\Delta \epsilon_\alpha = T \Delta S \left(\frac{1}{T_c} \frac{\partial T_c}{\partial \sigma_\alpha} \right), \quad (2c)$$

where ΔS is the entropy change, σ_s is the shear stress, ϵ_α is the generalized strain, and σ_α is the conjugate stress. The shear modulus change ΔG tends to be zero at T_c because the superconducting transition under zero field is a second-order transition, i.e. $\Delta S = 0$. Therefore, we can derive the relation between Young's sound velocity change and the specific heat jump from (1) and (2a):

$$\frac{\Delta Y}{Y} = \frac{2\Delta V_Y}{V_Y} = -B \frac{\Delta C_p}{T_c} \left(\frac{\partial T_c}{\partial P} \right)^2 \left(1 - \frac{Y}{3G} \right) \quad (3)$$

Using the reported values, $B = 170 \text{ GPa}$ [15], $\Delta C_p/T_c = 60 \text{ mJ/mol}\cdot\text{K}^2$ for single phase Hg-1223 [16], $\partial T_c/\partial P = 2.0 \text{ K/GPa}$ [17], $Y \approx G$, (3) yields $\Delta V_Y/V_Y \approx 95 \text{ ppm}$, which is in agreement with that obtained from our experiments.

We can also estimate Gruneisen parameter γ on the basis of the Mie-Gruneisen relation [18],

$$K_{V0} = \frac{\gamma \rho C_p}{\beta} \quad (4)$$

where $K_{V0} \equiv 1/\kappa = -V_0(\partial P/\partial V)_0$, which is taken as 83.8 GPa , is the adiabatic bulk modulus, i.e. the ambient-pressure moduli in Ref. [11], and β is volume thermal expansion coefficient, which is equal to the sum of linear thermal expansion coefficients along three symmetry axes, is about $5 \times 10^{-5} \text{ K}^{-1}$ in Fig. 3(a) and 3(b). The value of the specific heat can be taken as $1600 \text{ mJ/mol}\cdot\text{K}^2$ at 200 K [16]. Thus, we can estimate from (3) $\gamma \approx 1.8$, compared to 1.5 for LaSrCuO and 3 for YBaCuO [15, 18].

In addition, (2c) can be applied to the anomaly at about 250 K where $\Delta \epsilon_1 \neq 0$. The anisotropic stress derivatives of T_c , $\partial T_c/\partial \sigma_\alpha$ can be estimated if the entropy change ΔS is given. Unfortunately, the value of ΔS is not available at present. Further experiments are required for it.

In summary, We obtain a discontinuity of 110 ppm in Young's sound velocity at T_c for single-phase Hg-1223. Internal friction peaks and anomalies of lattice parameters are observed near 150 K and 250 K . The discontinuity in Young's second velocity estimated by a thermodynamics analysis is in agreement with the experimental data. Gruneisen parameter γ is estimated to be 1.8. Clearly, the identity of the anomalous behavior can only come from single-crystal data.

References

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