

Elastic constants and observation of significant elastic softening in superconducting $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals

Jin Wu, Yening Wang, Pingsheng Guo, and Huimin Shen

Department of Physics, Nanjing University, Nanjing 210008, People's Republic of China

Yifeng Yan and Zhongxian Zhao

Institute of Physics, Chinese Academy of Sciences, Beijing, People's Republic of China

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The complete temperature-dependent elastic constants of superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ ($T_c = 84$ K) are determined from sound-velocity data. The longitudinal modes C_{11} , C_{22} , and C_L (propagating along the [110] direction) monotonically increase as temperature decreases from room temperature to T_c . The shear-mode elastic constants C_{66} exhibit a weak variation ($< 1\%$) with temperature from 250 to 80 K except for three small softening minima around 240, 140, and 100 K. The in-plane shear mode $C' (= [(C_{11} + C_{22})/2 - C_{12}]/2)$ shows not only a relatively small absolute value, but also an overall trend of softening (11%) over a wide temperature range (200–100 K). The softening of the C' mode is consistent with the local-atomic-displacements model proposed from pulsed-neutron-scattering results. A possible relation between the softening of elastic constants and superconductivity is discussed.

I. INTRODUCTION

It is worth exploring whether phonons play an important role in high- T_c superconducting oxides and whether there exist structural instabilities above T_c as in $A15$ alloys.¹ Because elastic constants directly represent the behavior of the long-wavelength acoustic phonons and provide a sensitive probe of various phase transitions, there have been a number of elasticity studies on these materials in the prospect of providing some insight into the mechanism for the superconductivity,^{2–9} since the discovery of the high- T_c superconducting oxides. It was discovered by internal friction and ultrasonic attenuation measurements² that there exist phaselike transition peaks at tens of degrees above T_c and 200–250 K in Y-Ba-Cu-O, Bi-Sr-Ca-Cu-O, and Tl-Ba-Ca-Cu-O. However, most of these studies have been done on polycrystalline ceramic samples. Because of differences in sample porosity, the results for sintered materials vary considerably from sample to sample. It is desirable to make elasticity measurements on monocrystalline samples. Several reports on the elastic properties of superconducting single crystals have been published so far.^{10–22}

In an ultrasonic study of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (2:2:1:2) crystal, the sound velocities of the longitudinal mode C_L along the a - b plane, and the shear mode C_{44} associated with attenuation results were reported by Saint-Paul *et al.*¹⁸ A small softening of the longitudinal mode C_L was observed around the superconducting transition on an overall stiffening curve. Another study on the temperature dependence of Young's modulus and internal friction in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals has been reported by Nes *et al.*²⁰ They have found a discontinuity in Young's modulus around T_c and an observed peak in the internal friction Q^{-1} peak at 145 K. Boekholt *et al.*²¹ measured the room-temperature sound velocities of a

$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystal by means of Brillouin light scattering and determined a complete set of elastic constants from these sound velocity data. In our previous works,²² we reported the velocities and attenuations versus temperature of ultrasonic longitudinal and shear waves, respectively, propagating along the directions in the a - b plane at 10° to the \mathbf{a} and \mathbf{b} axes and the [010] axis in the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals. However, these two directions (propagating along the [010] axis and at 10° to the \mathbf{b} axis) are so close that it is not very reliable to accurately calculate the other C_{ij} 's.

In this work, first we reported the temperature dependence of the values of the sound velocities exactly along the \mathbf{a} axis ($V_{L[100]}$), \mathbf{b} axis ($V_{L[010]}$, $V_{s[010]}^{[100]}$), and the [110] direction ($V_{L[110]}$) belonging to longitudinal (denoted L) and shear mode (s) for single crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, respectively, determined by current ultrasonic measurements. And then, the elastic constants C_{11} , C_{22} , C_{12} , C_{66} , and C' (propagating along the [110] direction with the $[\bar{1}\bar{1}0]$ polarization) are derived from these sound velocities. Finally, implications of these results related to the property of the phaselike transition above T_c and the local distortion around carriers are discussed.

II. EXPERIMENTAL PROCESSES

Full details of the single crystalline $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ preparation via the self-flux method and of sample characterization have been given elsewhere.²³ The sample used for ultrasonic measurements is an irregular thin platelet with the maximum dimensions of $5.0 \times 4.0 \times 1.5$ mm³ in the \mathbf{a} , \mathbf{b} , and \mathbf{c} directions. X-ray Laue technique was used to determine the proper crystal structure as well as the crystalline orientations. The quality of the single crystal was surveyed by pseudo-Kossel diffraction pattern with a divergent x-ray beam. Every \mathbf{c} layer is a fairly

good single crystal, but there is a small tilt angle among c layers. Such defects have no significant influence on our results. The density (ρ) of the sample is $6.6 \text{ (g/cm}^3\text{)}$. Resistivity measurements employing an ordinary dc four-probe method showed that the zero-resistance temperature T_c is 84 K .

For the ultrasonic measurements, the samples were polished lightly to give flat and parallel surfaces. Ultrasonic pulses were generated and detected by x and ac cut (for longitudinal and shear waves, respectively) 5-MHz quartz transducers, bonded to the specimen using Nonaq stopcock grease. An oscilloscope was used to monitor the appropriate elastic wave forms. The round-trip transit time of waves was measured by the standard pulse-echo-overlap technique on a Matec 6600 capable of detecting changes in transit time to 1 part in 10^4 . Good quality ultrasonic signals could be propagated through the sample, there being at least five echoes at room temperature. The rate of temperature change was controlled within approximately 0.5 K/min or slower to ensure that thermal equilibrium was achieved.

We suppose a simple harmonic wave to be traveling through the crystal. The displacement $U_i(\mathbf{x}, t)$ of a point in the solid due to an ultrasonic wave is given by

$$\rho \frac{\partial^2 U_i}{\partial t^2} = C_{ijkl} \frac{\partial^2 U_l}{\partial x_j \partial x_k}, \quad (1)$$

for which a plane-wave solution

$$\mathbf{U}(\mathbf{x}, t) = U_0 \exp[j(\omega t - \mathbf{k}\mathbf{x})]$$

gives the Christoffel equations for a wave propagating along $\mathbf{n}(n_i)$:²⁴

$$\det|C_{ijkl}n_i n_j - \rho V^2 \delta_{ik}| = 0. \quad (2)$$

Here, C_{ijkl} are the elastic constants (when denoted C_{mn} , as used in the text, is the Voigt notation), the n_i 's represent components of the unit wave vector relative to crystalline axes, V represents the sound-wave velocity, and δ_{ik} is the Kronecker delta.

For the orthorhombic structure of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystal, from Eq. (2) we can obtain all elastic constants that are denoted the velocities of various longitudinal elastic modes V_L and shear modes V_s , as described in Eqs. (3)–(10):

$$C_{11} = \rho(V_{L[100]})^2, \quad (3)$$

$$C_{22} = \rho(V_{L[010]})^2, \quad (4)$$

$$C_{33} = \rho(V_{L[001]})^2, \quad (5)$$

$$C_{44} = \rho(V_{s[001]}^{[100]})^2, \quad (6)$$

$$C_{66} = \rho(V_{s[100]}^{[010]})^2, \quad (7)$$

$$C_L = \rho(V_{L[110]})^2, \quad (8)$$

$$C' = \rho(V_{s[110]}^{[1\bar{1}0]})^2. \quad (9)$$

If $C_{11} - C_{22} \ll C_{12} + C_{66}$, C_L and C' can be approximately represented by

$$C_L = \{(C_{11} + C_{22})/2 + C_{12} + 2C_{66}\}/2, \quad (8')$$

$$C' = \{(C_{11} + C_{22})/2 - C_{12}\}/2, \quad (9')$$

and

$$C_{12} = \{2C_L - (C_{11} + C_{22})/2 - 2C_{66}\}. \quad (10)$$

If a wave propagates along $[010]$ with the $[100]$ polarization, on solving Eq. (2) for orthorhombic symmetry we also obtain

$$C_{66} = \rho(V_{s[010]}^{[100]})^2. \quad (7')$$

Therefore, we take these modes, described in Eqs. (7) and (7'), to be symmetry equivalents.

III. RESULTS AND DISCUSSIONS

The velocities of the sound waves propagating along various directions $V_{L[100]}$, $V_{L[010]}$, $V_{L[110]}$, and $V_{s[010]}^{[100]}$ as a function of temperature in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals have been individually measured. At 250 K we obtained the velocities $V_{L[100]} = 4320 \text{ m/s}$, $V_{L[010]} = 4097 \text{ m/s}$, $V_{L[110]} = 4647 \text{ m/s}$, and $V_{s[010]}^{[100]} = 2762 \text{ m/s}$. These values are comparable with those measured by other authors^{15,21} using Brillouin light scattering, in which Baumgart *et al.*¹⁵ measured the sound velocity of the surface resonance of the longitudinal acoustic phonon (V_{LR}) on the (001) plane of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals at room temperature to be $V_{LR}(\mathbf{k}||a-b \text{ plane}) = 4650 \pm 300 \text{ m/s}$ and Bockholt *et al.*²¹ determined the velocities of a Bi (2:2:1:2) crystal to be $V_{LR}(\mathbf{k}||[110]) = 4386 \pm 184 \text{ m/s}$, $V_{LR}(\mathbf{k}||[100]) = 4380 \pm 194 \text{ m/s}$, and $V_{LR}(\mathbf{k}||[001]) = 3413 \pm 212 \text{ m/s}$.

Using Eqs. (1)–(8), the complete set of elastic constants can be determined from our current velocity data. As listed in Table I, the elastic constants at various temperatures for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals are summarized.

A. C_{11} and C_{22}

The temperature dependence of the C_{11} and C_{22} of superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, given in Fig. 1, illustrates the obvious elastic anisotropy in the c basal plane. The observed variation for Young's modulus with

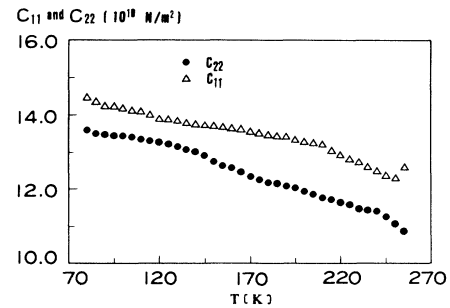


FIG. 1. The temperature dependence of the longitudinal-mode elastic constants C_{11} and C_{22} (in units of 10^{10} N/m^2 or 10 GPa) for the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

TABLE I. Elastic constants of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals in units of 10^{10} N/m^2 (10 GPa).

T (K)	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_L	C'
80	14.49	13.60				5.07	7.74		15.96	3.15
100	14.19	13.44				5.05	8.38		16.15	2.72
150	13.72	12.76				5.03	7.59		15.45	2.82
200	13.31	11.96				5.04	6.58		14.65	3.03
250	12.32	11.08				5.04	6.73		14.25	2.49
297 ^a	12.52		7.58	1.58			7.89	5.60		2.32

^aFrom Ref. 21.

different orientations in the a - b plane¹⁹ also suggested that the a - b plane of crystal $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ has considerable elastic anisotropy. Nomura and Yamada²⁵ found an anisotropy in critical-current densities within the a - b plane of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ single crystals under a magnetic field. The temperature dependence of anisotropic resistivity and Hall coefficient in the a - b plane for a single crystal $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ were observed by Honma *et al.*²⁶ All these are consistent with the fact that an incommensurate modulation appears only along the b axis in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals. Otherwise, both C_{11} and C_{22} show the overall stiffening trend with decreasing temperature.

B. C_{66}

Figure 2 shows the in-plane shear mode C_{66} as a function of temperature, which exhibits three obvious softening minima around 240, 140, and 100 K. Neither a stiffening nor a softening overall trend ($\Delta C/C < 1\%$) between 80 and 250 K can be seen obviously. In our previous works,²² three corresponding internal friction as well as attenuation peaks at the same temperature ranges were observed. The further frequency-dependent measurements²⁸ showed that no detectable shift of the attenuation peak positions could be found, whereas the height of the attenuation peaks is nearly proportional to the square of the frequency, showing the character of phase transitions instead of a relaxation. X-ray-diffraction measurements on the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ powder sample revealed that the lattice parameters have a step change around 140, as well

as 240 K, but with no structural symmetry change.²² A ultralow frequency (0.01–0.1 Hz) experiment^{27,28} carried out by tensile tests manifested that the feature of the internal friction peaks around 100, 140, and 240 K is amplitude dependent and frequency independent, indicating a static hysteretic loss property connected with first-order phase transitions, which results from the stress-induced motion of coherent phase boundaries, quite different from the mechanism of attenuation at MHz. The soft transverse phonon with a polarization vector parallel to the [010] direction propagating along the [100] axis, that is the C_{66} mode, has been observed using thermal diffuse scattering electron-diffraction pattern²⁹ for another cuprate high- T_c superconductor Tl-Ba-Ca-Cu-O ($T_c = 117$ K).

C. $C_{L[110]}$ and C_{12}

In Fig. 3 we show a set of data representing the temperature dependence of longitudinal $C_{L[110]}$ for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals. We find that the feature of the $C_{L[110]}$ as a function of temperature is in agreement with results measured by Saint-Paul *et al.*¹⁸ In order to obtain another important in-plane shear mode C' , we also calculate the independent elastic constant C_{12} from Eq. (10), plotted in Fig. 4.

D. In-plane shear mode C'

As described in Eq. (9), C' denotes an in-plane shear mode of sound waves propagating along the [110] direction with the $[\bar{1}\bar{1}0]$ polarization. The effects of tempera-

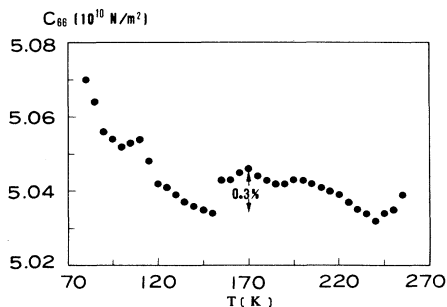


FIG. 2. The temperature dependence of the in-plane shear-mode elastic constants C_{66} (in units of 10^{10} N/m^2 or 10 GPa) for the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

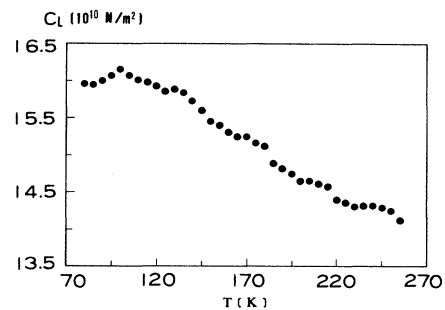


FIG. 3. The temperature dependence of the longitudinal mode $C_{L[110]}$ (in units of 10^{10} N/m^2 or 10 GPa) for the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

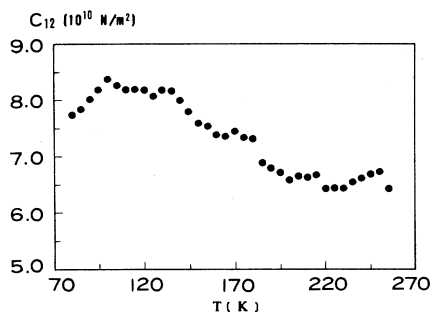


FIG. 4. The temperature dependence of the elastic constants C_{12} (in units of 10^{10} N/m² or 10 GPa) in the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

ture on the C' mode for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals are exhibited in Fig. 5. A striking feature of Fig. 5 is an overall trend of softening (11%) over a wide temperature range (100–200 K). In addition, its value is smaller than that of all other elastic constants. For instance, at 250 K the value of the C' is 24.9 GPa, and that of the C_{66} is 50.4 GPa (see Table I).

We believe that the most interesting result has to be seen in the temperature dependence of the shear mode C' which exhibits a significant elastic softening around 100, 140, and 240 K (much larger than that of C_{66}), just at the position of the corresponding attenuation peaks due to phaselike transition. The feature of these elastic anomalies reflects the property of structural instabilities and the shear-type phaselike transition in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ superconducting crystals. The property of shear type was confirmed^{27,28,30} by the ferroelastic behavior and shape memory effect appearing around the phaselike transition temperatures. Recently there have been several indications for the presence of these structural instabilities or phonon softening in these cuprate superconductors. The experiments of resonant neutron absorption spectroscopy determining the temperature dependence of the average kinetic energy $\langle E \rangle$ of the Cu in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystals have showed a phonon softening of the high-frequency in-plane Cu vibration near T_c .³¹

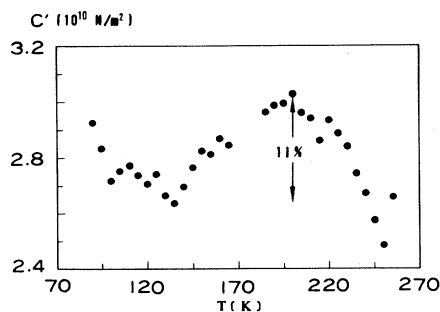


FIG. 5. The temperature dependence of the in-plane shear mode C' (in units of 10^{10} N/m² or 10 GPa) (propagating along the $[110]$ axis with the $[1\bar{1}0]$ polarization) for the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

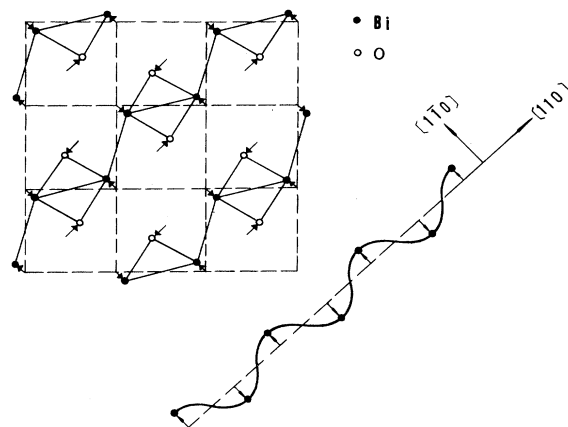


FIG. 6. Schematic representation for the model of the displacements of Bi and O atoms in the Bi-O plane for the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ from their ideal positions (corners and centers of the square) and the introducing periodic shears as indicated by the arrows. The atomic displacements model proposed by Dmowski *et al.* (Ref. 34) and Toby *et al.* (Ref. 33).

Such softening can be produced by a reduction of roughly 10% in the planar Cu-O interactions. Ion channeling in $(\text{Bi}_{1.7}\text{Pb}_{0.3})\text{Sr}_2\text{CaCu}_2\text{O}_y$ single-crystal measurements of the critical angle as a function of temperature revealed an abrupt, anomalous change near T_c in Cu-atom displacements perpendicular to the $[001]$ axis.³² Toby *et al.*³³ found that the actual atomic structure of superconducting oxides has significant local displacement from the crystallographic average structure, and the deviation shows an anomalous change through the superconducting transition by pulsed neutron scattering with the atomic pair distribution function (PDF) method. Monte Carlo simulation of the PDF for Tl (2:2:1:2) indicates that the anomaly is primarily due to the anharmonic displacements of both Tl and O atoms along the $[110]$ direction in the Tl-O plane and of oxygen atoms around Cu by about 0.2–0.3 Å along the c axis (see Ref. 34). They have also suggested that the Tl and coplanar O(3) atoms in $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ are not disordered but have locally correlated displacements that do not exhibit long-range order.³³

All these above-mentioned experimental results seem to be consistent with each other, suggesting that the existence of a local structural change or structural instability above T_c in superconducting oxides. These observations lead us to the question of what the possible explanation for these instabilities existing in these high- T_c cuprate superconductors is. The answer is the geometric misfit of the layers that stack on another.^{35,36} From a structural point of view, the common structural feature of these materials is the presence of CuO_2 planes. Individual CuO_2 planes are separated by metal atoms and are intercalated by a variable number of Bi-O or Tl-O layers. The lattice dimensions within the CuO_2 sheets are basically fixed by the Cu-O distance, which is 1.9 ± 0.1 Å. For the ideal structures, this constraint gives unreason-

ably long Bi-O or Tl-O distances. The mismatch between the dimensions of the CuO_2 and intercalating layers is so large that the structures display an incommensurate modulation or the shear-type phaselike transition. At high temperatures the entropy term can make up for the size mismatch, and some phases can have a small window of thermodynamic stability. At low temperatures these structures are unstable and would not ordinarily form. When these structures are cooling into the region where they are metastable, various complex distortions occur so as to achieve reasonable interatomic distances for some atoms. The softening of elastic constants may also be originated from the interaction of electron-lattice distortion.

Based on the atomic displacements suggested by the PDF studies and the soft-mode C' determined by our ultrasonic measurements, the model of the structural changes may be that both oxygen and bismuth atoms are displaced in the Bi-O plane above T_c as shown in Fig. 6. As a result, the following sequence was suggested: the distorted lattice in the Bi-O plane is formed by introducing periodic shears as indicated by the arrows in Fig. 6. It is considered that the production of the shears is due to the soft mode of the $[110]$ with a $[1\bar{1}0]$ polarization shear-type phonon. Meanwhile, these atomic displacements also affect the planar Cu-O interaction, reflecting a phonon softening of the in-plane Cu or O vibration (see Ref. 31) as well as the out-of-plane displacements (along the $[001]$ axis) of the Cu or O atoms in the Cu-O plane (see Ref. 32). The correlations among the displacements are apparently only short range, since they do not produce observable superlattice diffraction.

In addition, the further studies³⁷ of charge carriers effect on the internal friction plateau or background (Q^{-1}) at kHz for $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_7$ [$\text{Y}(\text{Pr})\text{BaCuO}$] and

$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ have showed that the Q^{-1} increases with the concentration of the carriers. Therefore, the Q^{-1} may be associated with the density of carriers. In view of the results of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{Y}(\text{Pr})\text{BaCuO}$, we proposed that the Q^{-1} may be caused by the distortion cloud induced by carrier just like the polaron but movable with carriers. The shear mode C' may be responsible for the generation of equivalent distortion variants, which have a preferential distribution under an applied stress and result in the mechanical loss Q^{-1} . In fact, the existence of such local distortions has been verified by photoinduced absorption experiments³⁸ and also by neutron-scattering experiments.^{34,35}

IV. CONCLUSIONS

The complete in-plane elastic constants as functions of temperature have been derived from ultrasonic velocity data. The signification softening of the C' mode as well as the C_{66} mode is in agreement with the results found by pulsed neutron scattering and electron diffraction, respectively. The softening minima of shear mode imply the shear-type transition at phaselike transition temperatures. A possible relation between the soft mode C' and the local distortion around carriers or the superconductivity is discussed.

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