A review of elastic properties of high-T_c superconductors and some related C_p results

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First a brief survey is given of what can be learned about important superconducting and normal-state properties by ultrasonic and other elastic measurements. Some of the characteristic elastic properties of the $La_{2-x}(Ba, Sr)_xCuO_4$ and $YBa_2Cu_3O_7$ systems are reviewed. In the La-based family it is shown how the elastic observations are closely related to structural and soft-mode properties. The physics of $YBa_2Cu_3O_7$ is shown to be more complex. Finally, our recent results on the fluctuation contribution to the specific heat near T_c in $YBa_2Cu_3O_7$ are discussed.

1. Introduction

We first outline some of the possibilities offered by measurements of the elastic properties of superconductors and normal metals. We then go on to discuss briefly how ultrasound couples to the structural order parameter. The thermodynamic relation between elastic constant and specific heat is mentioned. Following this review, we discuss the experimental situation in the high-T_c superconducting La- and Y-based families with respect to elastic properties. Recent work on fluctuation effects in C_p is included at the end.

2. A review of possibilities offered by ultrasound in superconduction research

• *Superconducting state measurements*

Ultrasonic methods have a long and important history in the study of superconductors and normal metals. In the hope that they may be applied also to the study of high-temperature superconducting properties, we first review some of these applications. Of particular interest are dynamic measurements, by observation of the attenuation α of sound interacting with the carriers of a metal in the normal (n) or superconducting (s) state. In the original BCS paper [1], confirmation of the theory was announced as far as the temperature-dependent energy gap $\Delta(T)$ was concerned, by use of the simple BCS expression for the ratio of superconducting to normal-state attenuation of longitudinal waves:

$$\frac{\alpha_s'}{\alpha_n} = \frac{1}{e^{\Delta(T)/kT} + 1}. \qquad (1)$$

The quantitative confirmation of the characteristic decrease

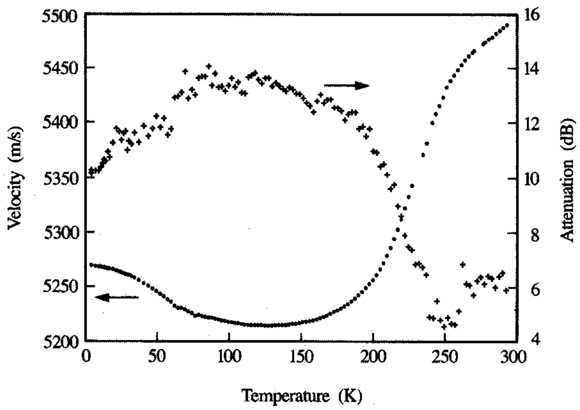

Figure 1

The ultrasonic sound velocity (•) and attenuation (+) measured at 24 MHz in $La_{1.85}Ba_{0.15}CuO_4$ (from [12]).

of attenuation below T_c implied by Equation (1) was therefore one of the first successes of the BCS theory.*

Later work showed [2, 3] that the attenuation of shear waves is governed by the coherence factors very close to T_c (in a 1–2-mK range) and the dynamic screening a little further away. The shear-wave attenuation due to electromagnetic coupling between elastic strain and the charge carriers disappears over only some 10 mK below T_c in a typical conventional superconductor. There are characteristic differences between low-frequency behavior, $q\delta \ll 1$, and high-frequency behavior, $q\delta \gg 1$, where q is the wave vector of sound and δ is the normal-state skin depth. The result given by Fossheim [2] for the ratio of attenuation of shear waves, α_s^s, in the superconducting state to that, α_n^s, in the normal state is

$$\frac{\alpha_s^s}{\alpha_n^s} = \frac{\sigma_{1s}/\sigma_{1n}[q^2\delta^2 + 1]}{[q^2\delta^2 + \sigma_{2s}/\sigma_{1n}]^2 + (\sigma_{1s}/\sigma_{1n})^2}, \quad (2)$$

where the normal-state and superconducting-state conductivities are symbolized by $\sigma_n = \sigma_{1n} + i\sigma_{2n}$ and $\sigma_s = \sigma_{1s} + i\sigma_{2s}$, respectively. Here σ_{2n} was neglected due to the low frequencies used in the corresponding experiments [2]. However, σ_{2n} may easily be included if it becomes important at very high frequency. The temperature and frequency dependence of Equation (2) can be evaluated numerically using BCS theory.

• *Fermi surface measurements*

At the Workshop on High-Temperature Superconductivity, we have been reminded of the need to map out the Fermi surfaces of the new superconductors. Again, ultrasound provides a well-established and accurate method for this, by means of the so-called magnetoacoustic effect [4]: The attenuation of sound as a function of magnetic field B shows oscillations which are periodic in $1/B$. The period $P(1/B_n)$ of these oscillations is directly related to the Fermi surface momentum $k_\perp$ at right angles to $\vec{B}$ and $\vec{q}$, as follows:

$$k_\perp = \frac{2\pi e\lambda}{h} \cdot \frac{1}{P(1/B_n)}. \quad (3)$$

Here λ is the acoustic wavelength, h is Planck's constant, and e is the magnitude of electronic charge. By making successive measurements at different angles of the magnetic field, the so-called effective zones [5] of the Fermi surface may be mapped out, eventually supplying enough information to describe the entire Fermi surface topology provided one has some reasonable idea to start with.

• *The $q\ell$ problem*

The required condition for observing the attenuation of sound due to electronic interaction is $q\ell > 1$, where ℓ is the electronic mean free path.

In the new high-T_c oxide superconductors [6], one would like to perform ultrasonic measurements at about 100 K, where ℓ is necessarily rather small. However, assuming, perhaps optimistically, that ℓ may be of the order of 100 Å, the required $q\ell$ condition will be met at ultrasonic frequencies approaching 100 GHz. This is a formidable frequency, but it is attainable by present-day technology [7]. Ultrasonic studies of the electronic properties hence may have a future in determining the energy gap $\Delta(T)$, the Fermi surface, the mean free path, and even the coupling in these substances. To date, however, no $q\ell$-dependent measurements have been carried out in the oxide superconductors.

• *Structural phase transitions: Statics and dynamics*

As has been well established over many years [8, 9], ultrasound is very sensitive to dynamic as well as static [10] properties of a structural nature. Perovskites have indeed been model systems for such research during the last 20 years. In perovskites the symmetry-allowed coupling between elastic strain ε and the soft-mode order parameter Q, typically representing some rotational motion of oxygen octahedra, is of the type εQ^2. This leads to a characteristic minimum in the static elastic constant (or sound velocity) at the structural transition temperature T_c, and concurrently to a peak in the attenuation [11]. The critical properties of these quantities have shed considerable light on the dynamics and statics of such phase transitions.

• *Thermodynamics at the superconducting transition temperature T_c*

From thermodynamics, one can derive simple relations connecting quantities such as the jump, ΔC_p, in the specific

* In general $\Delta(T)$ is anisotropic, and particularly so in the high-T_c oxide superconductors. Its measured value will therefore depend on the direction of the sound wave.

heat, the pressure dependence of T_c, dT_c/dp, and the shift of elastic constant ΔC at T_c. The fact that ΔC_p is positive on lowering the temperature through T_c requires the elastic constant to show a negative step at T_c. A useful relation is

$$\frac{dT_c}{dp} = \left[\left(-\frac{\Delta C}{C}\right)\frac{T_c}{C \cdot \Delta C_p}\right]^{1/2}, \tag{4}$$

where C is the bulk elastic constant. Attempts to test such relations are currently being carried out on a variety of new superconductors.

3. Experimental results, elastic properties

- *The $La_{2-x}(Ba, Sr)_xCuO_4$ family*

Early measurements [12] of the sound velocity and attenuation of compressional waves in polycrystalline $La_{1.85}Ba_{0.15}CuO_4$ showed unusual behavior. It was found that the lattice softened in a pronounced manner when the temperature was lowered from room temperature to about 190 K (see **Figure 1**). Concurrently, the attenuation increased in the same temperature range. Below this temperature the lattice remained soft (low velocity of sound) all the way down to 63 K, where a return to a stiffer lattice set in. Three temperature regions were thus identified, each carrying a different signature of the dynamics and statics of the lattice. It was concluded [12] that the upper region signified the approach to a structural phase transition, and that the middle region was one in which the lattice remained near instability, i.e., without fully recovering after the tetragonal-to-orthorhombic phase transition near 190 K. The third region, below 63 K, seemed to signal the onset of yet another structural behavior.

During the Workshop on High-Temperature Superconductivity, we have learned about the recent results of the Brookhaven group, as reported here by John Axe [13]. Those results confirm our original interpretation [12]: Corresponding to the three regions identified by ultrasonics, there are indeed three structures (for $x = 0.15$): the tetragonal one, existing from high temperatures down to somewhat below 200 K, a subsequent orthorhombic phase found down to near 60 K, and finally a new tetragonal phase below 60 K. The lower transition is first order, as seen from the symmetry [13].

Soft-mode behavior was also studied [13, 14]: The doubly degenerate mode softening toward approximately 190 K causes lattice instability as well as elastic softening and increased attenuation. This again is fully consistent with the early interpretation of the ultrasonic data [12]. Neutron measurements in La_2CuO_4 [13] show that after the degeneracy is lifted below the tetragonal-to-orthorhombic transition, one of the optical soft modes *remains quite low* over the entire region as far down as measured. This corresponds clearly to the wide temperature range of lattice

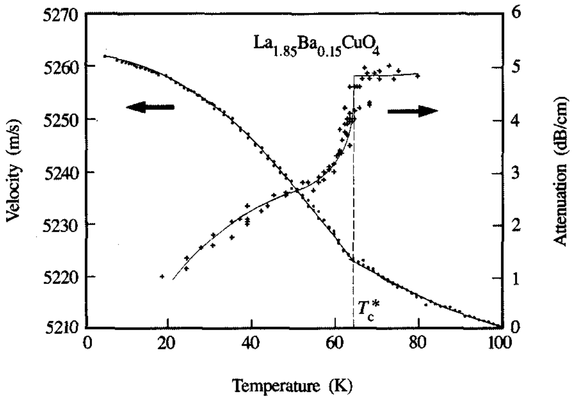

Figure 2

Close-up of the lattice stiffening behavior at 63 K as observed by ultrasound velocity in $La_{1.85}Ba_{0.15}CuO_4$, and the attenuation drop found at the same temperature. Two independent measurements of the sound velocity, one at 11 MHz and the other at 24 MHz, are included in the plot. The attenuation was measured at 37 MHz (from [15]).

instability in this region [12]: A low-lying optical mode corresponds to a system close to structural instability. This explains both the soft elastic constant and the high attenuation in the range from 190 K to 63 K.

Finally, we expect the return to a stiffer lattice at 63 K to correspond to the low-lying optic mode frequency jumping to a higher value. This prediction is made on the basis of careful measurements [15] of the acoustic attenuation near $T_c^* \simeq 63$ K (**Figure 2**). No peak in the attenuation is revealed at this transition, just a drop, as would be expected if the interacting soft-mode frequency jumps to a higher value at a first-order transition.

The physics of $La_{1.85}Sr_{0.14}CuO_4$ appears similar [16], but not identical. From the ultrasonic velocity data again three regions may be seen; hence we expect that even here there must be a third phase at low temperatures, just as in the Ba-doped case. This has not yet been confirmed by neutron or other experiments. Still, two features are at least quantitatively different: (1) The attenuation in the middle region is lower than in the Ba-doped case, and (2) there is a peak in attenuation at T_c^* instead of the drop seen in the Ba-doped case. We predict the following: (1) There is a third phase at low temperature in $La_{2-x}Sr_xCuO_4$, with $T_c^* \simeq 90$ K at $x = 0.14$. (2) The low-lying soft mode in the orthorhombic phase of $La_{2-x}Sr_xCuO_4$ is higher than in the Ba-doped case. (3) The lowest phase transition is either second order, or much closer to second order than the 63 K transition in the Ba-doped case ($x = 0.15$). (4) T_c^* is higher in the Sr-doped case than in the Ba-doped one (for the same value of x).

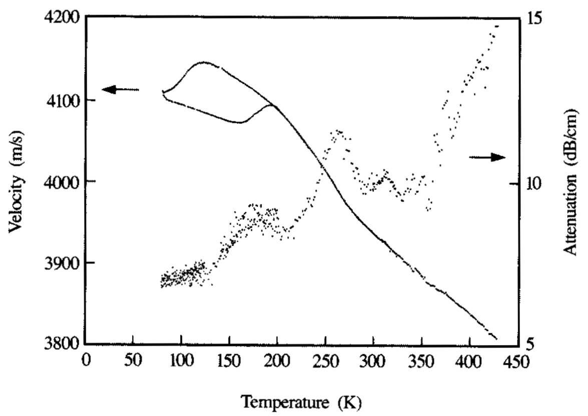

Figure 3

Ultrasonic sound velocity and attenuation in polycrystalline YBCO measured at 11 MHz. Several independent cooling and heating sequences are displayed (from [18]).

Finally, let us point out why the lattice softening occurring at the tetragonal-to-orthorhombic transition can be studied by compressional waves in a polycrystalline, fine-grained ceramic. As mentioned above, the coupling between strain and order parameter is of the general type εQ^2. Analysis of the symmetry properties shows that the coupling to the elastic degrees of freedom will affect the elastic constants $C_{11} + C_{12}$, C_{33}, and C_{66}. All of these quantities will therefore show dynamic and static anomalies at the tetragonal-to-orthorhombic transition. In the polycrystalline material used in [12], all of these elastic constants were probed. Even the C_{66} softening would contribute to the observed softening because compression of the individual grains is off-center, producing a shear in addition to the compression and dilatation.

We conclude that a qualitative understanding of the lattice-related elastic properties of the La_2CuO_4 family has been achieved. What remains is a more quantitative description by means of measurements on single crystals, studying one acoustic mode at a time. Such work is in preparation in a joint project between the authors and Brookhaven.

A number of measurements have been made to show the elastic step near the superconducting transition T_c. At this point we leave this problem for further clarification, i.e., until more precise measurements make possible an accurate examination of the thermodynamic consistency of various types of measurements and of results from different groups.

• *Elastic properties of $YBa_2Cu_3O_7$ (YBCO)*

In contrast to the situation in the La-based compounds, the elastic properties of YBCO appear much more complex. The problems are primarily the following:

1. Lack of a clear correspondence between structural and soft-mode observations on one hand and elastic behavior on the other.
2. Observation of a wide temperature range of elastic instability in ceramic material, probably related to the microstructure [17]. The instability has been found to persist [18] even down to LHe temperature.
3. Difficulty in observing the purely electronically driven shift of elastic constant near the superconducting transition. This is due to a very low value of dT_c/dp (the pressure dependence of T_c), as well as to additional effects discussed below.

In the following we outline the situation, beginning with the highest temperatures.

• *The tetragonal-to-orthorhombic transition*

The only simple case, as far as elastic properties in $YBa_2Cu_3O_7$ are concerned, seems to be the well-known tetragonal-to-orthorhombic transition. Depending on the oxygen concentration, the transition temperature may vary over several hundred degrees [19]. Tallon et al. [20] have carried out mechanical resonance experiments demonstrating that the phase transition can be sensitively recorded. Sharp minima, about 20% deep, were reported in the elastic moduli G and E. The results are typical for the εQ^2 coupling. The mechanical loss factor Q^{-1} showed a broad and strong maximum *below* the transition temperature which was ascribed to oxygen hopping [20].

• *The lattice instability in the 0–300 K range*

A number of elastic measurements have been carried out in this range. An example is shown in **Figure 3**. Many experiments reveal pronounced hysteresis of varying strength in the elastic properties.

Because of the lack of microscopic clarification of the detailed mechanism underlying the elastic hysteresis, various proposals have been made as to its origin. We have noticed the fact [18, 21] that the peak of attenuation, as well as the midpoint of the elastic stiffening on cooling from room temperature, appears to move with frequency. When the frequency is changed from the kHz range to the 10-MHz range, these features appear to shift from about 225 K to about 260 K. A similar situation is encountered in structural glasses. The possibility therefore seems to exist that a broad range of relaxation times are present, as in glasses. In our experiments, the shortest probing time is 10^{-7} s at 10 MHz, and the longest is a "settling time" several hours long, observed in the elastic hysteresis region of Figure 3. These observations, then, span dynamics of about 10 orders of magnitude in probing time, or frequency. It was suggested that the observations could be connected with some kind of ordering of the oxygen system. Further discussion of these points is found in [22].

A somewhat different view [21] takes the elastic behavior to signify a structural phase transformation at about 240 K. We do not feel that these issues can be resolved by data available at this time.

It is interesting to note that theoretical studies do in fact point to an intrinsic mechanism for a lattice instability. Oxygen ordering is the key issue here [23, 24]. In one case [23], two different orthorhombic phases are predicted, while in the other [24], a spinoidal decomposition of the tetragonal phase into orthorhombic and tetragonal regions is found.

• *Specific heat anomalies in the 200–240 K range of ceramic YBCO*

Closely related to the lattice instabilities revealed by elastic and microstructure work are observations of a specific heat peak [25] in ceramic YBCO. This anomaly is observed [26] only on heating, and *only* after the sample has been cooled into the hysteretic region shown in Figure 3. We have interpreted this [18] as a distributed latent heat of the metastable structure, appearing typically below 180 K in our samples. The size of the peak, the connection with the elastic hysteresis, and the absence of the peak on cooling lead to an important conclusion: The specific heat peak is expected to be of structural, microscopic (or microstructural) origin. In addition, we have tested for possible effects from the "addenda," GE varnish and Kapton foil, attached to the specimen. No effects were found. The content of CuO in samples where these effects are invariably found is often less than 1%. Although the absence of the peak during cooling and its connection with the lattice instability constitute proof that impurities or magnetic effects are not responsible, the additional test of CuO content is further confirmation of this.

• *Elastic anomalies near the superconducting transition*

A great number of papers describe a variety of anomalies in the temperature range close to T_c. Although claims [27] have even been made of gap measurements according to Equation (1), this does not seem to be a realistic possibility in view of the required $q\ell$ condition and the MHz frequencies applied.

On the other hand, one cannot in principle rule out the possibility of finding effects similar to those observed in heavy fermion systems where strong electron–phonon interaction is observable even at ordinary ultrasonic frequencies [28]. In a number of studies [18, 27, 29], a drop in the attenuation has been observed below T_c in YBCO. What complicates matters considerably is the following: Several studies find a strong increase in the temperature slope of the elastic modulus just below T_c (see for instance [30]). Furthermore, connected with this behavior is an attenuation *peak* below T_c [29, 30]. It has been suggested [30] that these effects are due to yet another structural instability. This possibility cannot be ruled out, especially in view of the weak anomaly in orthorhombic distortion observed both by means of X-rays [31] and neutrons [32]. Some very surprising observations of large steps, both up [33] and down [34], of the elastic constant on passing through T_c need further clarification. Until more work confirms these unusual features, we refrain from further comment.

Estimates of the expected downward step in the elastic constant at T_c related to dT_c/dp and ΔC_p [Equation (4)] give relative changes of only 10–20 ppm, well below the experimental resolution so far. Our observations, finding no step at all, with a resolution of about 1 m/s in sound velocity, are fully consistent with this.

In conclusion, several puzzles remain with regard to the elastic properties in the range from LHe to about room temperature in YBCO-type materials. Further refined work on the possible structural instabilities in this range is much needed. Hopefully, neutron work or other advanced structural tools can resolve these "mysteries."

• *Elastic measurements on single-crystal YBCO*

Recent measurements on single-crystal YBCO [35] have indicated that the elastic properties of such material may be simpler than those of polycrystals. However, the limited resolution obtained does not permit firm conclusions to be drawn. High-resolution elastic measurements on single crystals are needed.

• *Observations of fluctuation contribution to C_p near T_c*

Of major interest from a basic point of view is the possibility of observing a λ-type anomaly in the specific heat at the superconducting transition. We have recently attempted to observe such phenomena in YBCO [36]. Results of such measurements on a 3-mg monocrystalline sample are shown in **Figure 4**. It is apparent from Figure 4 that these data do not merely show a simple step in C_p. Rather, a λ-like behavior is found, as expected in the presence of fluctuations.

A thorough analysis of the data has been performed using the same background and the same exponent α above and below T_c, and the relation

$$\Delta C_p = A^{\pm} |t|^{-\alpha} + B, \tag{5}$$

where $A^{\pm}$ are the amplitudes above (+) and below (−) the transition, t is the reduced temperature, α is the specific heat exponent, and B is a constant. Equation (5) and more refined versions of critical behavior have been used to analyze for possible critical effects or Gaussian fluctuations [37, 38], similar to the analysis of data near the superfluid transition in liquid He. No BCS step should be included in the analysis.

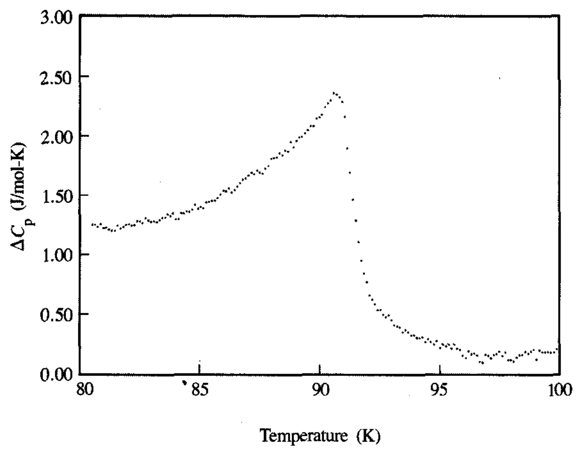

Figure 4

The anomalous specific heat, ΔC_p, at the superconducting transition. A background, $C_b = aT + bT^2$, was subtracted ($a = 1.184$, $b = 0.00195$) from the measured specific heat (from [36]).

Although it turns out to be possible to obtain an effective exponent of $\alpha \approx 0.7$ above T_c, the data do not allow a similar value below T_c, using the same parameters.

Our conclusion is the following: In spite of a much improved signal-to-noise ratio compared to that of Inderhees et al. [39], we find it impossible at this stage to draw conclusions either about the effective spatial dimension *d* or the number of order-parameter components *n*.

This is not to say that (critical) fluctuations cannot eventually be quantitatively described. So far the available crystal quality has, however, not allowed it. Again, we emphasize that fluctuation contributions to C_p *are* observed, but the quantitative aspects are not yet far enough advanced.

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