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VELOCITY AND ATTENUATION OF STRESS WAVES IN $GdBa_2Cu_3O_7$ NEAR THE SUPERCONDUCTING TRANSITION

by

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We have measured the Young's modulus (E) and quality factor (Q) using a resonant bar method in the ceramic superconductor $GdBa_2Cu_3O_7$ as a function of temperature and magnetic field. In contrast to previous pulse-echo and resonant ultrasound results for $YBa_2Cu_3O_{7-\delta}$, we find no dramatic stiffening below T_c. The associated Q increases smoothly by approximately an order of magnitude from ambient temperature to 100 K. Below T_c, Q is a strong function of magnetic field, suggesting significant attenuation from flux motion in the sample.

Thermodynamic probes have proven to be of value in the study of the superconducting phase transition because the free energy of superconductors can be determined independently of a microscopic formalism. The bulk thermodynamic properties derived from the free energy then constrain a microscopic description and provide a good test for its validity. The sound velocities, or more precisely, the elastic moduli, are particularly valuable because extremely small changes can be detected, on the order of 0.1 parts per million (ppm). This is sufficiently sensitive to enable observation of changes related to variations of the free energy near the superconducting critical temperature T_c.[1]

A more easily observed feature of conventional superconductivity is the rapid decrease in ultrasonic attenuation below T_c.[2] This is related directly to the quasi-particle density, because the principal loss mechanism for sound waves in metals at low temperatures is dissipation from electronic currents induced by the displacement of the ions by the sound field.

The recent interest in oxide superconductors has motivated several ultrasound studies of the compounds $(La\text{-}Sr)_2CuO_{4-\delta}$ and $YBa_2Cu_3O_{7-\delta}$.[3,4,5] Measurements of the bulk modulus B of $YBa_2Cu_3O_7$ show a substantial increase in $|dB/dT|$ below T_c, indicating that there are physical processes associated with the superconductivity that require a more complicated treatment than simple thermodynamic arguments can provide (the standard assumption being that only the difference in free energy of the electrons varies near T_c while the lattice is an unchanging background).

In this Communication, we report on acoustical measurements in sintered $GdBa_2Cu_3O_{7-\delta}$ using resonant ultrasound at frequencies near 320 kHz. This low frequency circumvents some of the problems, described below, associated with the granular nature of the samples. We achieved quality factors (the Q of the resonance) in excess of 10^3 at 100 K, enabling us to obtain a precision of slightly better than 1 ppm. In contrast, the higher frequencies used in pulse-echo measurements are attenuated heavily, most likely from dissipation and scattering at grain boundaries. Because good sensitivity to velocity changes depends on a large number of echoes, at best 1 part in 10^4 could be detected in our 30 MHz pulse-echo system.

The samples were prepared from the oxide powders Gd_2O_3 and CuO, and barium carbonate ($BaCO_3$). The starting materials were ground and fired twice at 950C, and then pressed into a right circular cylinder (6.25 cm x 6.00 cm) at a pressure of 3 kbar. We were able to achieve 84% density by this method. To ensure parallel faces for the acoustic measurements, the ends were machined with a special fixture on a milling machine. Sample characterization was performed with a Quantum Design[6] susceptometer, from which we found $T_c = 94.5$ K $\pm$ 0.5 K. The Meissner signal was 35% of $\chi = -1/4\pi$. Upon cooling to 10 K in zero field, complete shielding of the field in the interior of the sample was observed.

The transducers were $LiNbO_3$ from Valpey-Fisher,[7] 1/4" in diameter having coaxial electrodes (1/8" active area), with crystal axes oriented to excite the longitudinal modes of the sample. A transducer on one end was used to drive the sample and a second transducer on the other end was used to detect. The transducers were attached with Stycast 1266 epoxy. To prevent epoxy from diffusing in, 2000 Å Cu was evaporated on each end of the sample before the transducers were attached.

In the resonant method, we excite the fundamental compressional mode of the bar, and therefore the sound velocity v is approximately related to the resonant frequency f_0 by

$$\lambda f_0 = v \cong \left[\frac{E}{\rho}\right]^{1/2} \tag{1}$$

where $\lambda = 2L$ (L is the sample length), E is Young's modulus, and ρ is the mass density. This value of v is approximately 20% lower than for a sound beam in an infinite medium.[8] Moderate drive levels (>200 mV) caused some backbending in the tuning curves of the resonance,[9] so we always measured with less than 100 mV applied to the driving transducer. Although many resonances could be driven, we concentrated on the fundamental longitudinal mode which we were able to identify with the help of a pulse-echo experiment at ambient temperature. We obtained v = 4.4 ± 0.2 km/s from pulse-echo, and v = 4.3 ± 0.1 km/s from the resonant experiments. Our transducers could also weakly excite shear resonant modes. The shear velocity we obtained was 2.6 km/s ± 0.1 from the resonant method (no number was obtained from pulse-echo). These values are consistent with some recent static measurements.[10] The correction to the resonant frequency from transducer mass loading is small, reducing the resonant frequencies approximately 3%.[11] Geometry corrections were estimated to be less than 8%.[11]

In the course of our measurements, we often detected small unexplained shifts in resonant frequency. The highly anisotropic and randomly-oriented grains suggest that movement due to strain relaxation at the grain boundaries or alterations of the twin[12] structure at some arbitrary temperature could have caused such effects. We note that our noise floor was well below these jumps. In Fig. 1, we show velocity data for two different samples. The sample used for the upper plot (sample I) was cycled between ambient temperature and 40 K several times. The lower plot (sample II) is of measurements taken during the initial cooldown. We found it typical for the samples to undergo many small "earthquakes" at lower temperatures (~100 K) before stabilizing. This effect became less prevalent with further thermal cycling.

The data shown for sample I was taken during the final run. The 70 ppm discontinuous drop of the resonant frequency at T_c was observed on four separate experimental runs. On two other runs, small jumps occured at many temperatures, masking any effects at T_c. Just below the transition temperature, the resonant frequency f_0 increased at a rate of 30 ppm/K, with the slope decreasing in magnitude with decreasing temperature.

Purely thermodynamic arguments for a second order superconducting transition give, for the discontinuity in the bulk modulus B at T_c,

$$\frac{\Delta B}{B} = -\frac{B}{4\pi}\left[\frac{\partial H_c}{\partial P}\right]^2 \quad ; H = 0 \tag{2}$$

where ΔB is the change at T_c. If uniaxial stress is used as the thermodynamic variable, rather than pressure, in the derivation of Eq. (2), then a similar result is obtained for Young's modulus. For an isotropic solid, B = E/3(1-2σ) where σ = Poisson's ratio. Static measurements[10] suggest that σ = 0.26, so that B = (0.7)E. The thermodynamic critical field cannot be measured easily, so it is more useful to write Eq. (2) as

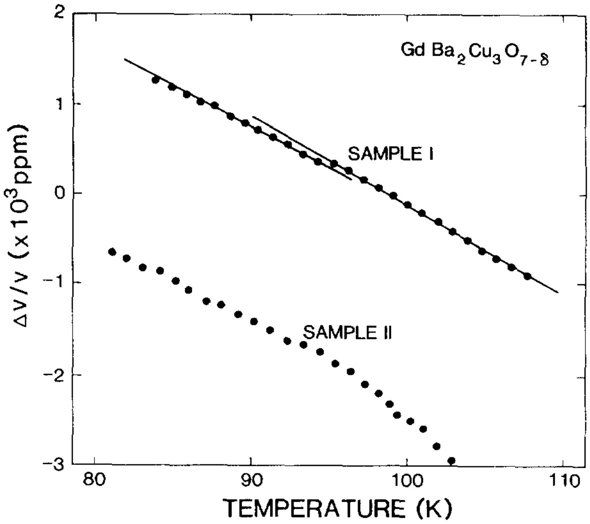

Fig. 1. Fractional change in resonant frequency near the phase transition for samples I and II.

$$\frac{v_s^2 - v_n^2}{v_n^2} \sim -\frac{\Delta C}{T_c} E \left[\frac{\partial T_c}{\partial P}\right]^2 \tag{3}$$

with v_s (v_n) the velocity of sound in the superconducting (normal) state, and ΔC the specific heat jump. Reeves et al.[13] have recently found ΔC = 3 9 J-mole^{-1}-K^{-2} (one mole = one mole formula unit of $GdBa_2Cu_3O_7$). Using the known unit cell parameters from x-ray measurements,[14] one obtains a specific heat jump of $3.7(10)^4$ J-m^{-3}-K^{-1}. Using our value of the bulk modulus at 100 K (B ~ 100 GPa) and $\partial T_c/\partial P$ = 0.113 K/kbar from Borges et al.[16] Equation 3 yields $\Delta v/v$ ~ 50 ppm at T_c. Not forgetting the limitations of polycrystalline samples, our measured shift of 70 ppm is reasonable and consistent with Borges, Reeves, and thermodynamics.

In Fig. 2 we show the temperature dependence of Q, defined as

$$Q \equiv \frac{f_o}{\Delta f_o} \quad , \tag{4}$$

with Δf_0 the full width at half power. For T > 95 K, the change in Q is linear with temperature, at a rate of -39 K^{-1}. Below 94 K, the slope increases dramatically, to -75 K^{-1}. Because both of our samples show this effect, we associate it with the onset of superconductivity. Although an increasing Q with decreasing temperature is expected, we cannot attribute this behavior to conventional BCS superconductivity. In fact, a discontinuity in the attenuation slope is expected for a BCS superconductor.

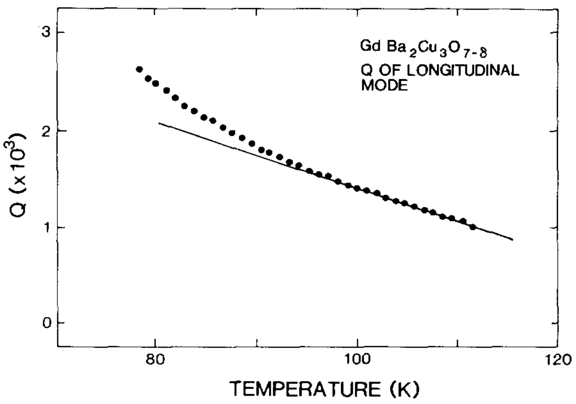

Fig. 2. Temperature dependence of the quality factor Q for the fundamental longitudinal mode of the cylinder for sample I.

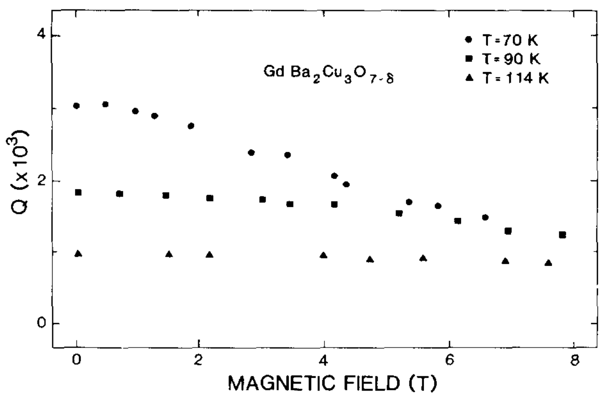

Fig. 3. Magnetic field dependence of Q at various temperatures above and below T_c for sample I.

The high critical temperature suggests that attenuation of the sound wave is mostly from thermal phonons. An estimation of loss indicates that the loss from phonon-electron coupling is only 10% of the loss compared to three-phonon processes. However, the present poor understanding of these materials gives us little confidence in this estimate. Experimentally, however, we note that the estimate is reasonable, because it has been found that the thermal conductivity K increases as the temperature is lowered below T_c.[16] We conclude that either phonon scattering by electrons is significant near T_c or the material is altered when the material becomes a superconductor (e.g. structurally or magnetically).[12,17]

Our original motivation for applying a magnetic field to the sample was to turn superconductivity off, thereby enabling us to subtract the normal background, facilitating the observation of small effects associated with the superconducting state. Because the changes in sound velocity near T_c were small and because the moderate fields (less than 8 T) could not suppress the onset of superconductivity by more than 4 K, we could not measure magnetic-field effects on sound velocity. However, very strong effects were observed for Q. These data are shown for a series of temperatures in Fig. 3. The lowest temperatures show a large drop in Q (more than a factor of three for T = 70 K) in approximately 7 T fields and almost no temperature dependence. The losses are far higher than a linear extrapolation made from the data above 94 K.

In summary, we have observed a small decrease (70 ppm) of the sound velocity in the oxide superconductor $GdBa_2Cu_3O_{7-\delta}$ at the superconducting critical temperature, consistent with other thermodynamic measurements and considering only arguments applicable to second-order phase transitions. This is in contrast to other work on $YBa_2Cu_3O_{7-\delta}$.[3,4] Scatter in our data arising from an intrinsic irreproducibility during the first few cooldowns indicates that the sintered samples are under a great deal of internal stress, which relaxes somewhat during each thermal cycle. Trapped stress and twin structures are commonly found together elsewhere. We find a change in dQ/dT which begins at T_c, perhaps from strong electron-phonon coupling near the transition temperature. Although the magnetic fields we applied do not destroy the superconducting state, a much lower Q is observed in the presence of magnetic fields, suggesting significant dissipation from flux motion, typical of type II superconductors.

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