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Authors
BROWN, SE
MIGLIORI, A
FISK, Z

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VELOCITY AND ATTENUATION OF STRESS WAVES IN GdBa2Cu3O7 NEAR THE SUPERCONDUCTING TRANSITION

by

S. E. Brown, A. Migliori, and Z. Fisk
Physics Division, Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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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 GdBa2Cu3O7 as a function of temperature and magnetic field. In contrast to previous pulse-echo and resonant ultrasound results for YBa2Cu3O7-δ, we find no dramatic stiffening below Tc. The associated Q increases smoothly by approximately an order of magnitude from ambient temperature to 100 K. Below Tc, 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 Tc.

A more easily observed feature of conventional superconductivity is the rapid decrease in ultrasonic attenuation below Tc. 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-Sr)2CuO4, and YBa2Cu3O7. Measurements of the bulk modulus B of YBa2Cu3O7 show a substantial increase in dB/dT below Tc, 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 Tc while the lattice is an unchanging background).

In this Communication, we report on acoustical measurements in sintered GdBa2Cu3O7-δ 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 10^4 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 Gd2O3, CuO, and barium carbonate (BaCO3). The starting materials were ground and fired twice at 950°C, 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 susceptibility meter, from which we found Tc = 94.5 K ± 0.5 K. Upon cooling to 10 K in zero field, complete shielding of the field in the interior of the sample was observed.

The transducers were LiNbO3 from Valpey-Fisher. 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 fo by
\[ \sqrt{\frac{E}{\rho}} = v \]  

(1)

where \( \lambda = 2L \) (\( L \) is the sample length), \( E \) is Young's modulus, and \( \rho \) 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 resonances, 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 \( v = 4.4 \pm 0.2 \) km/s from pulse-echo, and \( v = 4.3 \pm 0.1 \) 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 \( \pm 0.1 \) from the resonant method (no number was obtained from pulse-echo). These values are consistent with some recent static measurements.\(^9\)

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 occurred 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 \( T_c \),

\[ \frac{\Delta B}{B} = -\frac{B}{4\pi} \left( \frac{\partial T_c}{\partial P} \right)^2 ; \quad H = 0 \]  

(2)

where \( \Delta B \) 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, \( B = \frac{E}{3(1-2\sigma)} \) where \( \sigma \) = Poisson's ratio. Static measurements\(^6\) suggest that \( \sigma = 0.26 \), so that \( B = 0.7E \). The thermodynamic critical field cannot be measured easily, so it is more useful to write Eq. (2) as

\[ \frac{\Delta v_s}{v_s} \sim -\frac{\Delta C}{T_c} \left( \frac{\partial T_c}{\partial P} \right)^2 \]  

(3)

with \( v_s \) \((v_n)\) the velocity of sound in the superconducting (normal) state, and \( \Delta C \) the specific heat jump. Reeves et al.\(^{13}\) have recently found \( \Delta C = 3.9 \) J-mole\(^{-1}\)-K\(^{-2}\) (one mole = one mole formula unit of GdBa\(_2\)Cu\(_3\)O\(_7\)). Using the known unit cell parameters from x-ray measurements,\(^14\) one obtains a specific heat jump of \( 3.7(10^4) \) J-mole\(^{-1}\)-K\(^{-1}\). Using our value of the bulk modulus at 100 K \( (B \sim 100 \) GPa) and \( \Delta T_c/\Delta P = 0.113 \) K/kbar from Borges et al.\(^{16}\) Equation 3 yields \( \Delta v/v \sim 50 \) 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

\[ Q \equiv \frac{\Delta f}{\Delta f_0} \]  

(4)

with \( \Delta f_0 \) the full width at half power. For \( T \geq 95 \) K, the change in \( Q \) is linear with temperature, at a rate of \(-39 \) K\(^{-1}\). Below 91 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.
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 \( T_c \). 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).

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 \( \text{GdBa}_2\text{Cu}_3\text{O}_{7-\delta} \) 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 \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \). 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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