Ultrasound studies of the high- T_c superconductor La_{1.85}Sr_{0.15}CuO₄

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Sound-velocity, attenuation, and thermal-expansion measurements are reported on La_{1.85}Sr_{0.15}CuO₄. From room temperature to just above T_c the sound velocity is found to soften significantly as the temperature is reduced. At T_c there is a drop of \sim 150 ppm in accord with the thermodynamics of the superconducting transition, and below T_c , as the gap opens, there is a large increase in the sound velocity. The magnitudes of these effects are anomalously large.

The recent high interest in the high- T_c superconductors¹⁻³ has been fueled in part by the discovery of bulk superconductivity at 40 K in $La_{1.85}Sr_{0.15}CuO₄$.³ The potential for even higher bulk transition temperatures might be influenced by our understanding of the relevant mechanism for superconductivity in these materials. In this paper we report on measurements of the sound-velocity attenuation and thermal expansion on high-quality samples of $La_{1.85}Sr_{0.15}CuO₄$. Our data suggest a very strong electron-phonon interaction above T_c which disappears below T_c as the gap opens up. This is in accord with a recent theory of superconductivity in this material.

Our samples consist of sintered pellets of LaSrCu04 of approximately 95% solid density prepared as described in Ref. 3. Our best sample had a T_c as measured by ac susceptibility (see Fig. 3) of 41 K with a width $(10\% - 90\%)$ of ¹ K. Lithium niobate transducers were epoxied to the samples using the technique of Ref. 5. The transducers had a 10-MHz fundamental frequency, were oriented for longitudinal sound, and were polished for overtone operation. Ultrasound could only be propagated at low frequencies (10-30 MHz). At higher frequencies the size of sintered particles became comparable to the wavelength of the ultrasound, and Rayleigh scattering dominated. At 10 MHz good, clean echos were obtained, and the sound velocity C and attenuation could be measured using the standard methods. Ac susceptibility coils were mounted on the sample and the diamagnetic response, sound velocity, and attenuation could be measured simultaneously. Because of the nature of our samples we only measured a sound velocity corresponding to an average of the elastic constants of the tetragonal-orthorhombic K_2NiF_4 structure of the material. As measured by a time-of-flight technique the longitudinal sound velocity of technique the longitudinal sound velocity of $La_{1.85}Sr_{0.15}CuO₄$ was found to be 5×10^5 cm/sec. The thermal expansion data were obtained using the strain gauge technique described in Ref. 6.

In Fig. ¹ the sound velocity from 50 to 250 K is shown. The behavior is dominated by a softening of the lattice down to \sim 120 K, below which hardening sets in. The feature at 175 K can be associated with the tetragonal to orthorhombic structural phase transition investigated by Fleming, Batlogg, Cava, and Rietman.⁷ The nature of the softening, however, is not known and is unusual for metals, which generally harden upon cooling. We will return to this point later.

In Fig. 2 the sound velocity and thermal expansion data over a temperature range which includes T_c are shown. The most startling aspect of the data is the sharp dip in the sound velocity at T_c . Above T_c the sound velocity shows a marked slowing from \sim 100 to 40 K; the change is \sim 800 ppm. This occurs in a region where the thermal contraction changes by only 75 ppm. Therefore the changes are due to real changes in sound velocity and not a sample size change. At T_c this slowing stops and dC/dT reverses sign and, finally, at low temperatures C attains a value similar to that at \sim 100 K. This result is surprising both in its temperature dependence and magnitude. These are very large changes in sound velocity to be associated with the electronic properties of a metal.

In Fig. 3 an expansion of the data near T_c is shown. At T_c , as measured by the ac susceptibility, there is a drop in sound velocity of \sim 150 ppm. The thermodynamics of the phase transition demands that this change should be there, although in $LaSrCuO₄$ its magnitude is 5-10 times larger than seen in ordinary superconductors. The magnitude of this jump should be given by 8

$$
\frac{\Delta B}{B} = \frac{B}{4\pi} \left(\frac{\partial H_c}{\partial P} \right)^2.
$$

Using $V_c = \sqrt{B/\rho}$, $\rho = 7.4$ gr/cm³, and the sound velocity

FIG. 1. The sound velocity is shown from 250 to 50 K for $(LaSr)$ ₂CuO₄.

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FIG. 2. The thermal expansion and sound velocity are shown as a function of temperature. Note the large change in slope at T_c .

as measured we estimate an average value of $B \sim 1.85 \times 10^{12}$ dyn/cm². Using

$$
\frac{\partial H_c}{\partial P} = \frac{H_c}{T_c} \frac{\partial T_c}{\partial P}
$$

with $H_c = 4.5$ kG, $T_c \approx 37$ K, and $\partial T_c / \partial P = 0.4$ K/kbar (Ref. 9) we can then estimate $\Delta V/V$ as \sim 175 ppm at the transition. This estimate agrees well with our measured value.

One possible explanation for the low-temperature rise is given by a recent theory⁴ of the superconducting transition in this material which suggests that the phonons are very strongly coupled to the electrons. In the work by Weber it is shown that there is a strong electron-phonon coupling between the conduction electrons and the longitudinal acoustic modes in addition to coupling to bondlength changing modes.

An additional clue for understanding this behavior is found in specific-heat and neutron scattering studies¹⁰ of $La_{1.85}Sr_{0.15}CuO₄$ in which a marked enhancement of the phonon density of states is found at 10 meV \sim 115 K. Within this context the observed softening can be understood as relaxing the competition for phase space between optical-like phonon branches at 10 meV and the lowerlying acoustic branches. Above 120 K the optical branches are depopulated by cooling, thus relaxing constraints on acousticlike displacements. Below 120 K more

FIG. 3. The ac susceptibility, thermal expansion, and sound velocity are shown near T_c .

complicated behavior ensues as all phonon branches are depopulated at different rates. Since the nature of the displacements involved in these processes is not known, we can only speculate on the importance of these observations for superconductivity in this system. One possible connection is that the observed softening is a signature of an incipient structural transition involving strong-coupled phonon modes. Softening is interrupted as a gap of $2\Delta \approx 3.5k_BT_c \approx 120$ K opens up in the electron system, thus diverting vibrational degrees of freedom. A more quantitative comparison must await further theoretical work.

We have also measured the sound attenuation through the superconducting transition. We see no evidence for any change at T_c , although, given the high resistivity of the material (\sim 1000 $\mu \Omega$ cm) and low frequency of the experiment, this is hardly surprising.

In conclusion, we have measured the sound velocity, attenuation, and thermal contraction of $Sr_{1.85}La_{0.15}CuO₄$ through its superconducting transition. We find evidence for anomalously strong softening of the sound velocity above T_c and a hardening below T_c as the gap opens up. At T_c there is a drop in the sound velocity in agreement with the thermodynamic parameters of the transition.

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