Acoustic studies of $YBa_2Cu_3O_x$ single crystals

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The temperature dependence of the elastic modulus and that of the absorption of low-frequency sound ($\sim 10^4$ Hz) have been studied in $YBa_2Cu_3O_x$ single crystals over the temperature range 3–250 K. An abrupt change ($\Delta k / k \approx 10^{-4}$) has been observed in the elastic modulus at the superconducting transition temperature T_c. A maximum in the absorption of sound has been observed at $T \approx 120$ K.

An anomalous behavior of the elastic moduli of polycrystalline $YBa_2Cu_3O_x$ samples near the superconducting phase transition has been reported in several papers (e.g., Refs. 1 and 2). Because of the complexities introduced by the anisotropy and the grain boundaries, however, the actual situation can be resolved only by studying single crystals.

In this letter we are reporting a study of the elastic and relaxation characteristics of $YBa_2Cu_3O_x$ single crystals by a mechanical-resonance method over the temperature range 3–250 K.

While we were carrying out the experiments described below, two papers[3,4] appeared with reports of similar measurements. The data of Hoen *et al.*[3] and Shi *et al.*[4] agree qualitatively with our result concerning a softening of the Young's modulus of $YBa_2Cu_3O_x$ at the transition to the superconducting state, but quantitatively the results of all three studies are generally different. The $YBa_2Cu_3O_x$ single crystals were synthesized by spontaneous crystallization from a nonstoichiometric melt.[5] Single crystals of two types were used in the experiments: the original crystals (I), extracted directly from the melt; and samples which had been annealed in oxygen (II). The samples of type I were tetragonal crystals with an oxygen concentration $x \approx 6.3$ and an extremely wide (50–60-K) superconducting transition. It is important to note that the superconductivity of these samples is apparently not a bulk superconductivity and is instead associated with a thin (~ 1-μm) surface layer. The samples of type II ($x \approx 7$) had an orthorhombic structure with a clearly expressed twinning structure and were characterized by a fairly narrow ($\lesssim 1$-K) superconducting transition at $T = 91.8$ K.

The test samples were rectangular plates with dimensions of $0.03 \times 0.3 \times 1.5$ mm. One end of a sample was cemented to a massive copper block, so that the free part of the crystal (about two-thirds of its length) could be used as an acoustic resonator. In the course of an experiment, quarter-wave bending vibrations ($\nu \sim 10^4$ Hz) were excited in the sample by an electrostatic method.[6] We measured the effective Young's modulus Y corresponding to deformations of the crystal in the a-b plane, and we measured the attenuation of sound, δ, as a function of the sample temperature. Some typical results of these measurements are shown in Figs. 1 and 2 in terms of the variables $\nu^2 \sim Y$ and $Q^{-1} \sim \delta$, where ν is the resonant frequency of the resonator, and Q its quality factor.

A comparison of the data in Fig. 1 reveals that the curves of $\nu^2(T)$ for the samples of types I and II are quite different. The slope of the curve for the unannealed sample changes sharply at a temperature ~ 120 K, while the curve of $\nu^2(T)$ for an oxygen-saturated sample is more nearly linear. On the curve for a sample of type II, however, we see a slight anomaly which can be interpreted as a jump in the Young's modulus at temperatures near T_c.

To bring out this anomaly, we approximated the experimental data on $\nu^2(T)$ by

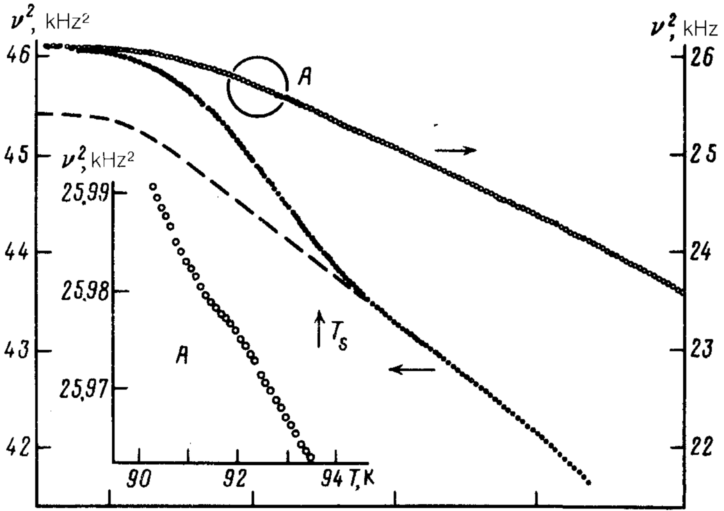

FIG. 1. Temperature dependence of the Young's modulus ($Y \sim \nu^2$) of (●) unannealed and (○) oxygen-saturated single crystals of $YBa_2Cu_3O_x$. Here T_s is the coordinate of the peak on the absorption curve for the unannealed sample (Fig. 2).

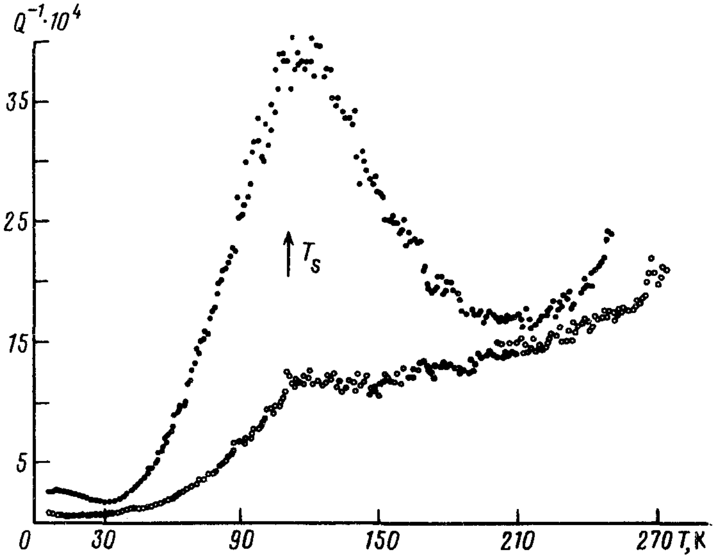

FIG. 2. Absorption of sound ($\nu\sim10^4$ Hz) in $YBa_2Cu_3O_x$ single crystals (the notation is the same as in Fig. 1).

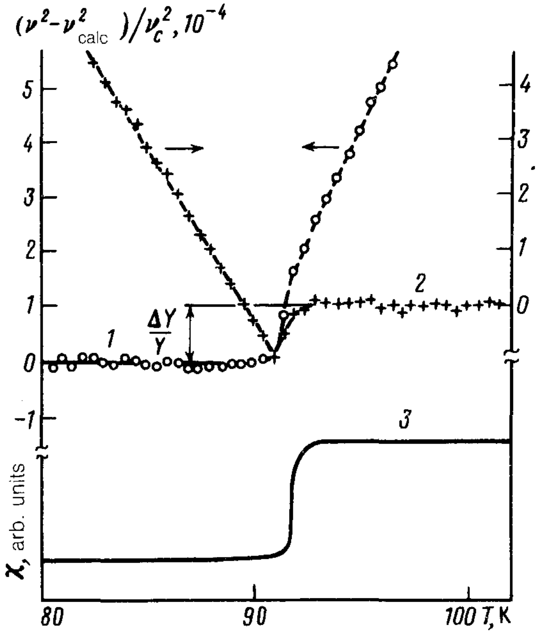

FIG. 3. Relative jump in the Young's modulus, $\Delta Y/Y$, of a $YBa_2Cu_3O_x$ single crystal which had been annealed in oxygen. This jump was found by approximating the experimental data at (1) $T<T_c$ and (2) $T>T_c$. Curve 3—Magnetic susceptibility of the sample near the superconducting transition.

polynomials of third degree in T over limited temperature intervals to the left (70–88 K) and right (93–110 K) of T_c. The results of the corresponding calculations (Fig. 3) show that the Young's modulus of the annealed sample undergoes a change in a comparatively narrow temperature interval (3–4 K), which is essentially the same as the interval of the most rapid change in the magnetic susceptibility of this crystal in the course of the superconducting transition. The relative size of the jump in the Young's modulus is 1×10^{-4}; the mean-square error of the approximation is[1] 1×10^{-5}.

It is interesting to compare the value found for $\Delta Y/Y$ with the result which follows from thermodynamic calculations. Using the Ehrenfest relation, we can write $\Delta Y/Y \approx \Delta k/k = -(dT_c/dp)^2 k\Delta c_p/T$. Substituting the values $dT_c/dp = 0.07$ deg/kbar (Ref. 7), $\Delta c_p = 40$ mJ/cm^3·deg) (Ref. 4), and $k = 2.4$ Mbar (Ref. 9),[2] we find $\Delta Y/Y \approx 5\times10^{-5}$, in fairly good agreement with the experimental value.

Despite some differences, the curves of the sound attenuation in the samples of types I and II (Fig. 2) do reveal a clearly defined peak in the absorption at $T\approx 120$ K. The height of this peak is inversely proportional to the oxygen concentration in the sample. The position of the absorption peak for a sample of type I corresponds roughly to the temperature at which the slope of the $v^2(T)$ curve begins to increase (Fig. 1). Note also the slight maximum in the absorption at $T\approx 8$ K on the absorption curve of this sample.

It appears that the anomalous absorption of low-frequency sound at $T\approx 120$ K is related in one way or another to an ordering of oxygen atoms. A comparison of the absorption curves for the oxygen-saturated and oxygen-deficient samples (Fig. 2) provides some support for this interpretation. In this connection we should call attention to the results of a study[5] of the electrical resistance of $YBa_2Cu_3O_x$ single crystals, which reveals deviations from a linear temperature dependence at $T \lesssim 140$ K.

We note in conclusion that it is not difficult to show that the relative jump in the bulk elastic modulus in the course of a superconducting phase transition would be $\Delta k/k \approx \alpha(\Delta/\varepsilon_F)^2$ according to the BCS model. Here Δ is the superconducting gap, ε_F is the Fermi energy, $\alpha = k/k_{\rm el}$, $k_{\rm el} \approx n\varepsilon_F$ is the electron component of the bulk elastic modulus, and n is the carrier density. Substituting the values $\Delta \approx 0.02$ eV,[3] $\varepsilon_F \approx 0.2$ eV,[4] $n \approx 10^{22}$ cm^{-3}, and $k = 1.2$ Mbar into this formula, we find $\Delta k/k \approx 2.5\times10^{-5}$. This result is fairly close to the experimental value. This approximate agreement may be evidence in favor of the standard (BCS) mechanism for the superconductivity of the cuprate superconductors.

[1] In general, the data of Ref. 8 suggest that the anomalous region of the $v^2(T)$ curve may have a fluctuation component. In order to distinguish it, however, we would need higher-quality samples and a substantially better measurement accuracy.

[2] This value corresponds to a quasi-2D compressional modulus in the *a-b* plane.

[3] Calculated from the values of the plasma frequency.

[4] Found from the results of tunneling experiments.

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