Evidence of low-energy tunneling excitations in the high-T_c superconductor $YBa_2Cu_3O_{7-x}$

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We have measured the internal friction Q^{-1} and the Young's-modulus sound velocity v_E at 1 kHz for $YBa_2Cu_3O_{7-x}$. Below 20 K the relative change in the sound velocity and the temperature dependence of the internal friction perfectly resemble the behavior obtained for Suprasil W. Below 1 K we observe a logarithmic temperature dependence for the sound velocity and between 1 and 5 K a well-defined plateau in the internal friction is obtained. We find quantitative agreement with the predictions of the standard tunneling model which is widely used to explain the low-temperature properties of amorphous materials. The implications of the existence of tunneling systems in these materials are discussed.

Acoustic experiments at audio frequencies are well suited to study different kinds of relaxation processes which affect the sound velocity and internal friction in a great variety of materials. In particular, using the vibrating-reed technique, Raychaudhuri and Hunklinger[1] have shown that the low-frequency elastic properties of different amorphous materials can be well understood with the standard tunneling model (TM).[2] This model, based on the existence of two-level systems with a broad distribution of energies and relaxation times, accounts for a formidable amount of low-temperature data obtained mainly in amorphous materials.[2,3]

Recently,[4] we have used a vibrating-reed technique to study the sound velocity and internal friction at $\omega/2\pi = 700$ Hz in the high-T_c superconductor $La_{1.8}Sr_{0.2}CuO_4$. In this material we have observed a well-defined plateau in Q^{-1} between 4 and 40 K which indicates a relaxation contribution from excitations with a wide distribution of relaxation times. The sound velocity also presents an anomalous behavior, as it decreases between 100 and 20 K having a minimum at $\simeq 20$ K. High frequency measurements in $La_{1.8}Ba_{0.2}CuO_4$ show partially the same behavior for the ultrasonic attenuation and longitudinal sound velocity, but at a higher temperature range.[5] Roughly speaking and taking into account that we are comparing two different materials and samples, the temperature shift of the plateau regime in the phonon attenuation, between the low- and high-frequency measurements, could be explained using the relation $\omega T^{-3} = \text{const}$, which is obtained assuming a relaxation interaction between phonons and tunneling systems (TS).[2,3] Some kind of tunneling excitations has also been proposed[6] to explain the phonon thermal conductivity in $La_{1.85}Sr_{0.15}CuO_4$. All these data and the widely observed low-temperature specific-heat linear term in these ceramics[7] could indicate the existence of low-energy excitations.

In this work we study the low-temperature elastic properties of the 90-K superconductor $YBa_2Cu_3O_{7-x}$ to search for evidence of a possible "glassy" behavior, which could also indicate the existence of tunneling excitations.

Samples of $YBa_2Cu_3O_{7-x}$ were prepared from the oxides of Y and Cu, and from Ba carbonate. The powders, thoroughly mixed with mortar and pestle, were lightly pressed into pellets, fired to 950 °C in an oxygen atmosphere for several hours, and slowly cooled to room temperature in 12 h. A second batch of samples was prepared with the same method, subsequently ground to a fine powder, pressed again into pellets applying 2.5 Kg/cm^2, and sintered once more, as described above.

The low-frequency measurements were carried out with the vibrating-reed technique described in Ref. 4. We have measured samples with thicknesses between 100 and 200 μm and resonance frequencies between 1 and 2 kHz. Three samples, characterized by electrical resistivity $\rho(T)$, mass density δ, and x rays, have been measured with the vibrating-reed technique. Sample A, from the first batch, showed $\delta = 2.9$ g/cm^3, a superconducting critical temperature (at 50% of the resistive transition) $T_c = 93.0$ K, superconducting transition width (10% to 90% of the transition) $\Delta T_c = 2.0$ K, and a ratio $\rho(270\ K)/\rho(100\ K) = 1.43$. For sample B, of the second batch, we obtained $\delta = 4.8$ g/cm^3, $T_c = 86.0$ K, $\Delta T_c = 3.0$ K, and $\rho(270\ K)/\rho(100\ K) = 1.81$. Sample C was obtained from B after a mild annealing process to remove a small amount of oxygen. This sample showed, within our error, the same density as B, $T_c = 85.6$ K, $\Delta T_c = 3.5$ K, and $\rho(270\ K)/\rho(100\ K) = 1.68$. All the samples showed the well-known superconducting orthorhombic phase as the main component. Small amounts of two parasite phases corresponding to Y_2BaCuO_5 and Ba_2CuO_4 were observed by x rays. Depending on the sample, these two phases were between 2%–3% to 10%–15% per mole of $YBa_2Cu_3O_7$. For samples studied, although they had different quantities of these parasite phases, no qualitative or quantitative differences have been observed in the low-temperature elastic properties, which could indicate a contribution other than from the main superconducting phase.

In Fig. 1 we show the relative change of Young's-modulus sound velocity v_E between 0.15 and 5.0 K. From the lowest temperatures up to 1 K a logarithmic decrease of v_E can be noticed. In Fig. 2 we plot the internal friction as a function of temperature. The internal friction increases between 0.15 and 1 K. Above this temperature Q^{-1} becomes temperature independent up to 5 K. No background contribution from the clamping of the sample

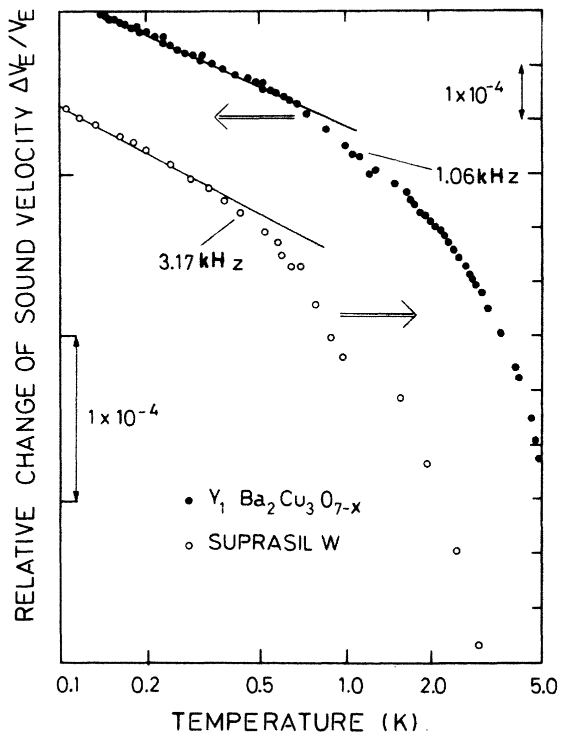

FIG. 1. Relative change of the Young's-modulus sound velocity against temperature for sample A. The data of Suprasil W are from Ref. 1. Note the different ordinate scales.

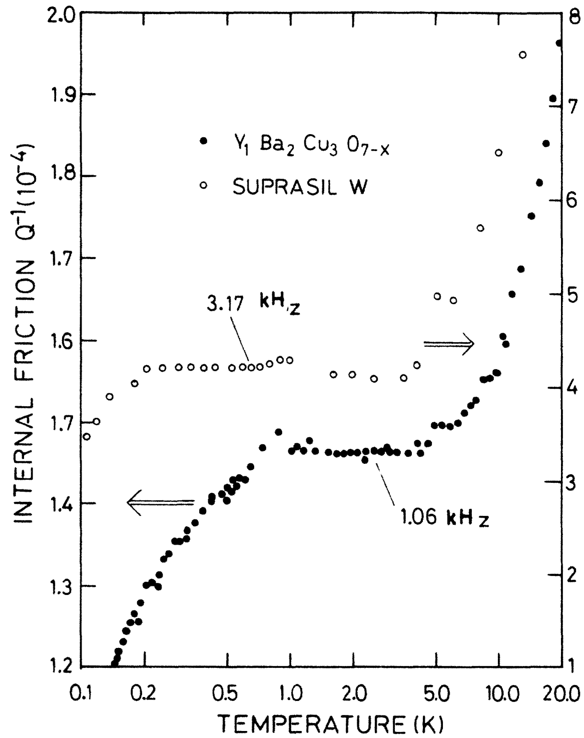

FIG. 2. Internal friction vs temperature for sample A.

was subtracted from the original results. The results obtained from Ref. 1 for Suprasil W, a water-free version of vitreous silica, are also plotted in Figs. 1 and 2. The similarities are evident and synthesize the main result of our report.

The similarities in Q^{-1} and v_E between both materials induce us to explain the observed behavior, as in Suprasil W, with the standard tunneling model. According to this model, and in our measured temperature range (we will assume that $\omega\tau_m \ll 1$ holds above 0.1 K, where ω is our experimental frequency and τ_m is the minimum relaxation time of TS due to the interaction with phonons)[1-3] the logarithmic temperature decrease in v_E should have a slope of $-C/2$, where $C=P\gamma^2/\delta v^2$, P is the density of states of TS, and γ the coupling constant between TS and phonons. From Fig. 1 we obtain $C=7.5\times10^{-5}$. The plateau value of Q^{-1} should be equal to $\pi C/2$. From Fig. 2 we obtain $C=9.5\times10^{-5}$, in good agreement with the TM predictions.[1-3] Samples B and C show the same sound velocity and internal friction behavior as A, characterized by the parameters $C=3\times10^{-5}$ and 6×10^{-5}, respectively. The internal friction of these two samples presents a broader plateau regime below 5 K.

The astonishing similarities with Suprasil W are not only qualitative but also quantitative: The ratio between the logarithmic slopes in v_E (note the difference in the ordinate scale) is exactly the same as the ratio between the plateau values of Q^{-1}. We are aware of the fact that such a perfect quantitative agreement may be partially fortuitous due to the unknown constant clamping contribution (in both measurements). The increase of Q^{-1} at $T>5$ K in Suprasil W is due to a relaxation absorption peak at 31 K (at 3.17 kHz) of still unknown origin. Surprisingly, as in Suprasil W, we also observe an absorption peak at 36 K of the same relative height compared with the plateau value. This high-temperature data will be published elsewhere.

The standard tunneling model could be an oversimplification of the real phenomenon in these crystalline ceramics. The observed large value of the linear term in the low-temperature specific heat C_p (Refs. 7 and 8) seems difficult to obtain from our low-frequency values for C. Taking into account the beginning of the plateau regime in Q^{-1} we obtain smaller values of γ (as is also the case for $La_{1.89}Sr_{0.2}CuO_4$), which would indicate a larger density of states of TS than in amorphous materials. Nevertheless, we obtain values of P 1 to 2 orders of magnitude smaller than C_P provides. This could be due to (a) the linear term could be very sensitive to the oxygen content (note the difference in the linear terms reported in Refs. 7 and 8); (b) C_P should be sensitive to all excitations which contributes to the linear term; (c) although the TM assumes no energy dependence in the coupling constant between TS and phonons, this simplification may not be valid in the ceramic materials. It is possible also that the low-frequency measurements test a lower scattering effective number of TS than observed by C_P measurements.

It is very tempting to try to correlate the origin of TS with some oxygen-related features, e.g., oxygen vacancies coupled to particular vibrating modes. The increase in the parameter C with a slight reduction of T_c from sample B to sample C indicates such a possibility. Moreover, new internal friction measurements in $La_{1.8}Sr_{0.2}CuO_4$ showed a large increase of the plateau value when the sample was annealed in vacuum, thus, with a lower oxygen content.[9]

In the case of $La_{1-x}Sr_xCuO_4$, oxygen is important for the superconductivity, as made evident by the recently discovered isotope effect.[10] We observe that oxygen vacancies in $YBa_2Cu_3O_{7-x}$ could be related to the amount of TS, and also could be important to trigger superconductivity. Nevertheless, no oxygen isotope effect has been observed in this ceramic.[11,12] It is possible that the vacancies are in the chains,[13] while superconductivity would be more related to the CuO_2 planes.[14] If this were the case, the vacancies could be held responsible for the observed twinning of the crystals. The multiple grain boundary generated by this microtwinning can be the origin of the TS.

Localized oxygen vacancies could create regions of the $YBa_2Cu_3O_{6.5}$ phase or perhaps favor some other type of local crystallographic distortion. The distortions induced by the disorder oxygen vacancies do not need to have long-range order, i.e., it would be like a polaronic or a CDW glass. This indeed happens with a certain class of perovskite ferroelectrics.[15] These materials undergo an anomalously smooth transition to a ferroelectric state showing at low-temperatures properties typical of amorphous systems. If this disordered state exists in our material, however, the structural instability that causes it will probably not be related to the high-temperature superconductivity, as evidenced by the lack of an isotope effect. Tunneling systems due to bipolarons have also been detected in nonmetallic vanadium bronzes.[16] In our case, however, if, as Chakraverty[17] affirms, polarons do not form bipolarons in the square CuO_2 lattice, such tunneling bipolarons can only appear in the CuO_3 chains.

From our measurements we conclude that tunneling entities exist in these ceramics which should be related to the oxygen content. The low-temperature behavior of the sound velocity and internal friction can be understood with the tunneling model. An important conclusion is that the linear term in C_P cannot be trivially attributed to excitations of the resonating valence-bond model[18] and could be partially or totally due to tunneling excitations. Any further analysis must be strictly quantitative.

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