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Tunneling systems in superconducting $YBa_2Cu_3O_7$

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Low-energy atomic tunneling has been observed by acoustic methods in the superconductor $YBa_2Cu_3O_7$ at very low temperature. An estimate of the tunneling-system density of states yields a magnitude sufficient to account for the linear term observed in recent heat-capacity measurements.

The discovery of superconductivity above 90 K in the $YBa_2Cu_3O_7$ system[1,2] has raised questions concerning the mechanism responsible for the high transition temperatures. The inhomogeneous microstructure of these compounds, as well as for the (La,Sr,Ba)CuO_4 series,[3] has made it difficult to infer fundamental electronic and vibrational parameters from thermodynamic, transport, and spectroscopic data. The granular character, the importance of interfacial properties, and the critical role of oxygen stoichiometry lead one to suspect that disorder may be a significant factor in determining the unusual properties. Recent measurements of low-temperature heat capacities C_p have consistently revealed two unusual features: (1) a low-energy vibrational mode of considerable weight centered near 10 meV (Refs. 4-6) and (2) a relatively large quasilinear term.[4-10] The latter contribution has been interpreted generally as arising from normal electrons. A linear specific heat is also a manifestation of the proposed resonating-valence-bond state.[11]

In order to clarify the character of the low-energy excitations in $YBa_2Cu_3O_7$, we have measured the low-frequency sound velocity and damping below 1 K. We find evidence for atomic tunneling processes in $YBa_2Cu_3O_7$. An estimate of the tunneling-system (TS) density of states shows that the TS's are able to exhaust the quasilinear term in C_p. Therefore it does not appear necessary to invoke substantial amounts of normal metal to explain the low-temperature properties. Moreover, we suggest that the tunneling systems involve oxygen and propose experimental tests of their locations.

Preparation of the $YBa_2Cu_3O_7$ samples has been described previously.[2] The material ($T_c = 92$ K) was cut and polished in the form of a rectangular bar and the surfaces were coated with a thin gold film. The sample was fixed at one end and excited into a flexural mode near 2 kHz using electrostatic transducers. The device was attached to the mixing chamber of a ^{3}He-^{4}He dilution refrigerator and could be cooled to 5 mK.

The temperature dependence of the 2-kHz sound velocity below 10 K is shown in Fig. 1. The dominant features are (1) a peak at 350 mK and (2) a log-T dependence below the peak for two decades in T. The log-T term has been studied extensively in disordered normal[12] and superconducting metals[13] and arises from the resonant interaction of sound with low-energy tunneling systems. A pseudo-spin-$\frac{1}{2}$ Hamiltonian describing the TS's and their coupling to phonons and electrons is given by[14]

$$H = ES_z + 2MS_x\epsilon + DS_z\epsilon + \frac{1}{N}\sum_{kk'} V_\perp S_x c_k^\dagger c_{k'} . \qquad (1)$$

The first term describes the static splitting of the zero-point energy of a particle in a double-well potential, the second and third term, the off-diagonal and diagonal coupling to strain ϵ with deformation potentials M and D, respectively, and the last term the interaction with conduction electrons. This term, with electron-TS coupling parameter $V_\perp$, allows for electron scattering from state k to k' by a TS undergoing a spin flip and leads to a greatly enhanced TS relaxation rate[12] as well as enhanced resistivity.[14] Nevertheless, in a superconductor well below T_c the exponentially small quasiparticle density leads to a negligible contribution from this term[15] which is herewith neglected.[16] We shall thus assume that TS decay is determined by a one-phonon process below 1 K, where

$$T_1^{-1}(E) = \frac{M_t^2 E^3}{\pi\rho v_t^5 \hbar^4}\coth(\beta E/2) . \qquad (2)$$

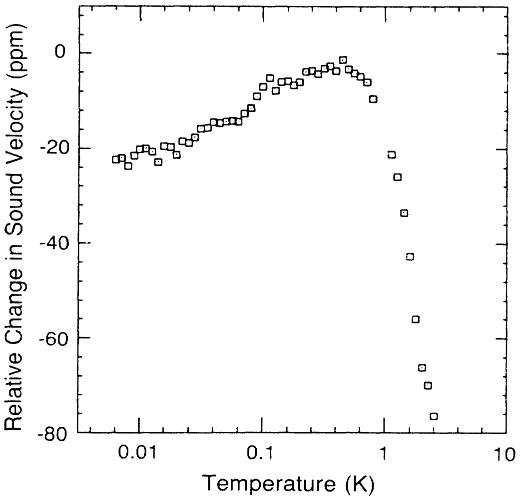

FIG. 1. Sound velocity change (in parts per million) of a 2-kHz flexural mode in $YBa_2Cu_3O_7$ below 1 K. The logarithmic temperature dependence below 0.35 K is the signature of resonant acoustic coupling to tunneling systems.

According to standard TS theory,[17] the second term in Eq. (1) leads to a contribution to the sound velocity

$$\frac{\Delta v}{v} = \frac{\bar{P} M_i^2}{\rho v_i^2} \ln(T/T') \ , \tag{3}$$

where $\bar{P}$ is the symmetric TS density of states and T' is an arbitrary temperature. Therefore the data in Fig. 1 allow us to calculate $\bar{P} M^2 = 5.1 \times 10^6$ erg cm^{-3} using $\rho = 6.3$ g cm^{-3} and $v = 3.8 \times 10^5$ cm s^{-1}. In the absence of coherent techniques which allow an independent evaluation of M,[18] the following approach yields an estimate of the coupling parameters. The third term in Eq. (1) generates a relaxational interaction between the TS and phonons of frequency ω which scales with ωT_1 and which is additive to the resonant interaction discussed above. It is the relaxational term which dominates the velocity change above the peak. Nevertheless, this contribution becomes largely temperature independent at very low temperatures, where $\omega T_1 \gg 1$ and we shall take the velocity peak as signifying $\omega T_1 = 1$. Using the above expression for $T_1^{-1}(E)$ and the values $E = 2k_B T$, $v_t = 2.8 \times 10^5$ cm s^{-1}, $M_1^2 = 2M_t^2$, we find $M_t^2 = 12 \times 10^{-28}$ erg^2 ($M_t = 0.02$ eV), and $\bar{P} = 4.2 \times 10^{33}$ erg^{-1}cm^{-3}. The specific heat of the two-level systems in $YBa_2Cu_3O_7$ is

$$C_p = aT = \frac{\pi^2}{6\rho} k_B^2 \bar{P} T \ . \tag{4}$$

We obtain $a = 1.4$ mJ K^{-2} (mole f.u.)$^{-1}$. Values reported for a (in the above units) are $YBa_2Cu_3O_7$, 3.0,[19] 3.5,[5] and 20;[6] $La_{1.85}Sr_{0.15}CuO_4$, 1.6–5;[5-8] and $La_{1.85}Ba_{0.15}CuO_4$, 3.6–5.[6,9,10] The presence of an additional, possibly magnetic impurity contribution in $YBa_2Cu_3O_7$ has been noted[5,6] and may account for the large value reported in Ref. 6 even in the presence of a 7-T magnetic field. In spite of the uncertainty in our $\bar{P}$ estimate and the variability in measurement and sample preparation methods the agreement is striking.

One aspect of the tunneling process subject to test is the presence of a time dependence to C_p. For a standard distribution function, C_p varies as $\ln t$ and we would estimate an enhancement from the short-time value quoted above by a factor of 5 or so at 1 s. However, the distribution may be unusual; even though the high-T_c superconductors possess significant numbers of TS's they are *not* glasses. For example, the coupling parameter M_1 is two orders of magnitude smaller than in amorphous SiO_2,[18] but only a factor of 6 smaller than in $KBr_{0.5}KCN_{0.5}$,[20] a crystalline orientational glass. In spite of the weak coupling to long wavelength phonons, the large temperature coefficient of the velocity above 1 K, where v is nearly linear in T, indicates strong anharmonicity at energies above a few degrees kelvin.

We have neglected electron-TS interactions. In normal metals the occurrence of a velocity maximum is not associated with the ωT_1 condition but rather with a transition from primarily phonon to electronic relaxation as T decreases. In this case the logarithmic slope of $\Delta v/v$ is $C/2$. However, if fast electronic relaxation dominated, the condition $\omega T_1 \ll 1$ would be satisfied at all temperatures and no decrease in the acoustic damping should occur below the peak. This contrasts with our observation of a small decrease in the damping below 100 mK. Nevertheless, we have noted a relatively large and essentially temperature-independent background damping. It is possible that the background could occur if there were a large TS-electron interaction in a small amount of normal metal.[21]

The most likely candidate for the tunneling species is oxygen. If the distribution is uniform and extends to a typical oxygen annealing temperature, ~500–1000 K, then at least 1% of the oxygen can tunnel. It appears likely that defects in the oxygen coordination are responsible for these modes. In two samples of $La_{1.85}Sr_{0.15}CuO_4$, it was noted that the linear term increased as T_c decreased,[5] with a lower T_c presumably due to incomplete oxygenation. The most convincing test should occur in $YBa_2Cu_3O_{7-x}$, where the linear chain oxygens are particularly mobile. Measurements of the linear term as a function of x, and the degree of oxygen ordering, would be instructive.

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