Title

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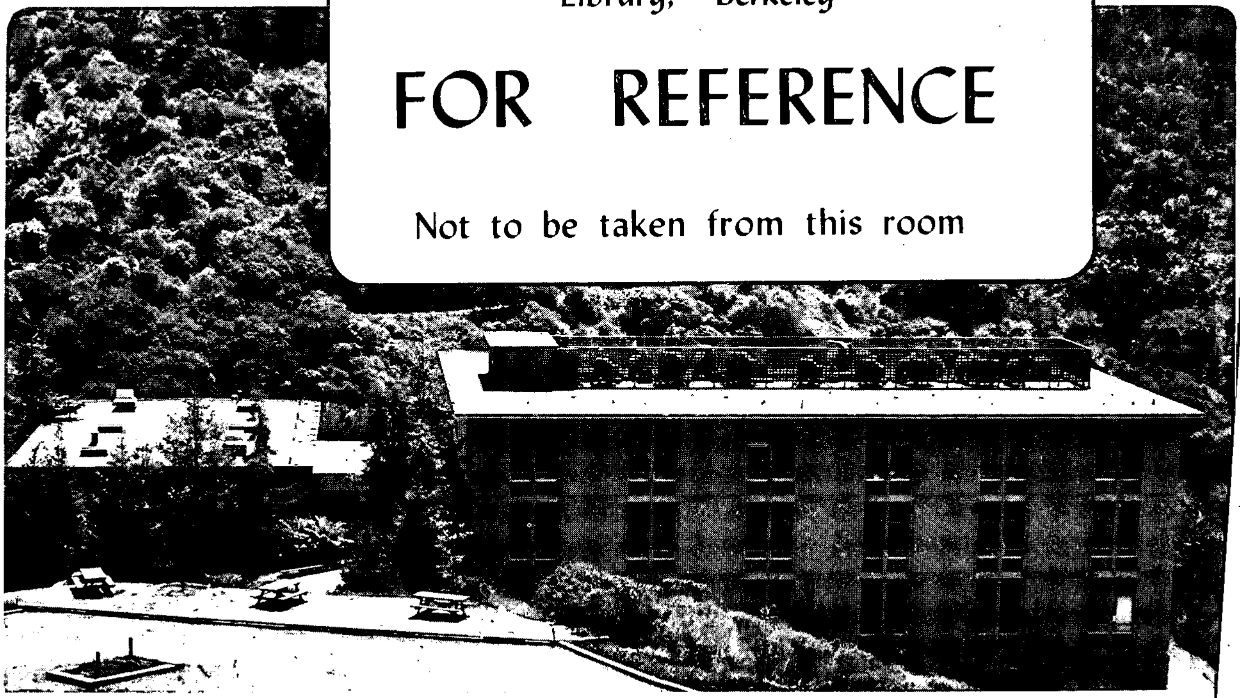

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ABSTRACT

Specific-heat measurements, on polycrystalline samples of $YBa_2Cu_3O_{7-\delta}$, YBCO, have shown sample-to-sample variations in the volume fraction of superconductivity, f_s, which is correlated with the concentration of Cu^{2+} magnetic moments in the YBCO lattice. At low temperatures the lattice specific heat also varies with f_s, but these variations do not persist above ~20K. The low-temperature data show that Θ_o^{-3} varies linearly with f_s, and give values of 520 and 390K for Θ_o for fully-superconducting and "fully-normal" YBCO, respectively. These results suggest that the long wavelength phonon modes are altered when Cu^{2+} magnetic moments are present in the lattice. The fact that different samples have the same lattice specific heat at ~20K and above T_c indicates that the higher energy phonon modes are insensitive to these Cu^{2+} moments.

The size of the anomaly at T_c in the specific heat, C, of polycrystalline $YBa_2Cu_3O_{7-\delta}$, YBCO, as measured by $\Delta C(T_c)$, shows a wide sample-to-sample variation[1,2]. It has been argued[3,4] that this result is evidence for a corresponding sample-to-sample variation in the volume fraction of superconductivity, f_s. Two other quantities[3,4,5] that might also be expected to be proportional to f_s, ΔS, the change in the entropy near T_c produced by the application of a magnetic field, H, and $d\gamma^*/dH$, where γ^* is the coefficient in the linear term in C, are proportional to $\Delta C(T_c)/T_c$ as shown in Fig. 1. These quantities have been used to determine f_s, as is shown in Fig.2a, where each quantity has been suitably scaled and averages of the three have been used to define relative values of f_s. Figure 2b shows that f_s decreases approximately linearly with increasing n_2, the concentration of Cu^{2+} magnetic moments in the YBCO lattice[3,4]. Whether these moments themselves create regions of normal material via a magnetic pair-breaking mechanism, or whether their concentration is simply proportional to some other defect responsible for creating the

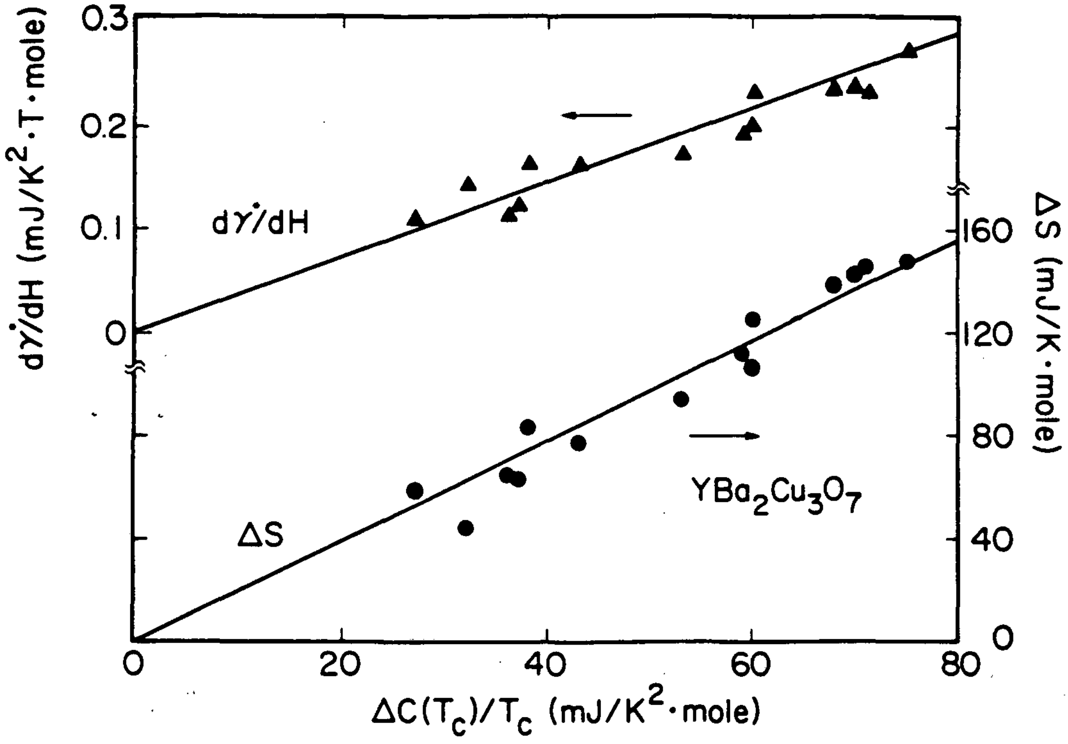

Fig. 1. The correlation of $d\gamma^*/dH$ and ΔS with $\Delta C(T_c)/T_c$.

normal regions, is not known. However, in view of the correlation of f_s with n_2, it is reasonable to associate the limit $n_2=0$ with $f_s=1$, thus establishing absolute values of f_s. The interpretation of the properties of YBCO as reflecting the coexistence of superconducting regions and normal regions associated with Cu^{2+} magnetic moments, first reported in Ref. 3, is supported by very recent NMR measurements[6].

In this paper it is pointed out that Θ_0^{-3}, where Θ_0 is the Debye temperature obtained from the T^3 contribution to the low-temperature specific heat, is a linear function of f_s. Figure 3, a graph of Θ_0^{-3} versus f_s, indicates that $\Theta_0 \approx 520K$ for a fully-superconducting sample of YBCO and $\Theta_0 \approx 390K$ for a fully-normal one. Swenson et al.[7] have reported a variation of Θ_0 with $\gamma^*(0)$, a result that is qualitatively in agreement with the correlation reported here. It should be noted that values for Θ_0 reported in the literature (see the compilations in Refs. 1 and 2) vary from 290 to 510K. While some of these reported variations undoubtedly arise because of the way in which Θ_0 was derived from C, e.g., the assumption of T^3 behavior for the lattice specific heat for $T>5K$ (which results in too small a value of Θ_0), not all of the difference can be explained in this way.

Figure 4a is a graph of C_ℓ/T^3 versus T from ~2 to 20K for two polycrystalline samples [C_ℓ is the lattice specific heat and is equal to $C-\gamma^*(0)T-C_m$, where C_m arises from the magnetic interaction among the Cu^{2+} moments. $C_m \sim A_{-2}T^{-2} + A_{-3}T^{-3} + \cdots$]. It is evident that for both samples the T^3 region does not extend above ~5K. It is also clear from Fig. 4a that whereas C_ℓ is markedly different in the low-temperature region, this difference essentially disappears by ~20K. Figure 4b is a graph of C/T versus T for the same two samples in the vicinity of the anomaly at T_c. It should be noted that above T_c the specific heats of the two samples are the same to within experimental accuracy. Because a larger fraction of one sample (triangles) undergoes

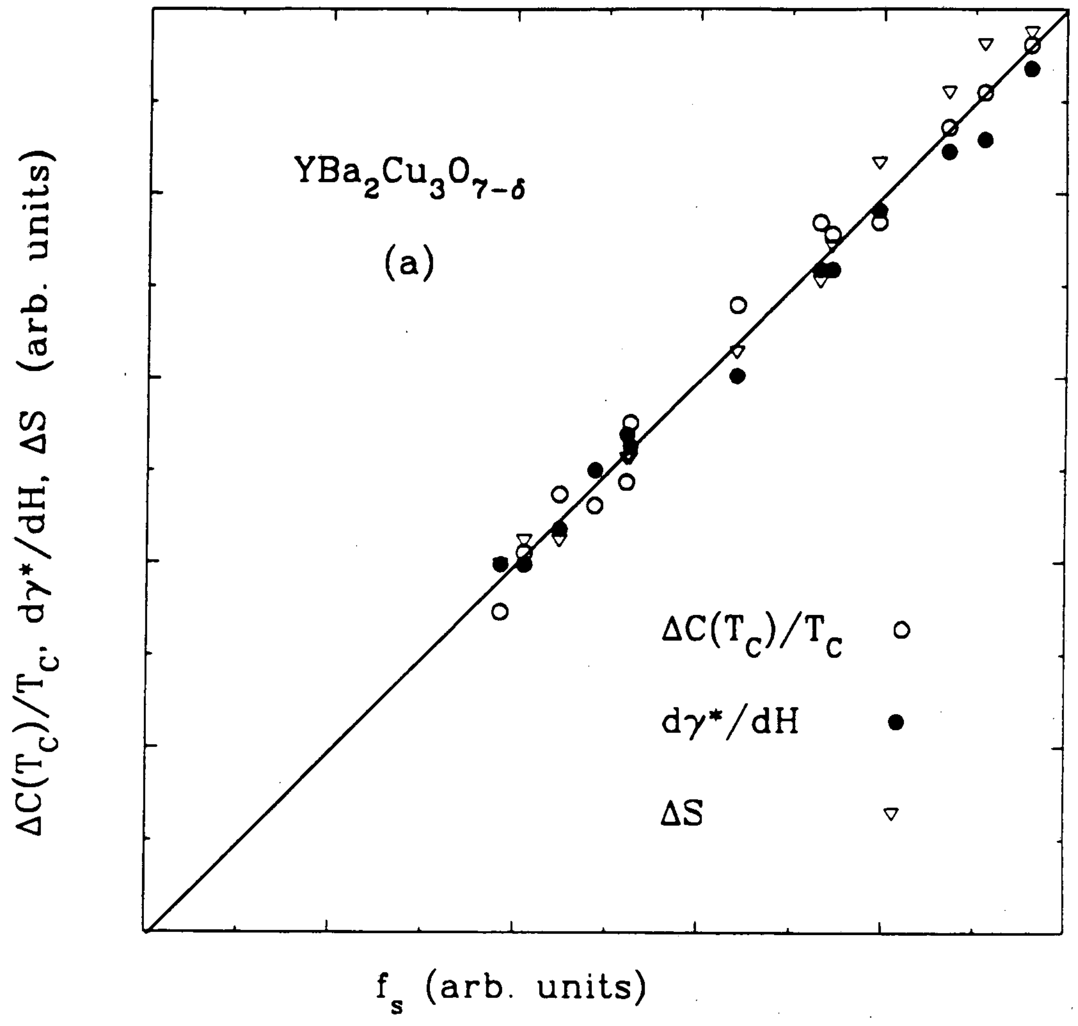

Fig. 2a. The correlations among $\Delta C(T_c)/T_c$, $d\gamma^*/dH$ and ΔS, scaled to fall on the same line, for a number of polycrystalline YBCO samples. For each sample the three quantities are plotted at the relative value of f_s that best represents all three.

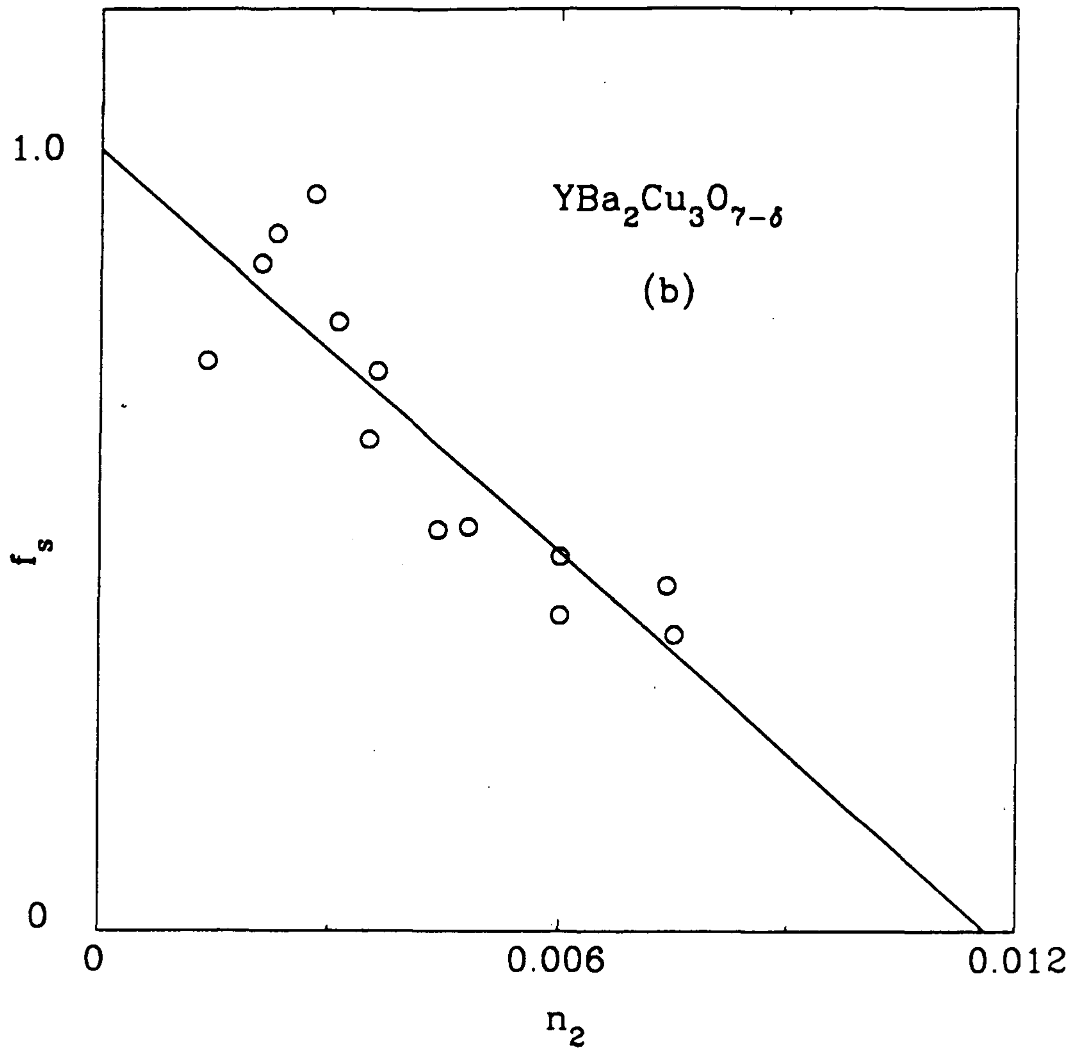

Fig. 2b. The correlation of the relative values of f_s with n_2, which suggests that n_2 measures the concentration of a defect that suppresses the transition to the superconducting state. The association of $n_2 = 0$ with $f_s = 1$ puts the values of f_s on an absolute scale.

the superconducting phase transition, that sample's specific heat is larger immediately below T_c than that of the second (circles), but as T approaches 70K the two specific heats become more nearly equal. There are no data for one of the samples between 22 and 70K. However, since the two specific heats are equal to within experimental accuracy at the lower temperature, it seems reasonable to assume that for temperatures between 22 and ~65K, the specific heats of the two samples would differ only because their values of $\gamma^*(0)$ are different and, possibly, because impurity contributions to C may also be different for the two samples.

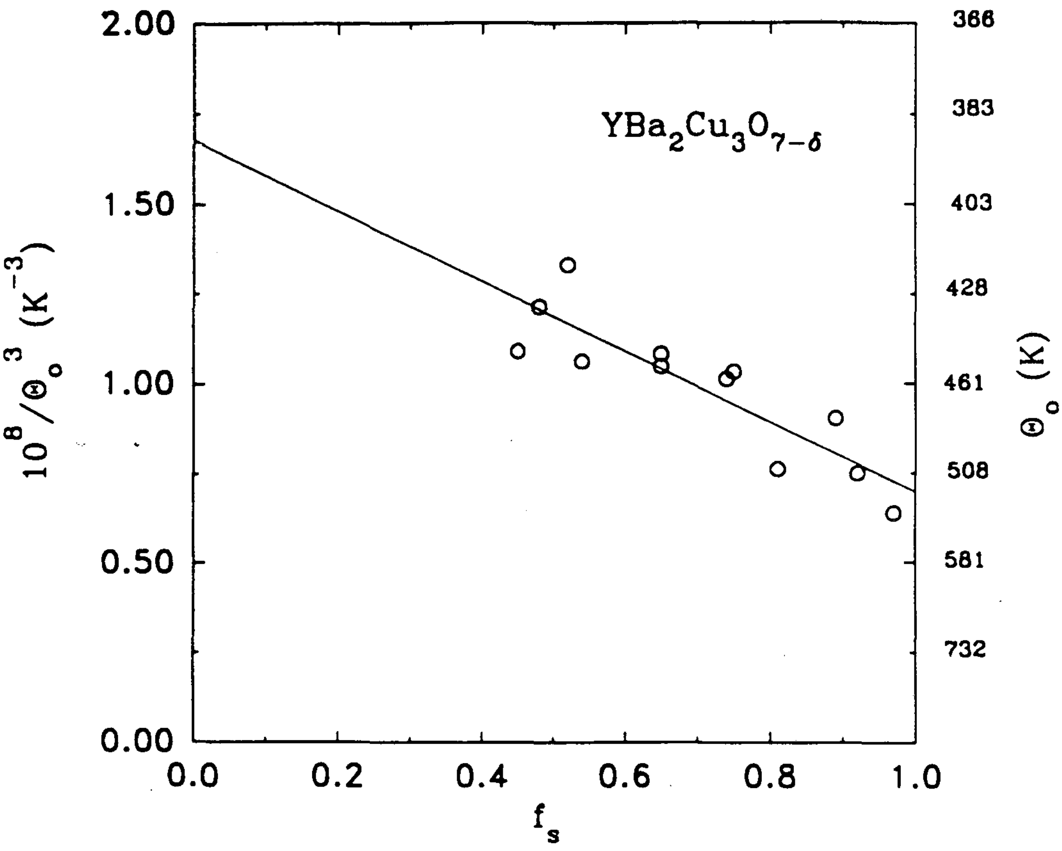

Fig. 3. The correlation of Θ_0^{-3} with f_s.

It appears, then, that the difference in the low-temperature lattice specific heats has disappeared at temperatures above ~20K. If we associate the low-temperature difference with different fractions of volume superconductivity in the two samples, it is logical to infer from these data that the difference in the phonon spectra of the superconducting and the "normal" phases lies in the very long wave length modes. The "normal" phase presumably contains Cu^{2+} magnetic moments that probably correspond to alterations in the bonding characteristics of the lattice. Thus, this "normal" material may well have a Θ_0 that is different from the Θ_0 of the superconducting material, and from that of the normal phase to which the superconducting material transforms at T_c.

Information about the low-frequency modes can, in principle, also be obtained from sound-velocity, v, measurements. There are numerous reports of small increases in v near T_c[8,9]. However, these changes are far smaller than the 30% difference in Θ_0 obtained from the intercepts in Fig.3. This apparent discrepancy may arise because the change in v is associated with the superconducting transition in regions that do not contain Cu^{2+} magnetic moments, whereas the difference in the intercepts in Fig.3 is associated with the difference between regions that contain such magnetic moments and regions that do not. On the other hand, the sample-to-sample variations in v reported in the literature may result, in part, from sample-to-sample variation in the concentration of Cu^{2+} magnetic moments, which affect f_s.

The sound-velocity measurements sample the very long wave length phonon modes, and do not test the proposition that the higher energy modes are unaffected by the superconducting phase transition. It is precisely the higher energy modes, however, that are sampled by inelastic-neutron scattering data. The results of Rhyne et al.[10] show there is a negligible difference in the YBCO phonon density of states above and below T_c. Although somewhat contradictory results have also been reported[11], the data in Ref. 10 provide support for the inference drawn from the specific-heat data that the higher energy phonon modes are relatively unaffected by the superconducting transition. The neutron-scattering experiments do not effectively sample those phonon modes that contribute to the low-temperature specific heat and

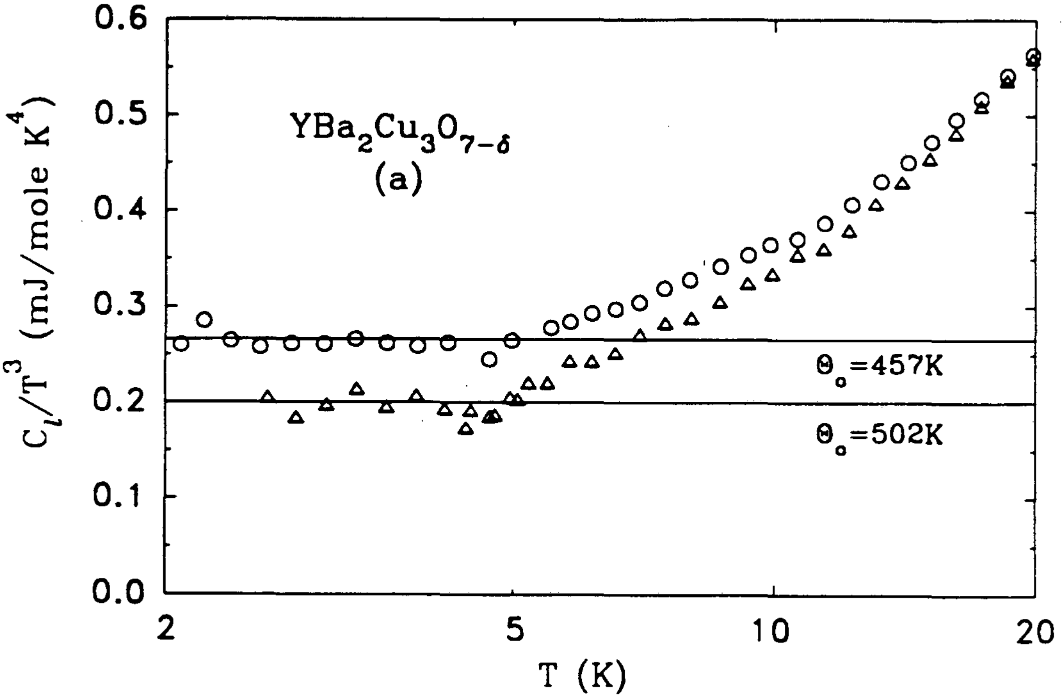

Fig. 4a. The lattice specific heat for two polycrystalline YBCO samples between 2 and 20K.

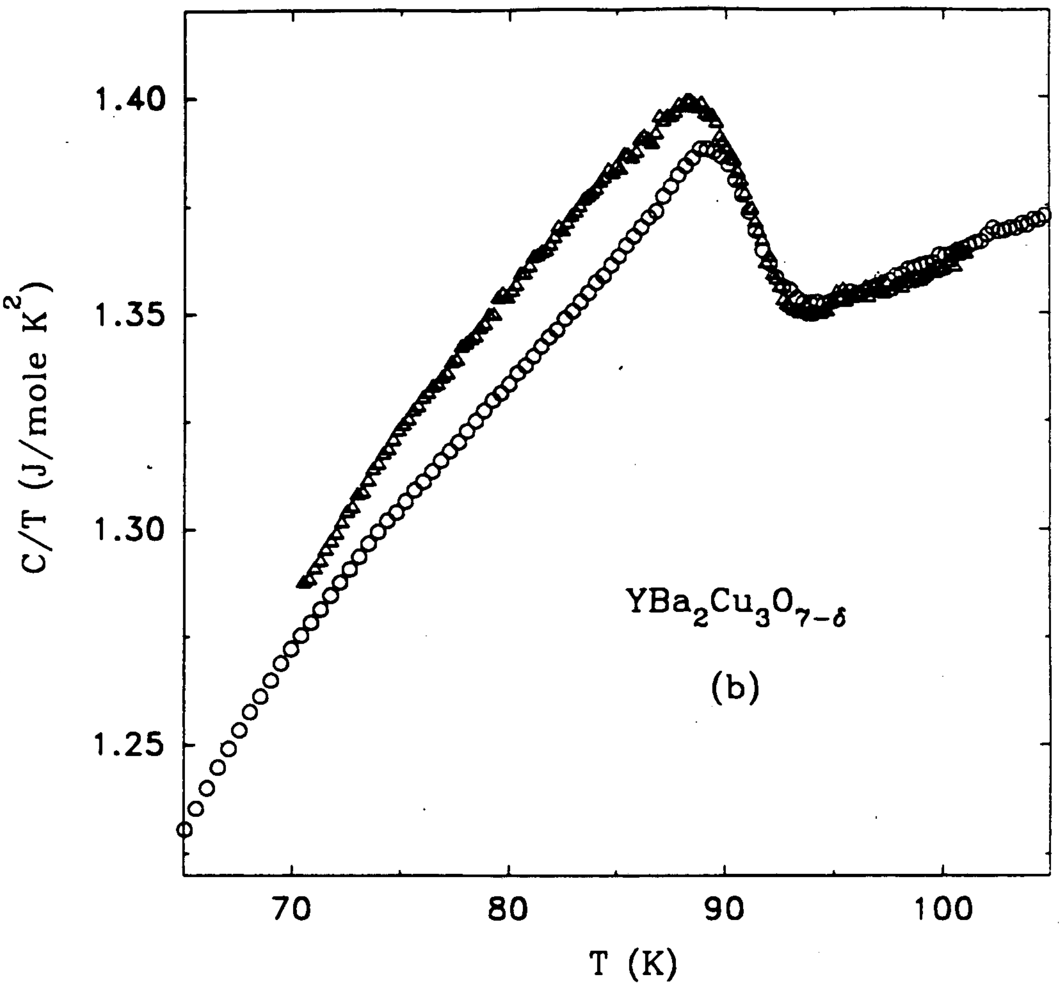

Fig. 4b. The specific heat for the same two polycrystalline YBCO samples shown in Fig. 4a in the vicinity of T_c.

therefore do not contradict the conclusion that these low-lying modes are affected by the presence of Cu^{2+} magnetic moments and, possibly, by the superconducting phase transition.

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