LOW TEMPERATURE INTERNAL FRICTION PEAK IN HIGH Tc SUPERCONDUCTORS

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We measure the internal friction Q^{-1} and Young's modulus sound velocity v_E in $YBa_2Cu_3O_{7-x}$ and $Bi_2Sr_2CaCu_2O_{8+x}$ samples. At temperatures near 35 K for KHz frequencies we observe an attenuation peak. In the case of $YBa_2Cu_3O_{7-x}$, assuming a thermally activated relaxation behaviour, we are able to show that it corresponds to the same process than other observed peaks at higher frequencies, probably involving impurity diffusional jumps. For $Bi_2Sr_2CaCu_2O_{8+x}$ this is not the case, and the origin of the peak, found in all the measured samples, remains unknown.

Acoustic measurements are a very sensitive tool for investigating elastic properties of matter. In the case of high temperature superconductors they have been decisive [1-4], toghether with specific heat and thermal conductivity measurements at very low temperatures to demostrate the existence of tunneling systems in these compounds. At other temperatures including those around the superconducting transition temperature Tc, different anomalies in the internal friction Q^{-1} and sound velocity v_E [3-7] have been reported by several groups in $YBa_2Cu_3O_{7-x}$ of different grain size and densities at low (vibrating reed) and high (ultrasound) frequencies. An important issue is the existence or not of an elastic anomaly accompanying Tc. It has been suggested [7] that the anomalies found near Tc are caused by a classical relaxation process with an activation energy of 0.16 eV, and not by the onset of superconductivity. In fact this is roughly the energy found from measurements at different frequencies by the vibrating reed technique [4]. However, the evidence accumulated for a structural anomaly [8,9] toghether with sound velocity anomalies [4,10-12] at different frequencies for single crystals and ceramic samples conclusively shows that there is an effect of the superconducting state on the lattice which is detected by elastic measurements. The attenuation peak associated with this anomaly should be, as in the case of charge density wave transitions [13], much smaller than the measured anomaly and could be overshadowed by a larger relaxation peak. Other attenuation peaks have been detected but to the best of our knowledge a systematic study has not yet been published.

In this report we present vibrating reed measurements performed on $YBa_2Cu_3O_{7-x}$ (YBCO) and $Bi_2Sr_2CaCu_2O_{8+x}$ (BSCCO) ceramic samples. In both, we observe an attenuation peak at $\simeq$ 35 K for KHz frequencies. In the case of YBCO we show that it can be related to a relaxation process with activation energy U = 63 meV, when it is compared to the existing measurements at different frequencies. In BSCCO, however, the behaviour observed does not correspond to a thermally activated process. The existing evidence points towards interstitial impurity hopping in the case of 1-2-3 compounds while no satisfactory explanation was obtained for the BSCCO samples.

The YBCO samples were prepared in the way described previously [4], while for the Bi-Sr-Ca-Cu-O compound reagent grade oxides (Bi_2O_3, CuO) and carbonates (CaCO, SrCO) in quantities to satisfy the formula $Bi_4Sr_3Ca_3Cu_4O_{16+x}$ were mixed in an agate mortar, lightly pressed (1000 Kg/cm^2) and heated in air at 800 °C for 18 hours. The reacted material was quenched in air, ground and pressed again into pellet shape with a load of 2500 Kg/cm^2 and kept for another 18 hours at 875 °C. X-ray diffractograms were fitted assuming a tetragonal cell with parameters a = 3.81(1) A and c = 30.68(1) A. EDAX analysis gives for the resulting compound the formula $Bi_2Sr_2CaCu_2O_{8+x}$. The electrical resistivity as a function of temperature showed a metallic like behaviour and the superconducting transition had its midpoint at 81 K.

The vibrating reed was excited and detected electrostatically, and the displacement at the end of the reed was estimated to be around 0.1 microns. The experiments were perfomed in a conventional cryostat inside a vacuum can and the temperature was controlled by an electrical heater driven by a temperature regulator whose input was a Germanium resistor. Temperature could be swept (typically at 0.2 K/min) or discrete points could be taken by stabilizing the temperature with the controller. Both methods give essentially the same results. We have measured samples 100-200 μm thick, 2.0-3.0 mm wide and 6-12 mm long. For the YBCO sample we obtained a value of v_E ($=(E/\rho)^{1/2}$, where E is the Young's modulus and ρ is the density) similar to the one reported previously [4], while for the BSCCuO samples $v_E = 2.0 \times 10^5$ cm/s.

In figure 1 we show the low temperature internal friction curves for the YBCO sample for which the two first oscilation modes were measured. We observe, as in the samples of reference 4, that there is a small maximun at ≃ 37 K which moves with frequency. On the hypothesis that it is caused by a thermally activated relaxation phenomena we plot in figure 2 in a semi-logarithmic scale the frequency as a function of inverse temperature, together with other data published [4-6]. We observe that all the points follow roughly the same activation energy H = 63 eV. On the other hand if we plot the points corresponding to the maxima observed in the range 60-70 K in experiments at ultrasonic frequencies [6,7] we see that they correspond to the same activation energy. The extrapolated attempt frequency is a reasonable 9×10^{11} Hz. Though it is difficult to say if it corresponds to some intrinsic property of this material or some impurity, it seems to be present in samples with different origin [4-7].

In figure 3 we show the internal friction temperature dependence for two different samples of BSCCO. Sample BF1 has typical grain size d ≃ 1 μm while in sample BT, which was made with a method to ensure small grain size, d ≃ 0.1 μm. On the other hand, while samples BF1 have a narrow superconducting Tc with no apparent amount of the 110 K $Bi_2Sr_2Ca_2Cu_3O_{10+x}$ phase, sample BT shows a small jump at 110 K followed by a complete superconducting transition at 80 K. We see that in all the samples there exists a very large and undefined maximun around Tc and a larger and more defined peak at higher temperatures. Other such as peaks have been reported [6] and their origin still remains unclear. We also observe a small anomaly at ≃ 30-35 K in the samples, similar to the ones that appear in the 1-2-3 materials. We also observed this anomaly in a third BSCCO sample measured. Plotting the points in figure 2 corresponding to the bismuth samples, we find that these maxima do not indicate a thermally activated process. This strongly suggests that the anomalies in the 1-2-3 materials and in the bismuth compounds are not due to the same phenomena. In the 1-2-3 compounds atomic diffusion is a probable cause, due to the activated behaviour of the relaxation peak. Oxigen atoms are able to diffuse in the plane corresponding to the Cu-O chains [15]. The activation energy for this phenomena is, however, about 1 eV, one hundred times higher than the energy corresponding to the process at the origin of the ≃ 35 K peak. On the other hand, we have perfomed preliminary measurements desoxygenating the YBCO sample to study the effect of an increase of vacancy concentration on the measured peak. An increase in the number of oxygen vacancies should favour oxygen diffusion with an enhancement of the amplitude of the internal friction peak. We observe, however, a disappearence of the peak. This confirms that it is due to other process, probably impurity interstitial diffusion that are eliminated by the anneal at 500 °C. The internal friction peak observed at ≃ 60 K for ultrasound (MHz) measurements and at ≃ 35 K for acoustic (KHz) measurements seems thus not to be an intrinsic property of 1-2-3 high temperature superconductors.

The situation is different in the BSCCO compound. The measured internal friction peak appears with different magnitude and is not a thermally activated process. It is known that Bi-Sr-Ca-Cu-O can always contain small percentages of 2201 phase with a Tc ≃ 7 K. Intergrowths between this and the 2212 phase will probably have Tc's in between. We can speculate that, in this picture, the observed peak is due to the superconducting phase transition of the intergrowth portions of the sample. However, it is more plausible that different types of intergrowths have different Tc's resulting in a continuous anomaly, not a defined peak. On the other hand the peak on sample BF1, which was used for the study of "vortex melting" [16] disappeared after about twenty temperature cycles. Though it could be that the peak was thermal history-dependent, cycling for 2-4 times the other samples did not show any effect on the anomaly.

In conclusion we have detected a maximun in the internal friction temperature dependence in both $YBa_2Cu_3O_{7-x}$ and $Bi_2Sr_2CaCu_2O_{8+x}$ at temperatures of ≃ 35 K for KHz frequencies. This relaxation process comes from a thermally activated mechanism in $RBa_2Cu_3O_{7-x}$ with the same activation energy as observed by different experimental groups at other frequencies. The most plausible explanation is that this is caused by impurity atomic diffusion jumps. On the other hand the peak in $Bi_2Sr_2CaCu_2O_{8+x}$ seems not to be thermally activated and its origin remains obscure.

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FIGURE CAPTIONS:

Figure 1: Low temperature internal friction peak in 123 compounds:

a) YBCO, this work, (■) f=1.5 KHz, (□) f= 8.9 KHz,

b) YBCO, ref. [4] f= 1.92 KHz,

c) YBCO, ref. [4] f= 1.05 KHz,

d) $EuBa_2Cu_3O_{7-x}$, ref. [4] f= 1.35 KHz.

Figure 2: Frequency vs. inverse of the temperature.

YBCO: (●) this work; (+) ref. [4]; (□) ref. [5];

(X) ref. [6]; (O) ref. [7]

EuBaCuO: (Δ) ref. [4]

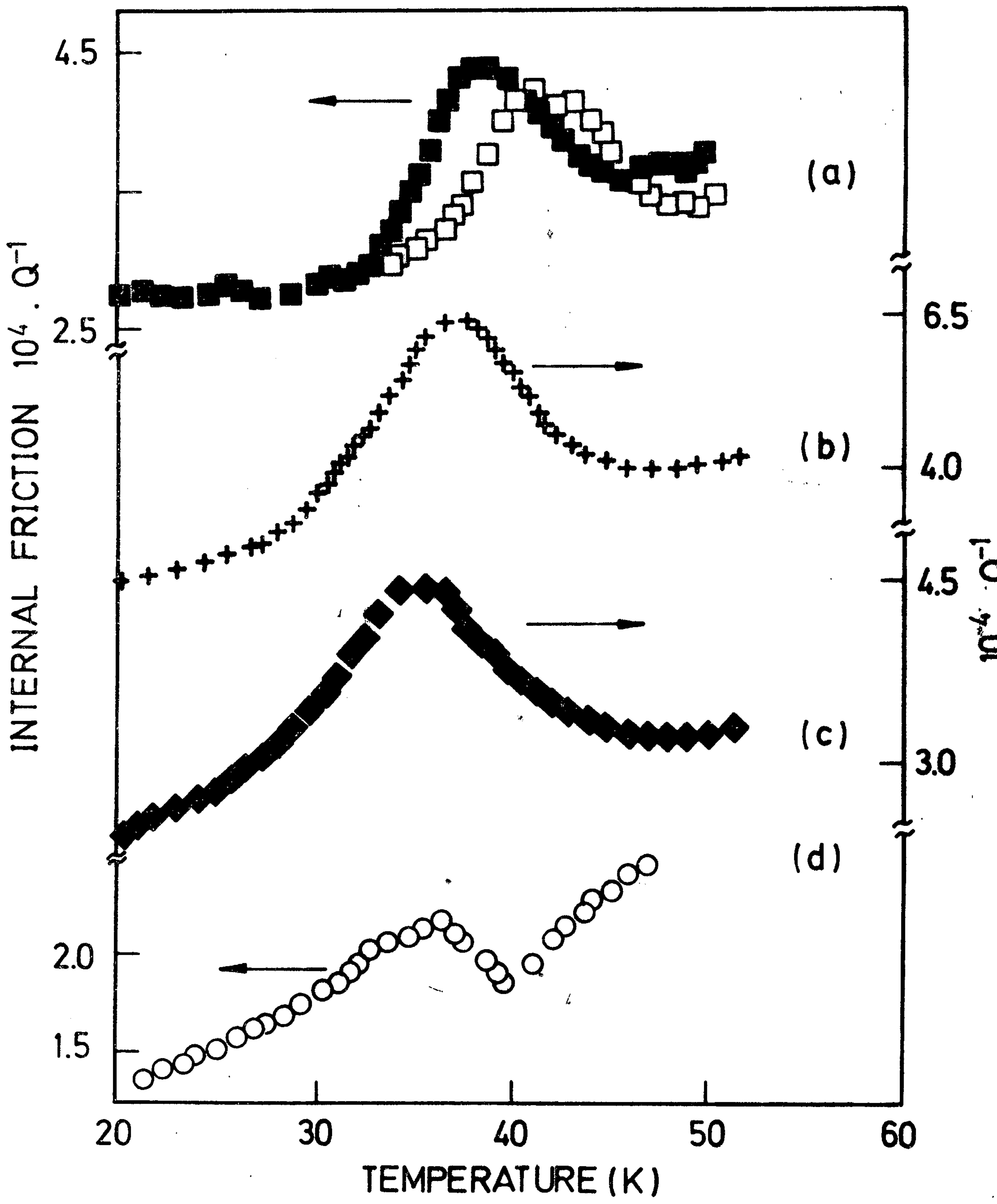
BSCCO: (■) this work.

The line is the fit $f = f_o \exp(-U/kT)$ using only the 1-2-3 [illegible] data.

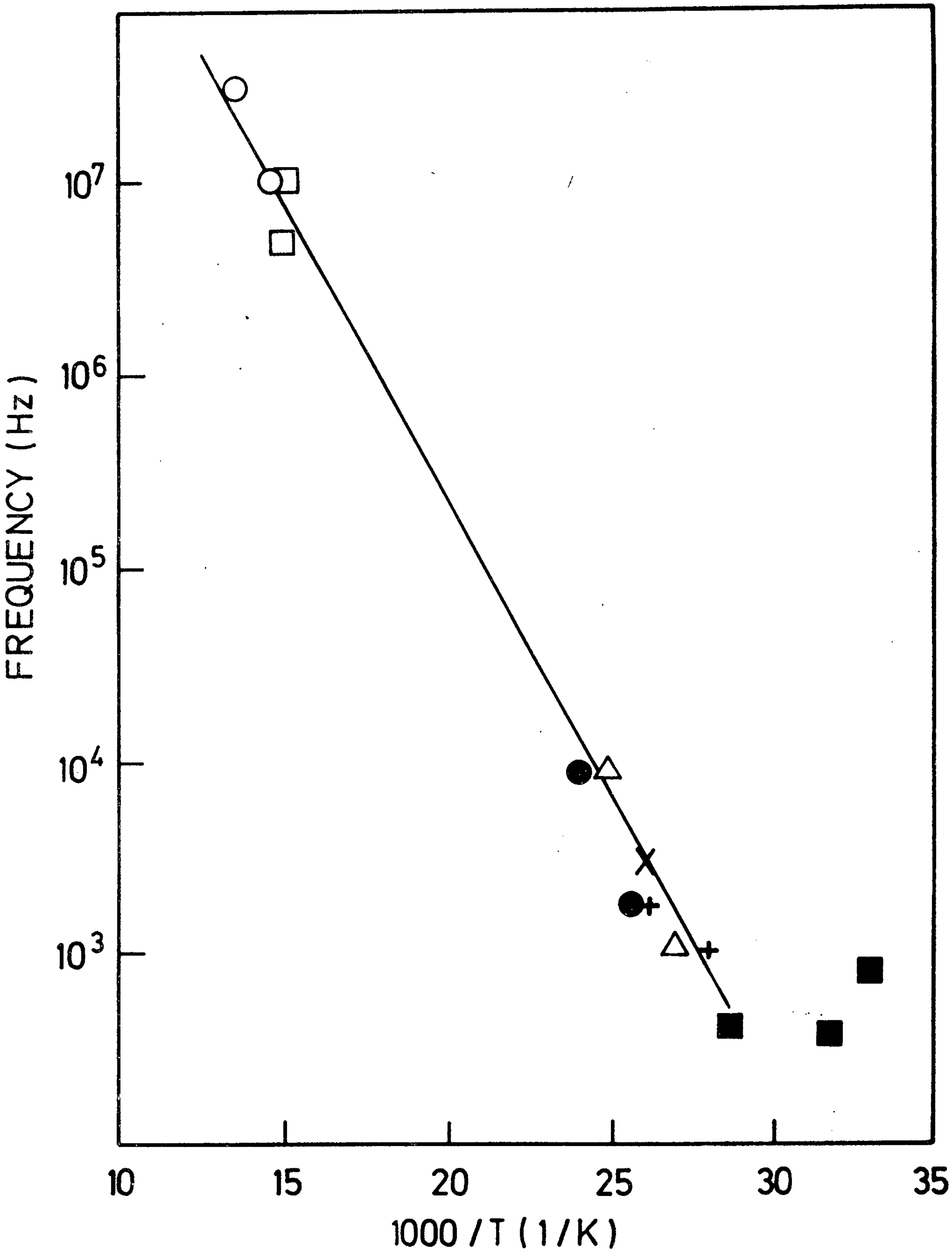
Figure 3: Internal friction of $B_2Sr_2CaCu_2O_{8+x}$ vs temperature.

(BTH) f= 0.41 KHz , (BTT) f= 0.39 KHz.

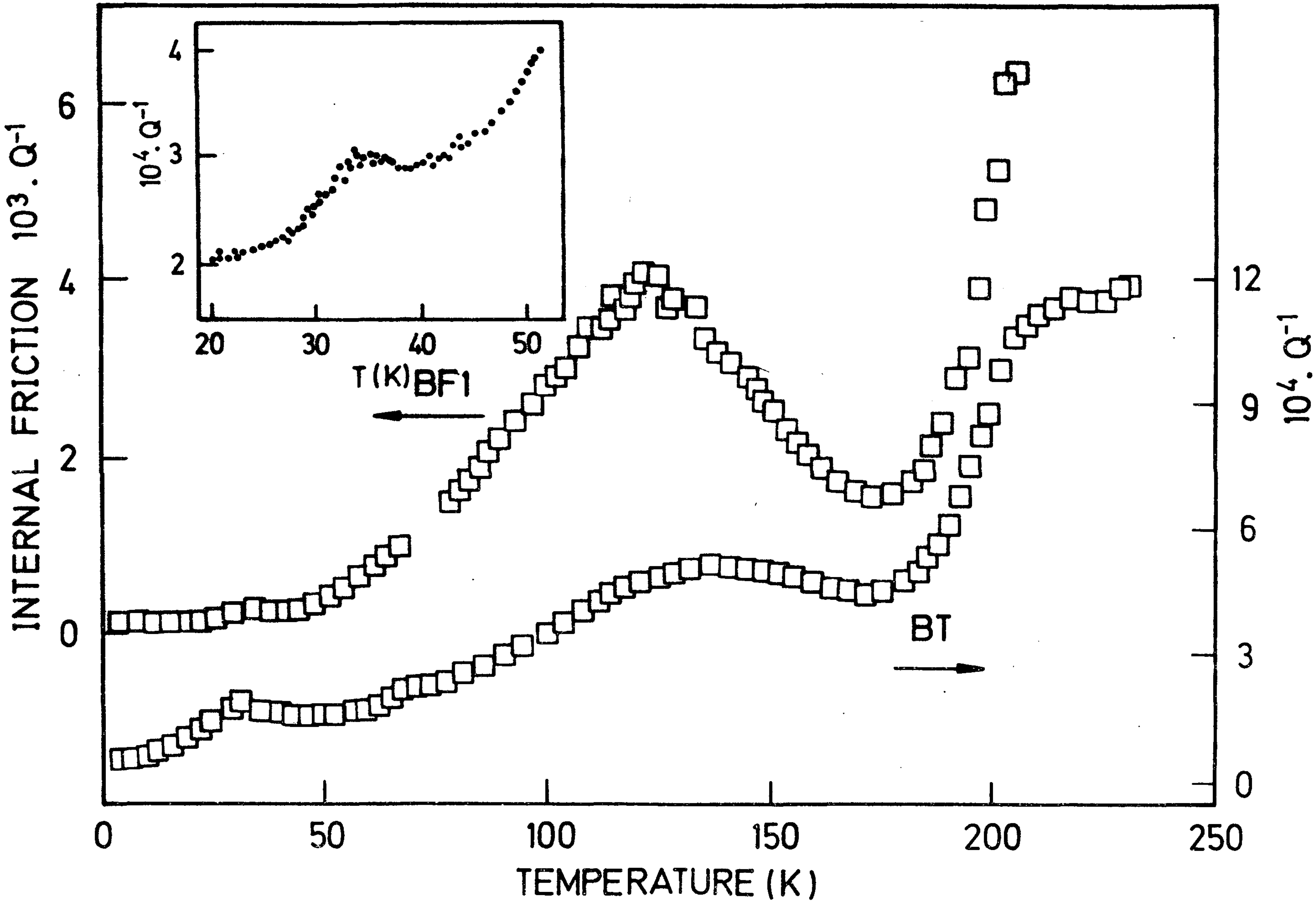
Inset: low temperature attenuation peak in BF1 vs. temperature, in a amplified vertical scale.



INTERNAL FRICTION $10^4 . Q^{-1}$
4.5
2.5
2.0
1.5
(a)
(b)
(c)
(d)
6.5
4.0
4.5
3.0
$10^{-4} Q^{-1}$
20
30
40
50
60
TEMPERATURE (K)



FREQUENCY (Hz)
10^7
10^6
10^5
10^4
10^3
10
15
20
25
30
35
1000 / T (1 / K)



INTERNAL FRICTION $10^3.Q^{-1}$
TEMPERATURE (K)
$10^4.Q^{-1}$
T(K)
BF1
BT