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FREQUENCY DEPENDENT SUSCEPTIBILITY OF POLYCRYSTALLINE $Bi_{0.7}Pb_{0.3}SrCaCu_2O_x$

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The low field complex ac susceptibility of the ceramic high - T_c superconductors has been investigated close to T_c for frequencies from 1.75 kHz to 56 kHz. There is shown that the critical state model is not sufficient for description of the results. Analysis of the data suggests the existence of the relaxation processes which manifests in strong frequency dependence of susceptibility and are possibly connected with the weak flux pinning at low fields. The analogy with spin-glass-like behaviour of susceptibility is discussed.

1. Introduction.

In our early papers[1] the frequency dependence of the complex ac susceptibility of YBCO samples has been investigated in the very low ac fields. It was found that the maximum of $\chi''(T)$ moves to higher temperatures as the frequency increases. The spin-glass-like scaling of the frequency dependence of that temperature has been found.
The maximum value of $\chi''(T)$ is close to $0.212/4\pi$ at large ac fields, in aggreement with predictions of the simple critical state model[2]. Higher harmonics in ac susceptibility are observed as evidence of strongly nonlinear M(H) dependence of the small hysteresis loop[3,4] and the temperature of the onset of these harmonics is easily determined. The onset of higher harmonics might be interpreted as the onset of the phase-coherence between different superconducting grains interacting through the weak Josephson junctions and the occurrence of the critical state in the bulk superconducting sample[3].
In Bi-based ceramics we meet more complex situation. The maximum in $\chi''(T)$ is about twice smaller than it follows from the simple critical state model. We observe the higher harmonics, too. This is the evidence on the nonlinear M(H) dependence. But the origin of that nonlinearity is not clear for us: is it connected with the existence of the magnetic hysteresis or with the strong field dependence of the current density at the very small fields. It is not possible to define sharp onset of the higher harmonics as a function of temperature, contrary to the situation in YBCO ceramics. In order to study systematically these differences the investigations of the frequency dependence of the ac susceptibility of two nominally $Bi_{0.7}Pb_{0.3}SrCaCu_{1.8}O_x$ samples have been carried out. We find the similarities of $\chi(\omega)$ dependence to the results observed in spin-glasses. We analyse the data in the terms of the stretched exponential response function used lately for describtion of the magnetization relaxation in some superconductors.

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2. Details of the measurements.

The samples were produced in the standard solid-state reaction method[5]. Both are cylindrically shaped with the length 5 mm and 3 mm in diameter. One of these samples has been exposed to the irradiation by the fast neutrons with a dose 6.6×10^{17} n/cm^2, which caused the strong destruction of their superconducting properties in the atomic scale level[5].
The measurements were carried out using ac bridge constructed from two identical coils and stable, temperature insensitive resistors. The ac field amplitude was about 0.1 Oe and was constant for all frequencies. Both components of ac susceptibility have been registered simultaneously during the slow cooling down. The curves of $\chi'(T)$ for each frequency have been normalized to the same value at low temperature (80 K) and proportionally normalization of $\chi''(T)$ has been obtained. This is justified by the weak temperature dependence of susceptibility at low T.

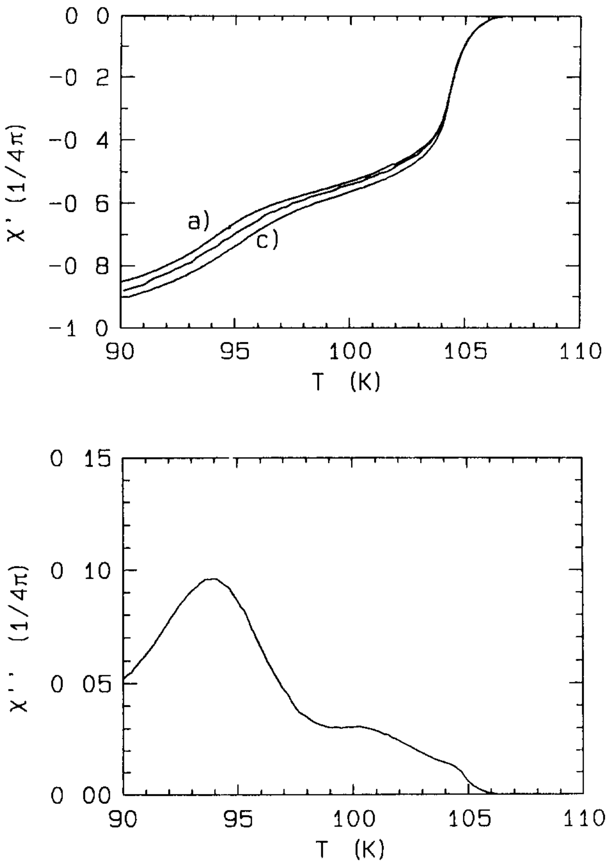

Figure 1. Temperature dependence of the ac susceptibility for nonirradiated sample: upper curves - χ', bottom - χ''. Letters a) to c) refer to the measuring frequency: 1.75, 7 and 28 kHz, respectively.

3. Experimental Results.

Figures 1 and 2 present measured frequency dependent susceptibility of both samples. We observe strong frequency dependence of real and imaginary parts of susceptibility for both samples. The inflection point in χ'(T) is observed when χ''(T) reaches maximum. The position of the maximum in χ'' shifts toward higher temperatures with increase of frequency. The maximal value of χ'' is 2-3 times smaller than it would follow from the critical state model. Very interesting feature is the double maximum observed in χ''(T). At such small ac fields we expected only one maximum connected with the occurrence of the bulk superconductivity due to the Josephson interaction between the grains. That maximum should be located roughly at the position of the lower of these observed maxima. Systematic studies of the series of neutron irradiated samples leads to conclusion that the higher-T maximum is connected with the damage of the sample. The value of χ'' at higher-T maximum is systematically increasing with the neutron irradiation. It points on the connection of the higher-T maximum with the intra-grain superconductivity. In order to test this assumption we have carried the measurements on the same irradiated sample after crushing it to the powder of the grains of dimension less then about 30 μm. Then the first maximum in χ''(T) has been still observed while the second one was absent.

4. Analysis and Discussion.

Despite we don't understand the whole of the observed results we are able to carry their phenomenological analysis.
Magnetic relaxation effects in low dc fields were studied in HTCS materials[6,7] and in heavy-fermion superconductor[8] UPt_3. The authors have pointed out on the similarities of the relaxation effects to those found in disordered materials, where the Kohlrausch law is often used to describe the relaxation processes:

$$M(t)=M(\infty)+(M(0)-M(\infty))\exp(-(t/\tau_p)^{\beta}) \quad (1)$$

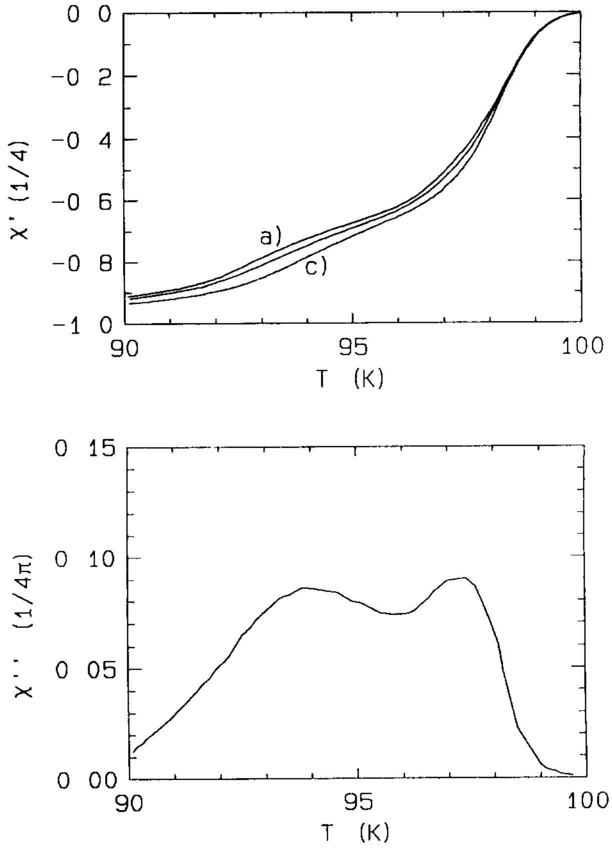

Figure 2. The same results as in Fig.1 but for the sample irradiated by neutrons.

Another approach to describe the relaxation processes in disordered materials is to use the concept of distribution of the relaxation times. However, we prefer the first method as we believe, there is a common description of relaxation effects in wide class of materials and the use of the concept of distribution of relaxation times may lead to missing a large portion of physics. But both approaches are equivalent usually. Kohlrausch law can be approximated strictly by some distribution of relaxation times.

There are many theoretical derivations of the formula (1)[11]. For instance Campbell et al discuss stretched exponential relaxation in terms of the number of available states in configurational space, changing as the glass transition is reached.

For response function (1) the frequency dependent susceptibility can be approximated by the Williams-Watts series, when $\omega \ll 1/\tau_p$ and by the series[10]:

$$\chi' = \chi_S + (\chi_T - \chi_S) \sum \Gamma(\beta m)/\Gamma(m) \cdot \beta x^m \cos(\frac{\pi}{2}\beta m) \quad (2)$$

$$\chi'' = -(\chi_T - \chi_S) \sum \Gamma(\beta m)/\Gamma(m) \cdot \beta x^m \sin(\frac{\pi}{2}\beta m) \quad (3)$$

when $\omega \gg 1/\tau_p$. Here Euler's gamma function is used, χ_T and χ_S are isothermal and adiabatic (equilibrium and immediate) susceptibilities of the medium and $x=(\omega/\omega_p)^\beta$. In the limit of large frequencies we have:

$$d\chi'/d\ln\omega = \beta/tg(\pi/2 \cdot \beta) \cdot \chi'' \quad (4)$$

In the limit of very broad distribution of relaxation times (or equivalently when $\beta \ll 1$) eq. (4) becomes:

$$d\chi'/d\ln\omega = 2/\pi \cdot \chi'' \quad (5)$$

-this result is observed experimentally in spin-glasses at $T \ll T_g$.

We have checked if the relation (4) is valid for our data. The experimental $\chi'(T)$ curves for each frequency measured (from 1.75 to 56 kHz) have been interpolated for a discrete set of temperature points, with a step of 0.25 K. Then the $\chi'(\ln \omega)$ has been found for each temperature. In the whole investigated temperature and frequency range that relation was linear, within the experimental accuracy, with some scatter of the data close to T_c only. The figure 3 presents the fitted slope of $\chi'(\ln\omega)$ as a function of temperature for both samples. There is a good correspondence between these results and the shape of $\chi''(T)$ for both samples. That implies that the relation (4) holds. The coefficient $\beta/tg(\beta\pi/2)$ determined for the results in figure 3 using (4) is 2-3 times smaller then $2/\pi$. This can be explained by large value of β (about 0.8 at 98K and 0.6 at 90K for irradiated sample; β should slowly decrease with lowering temperature[11]).

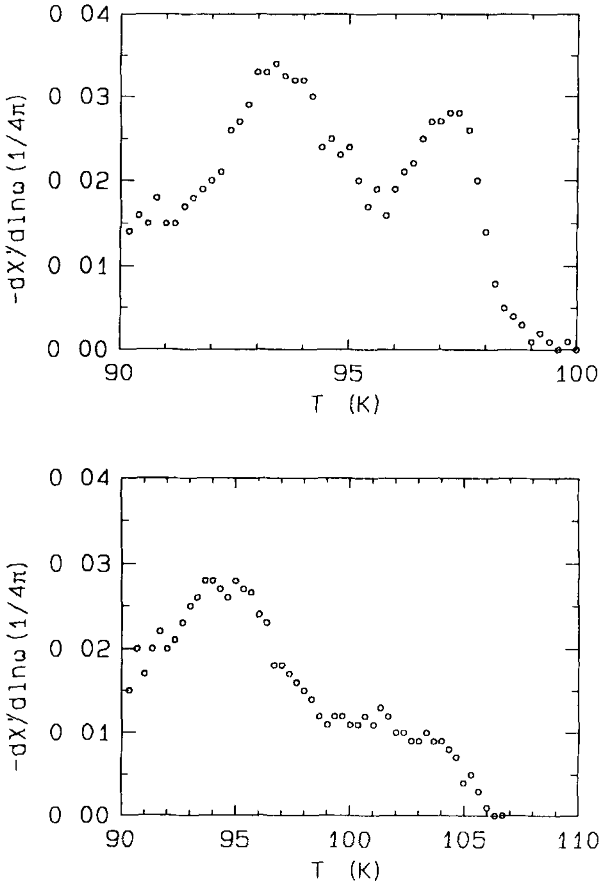

Figure 3. $d\chi'/d\ln\omega$ in units of $1/4\pi$. a)-nonirradiated sample, b)-irradiated sample. Each point in the figure has been determined from the sets of the data similar to these presented in Fig.1 and 2.

5. Conclusion.

The strong frequency dependence of the complex ac susceptibility of $Bi_{0.7}Pb_{0.3}SrCaCu_2O_x$ ceramics was observed. The determined $d\chi'/d\ln\omega$ (T) is similar to $\chi''(T)$. χ'' at the maximum is about 2 times smaller than it follows from the critical state model. That implies that the imaginary part of susceptibility is connected with the relaxation effects during the flux penetration into the sample. The measured frequency dependence is similar to that observed in spin-glasses and other disordered systems. The double maximum in $\chi''(T)$ occurs when the intragrain disorder is induced by the neutron irradiation. The detailed mechanism of the observed effects is not clear for us. The understanding of ac susceptibility measurements is extremely important as it is one of the most common methods of investigation of superconductors and there is a lot of unresolved problems connected with this method.

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11. In spin-glasses the β is decreasing as the glass transition is approached from above and β is close to 1/3 at the critical temerature.