Physica C 159 (1989) 281–286
North-Holland, Amsterdam

FREQUENCY DEPENDENT SUSCEPTIBILITY OF THE CERAMIC $Y_1Ba_2Cu_3O_{7-\delta}$ – THE SPIN-GLASS-LIKE APPROACH

Z. KOZIOŁ
Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Warszawa, Poland [#]

Received 25 April 1989

The AC complex magnetic susceptibility of the ceramic $Y_1Ba_2Cu_3O_{7-\delta}$ was measured close to the superconducting transition temperature as a function of a superimposed DC field and frequency of the AC field. Temperatures T_J of the maxima of the imaginary part of the susceptibility are connected with the occurrence of the phase locking of the grains order parameter through the Josephson weak links interaction. The standard spin-glass-like critical slowing down of this transition temperature was obtained. From the DC field scaling of T_J critical exponents were determined which are similar to these ones in spin-glasses.

1. Introduction

There is a large amount of works investigating spin-glass-like properties of superconductors, from the granular mixtures [1] of the classical low T_c superconductors and nonsuperconducting materials through the lithographically fabricated networks of the Josephson junctions [2] to the high T_c oxide materials [3,4]. In the ceramics and single crystals the remanent magnetisation has been measured [5], history dependent AC susceptibility was observed [6], DC field scaling of the spin-glass-like temperature in magnetisation [7] was obtained and one of the most basic properties of spin glasses, the existence of memory effects in magnetisation [4] was shown and strong indications for aging effect were suggested due to the AC susceptibility measurements [8].

The early interpretation of Müller et al. [9] of the critical irreversibility temperature dependence in magnetisation measurements as the existence of the de Almeida–Thouless $H^{3/2}$ line has been explained in the frame of the giant flux creep effect [10]. But there is one other transition temperature T_J connected with the occurrence of the bulk superconductivity in the sample due to the interactions between the superconducting entities through the Josephson weak links. Although the existence of the Josephson junctions between the superconducting grains in the ceramic materials and in the thin films [11] is obvious the true origin of the spin-glass-like properties is unclear. It seems, that the weak links can be formed inside the grains, probably at twinning boundaries [9,12,13]. The role of a possible Kosterlitz–Thouless transition should also be taken into account in the analysis of all transport (and magnetic) properties of HTCS [2,14]. The temperature T_J manifests itself in the imaginary part of the AC susceptibility [8], one observes a maximum in χ'' (T) at T_J with T_J dependent on H_{AC} and H_{DC}. It is possible to observe the second maximum in χ'' (T), connected with the flux-creep effect, at T more close to T_c than T_J, where the existence of some activation energy for the flux-flow causes field and frequency dependence of the temperature of the maximum in χ'' [15]. This effect is related to the time and history dependence of the magnetisation. We have studied these phenomena in $Bi_{1-x}Pb_xSrCaCu_2O_x$ samples [16]. The most probable situation is that at low temperatures one observes both contributions to the time dependent magnetisation: one connected with the flux-creep and the other one connected with the spin-glass-like properties caused by the disordered nature of the high T_c superconductors.

[#] Present address: Natuurkundig Laboratorium der Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, Postbus 20215, 1000 HE Amsterdam, The Netherlands.

0921-4534/89/$03.50 © Elsevier Science Publishers B.V.
(North-Holland Physics Publishing Division)

In the present work we are dealing only with the scaling properties of T_J, temperature of locking of the "grains" order parameter through the Josephson interaction described by the Hamiltonian written in the form:

$$\mathscr{H}=\sum_{i,j} J_{ij}\cos(\Phi_i-\Phi_j-A_{ij}) \qquad (1)$$

where Φ_i is the phase of the ith grain and A_{ij} is the proper path integral from the vector potential of the magnetic field between the grains. The parameter J_{ij} is proportional to the critical Josephson current density for the junction. It is easy to understand the equivalence of the Hamiltonian (1) to the X–Y spin-glass Hamiltonian with the same distribution of J_{ij} [3]. Frustration is introduced into (1) through the random Φ_i, Φ_j and by the magnetic field.

The phase locking temperature T_J is related to the interaction energy in (1). Field and time scaling of the measured T_J corresponds to the field and time scaling of the apparent interaction energy. With this idea we present the measurements of the field and frequency dependence of $\chi(T)$ and we discuss the scaling properties of T_J following the work of Svedlindh et al. [17] and using well accepted phenomenology of the spin-glass transition. These results are the first one presentation of the frequency dependence of susceptibility although many authors have claimed for the existence of this effect. The shift of $\chi'(T)$ and $\chi''(T)$ to higher temperatures with frequency resemble the similar behaviour of susceptibility in spin-glasses. We will try to explain phenomenologically the origin of this analogy.

2. Experimental details

Ceramic samples of 3 mm in diameter and 5 mm long were obtained in the standard solid-state reaction method. Complex magnetic susceptibility was measured using a self-made AC bridge. Temperature was monitored with the help of the carbon resistor with relative accuracy of about 0.05 K. The data were gathered by the computer system controlling the experiment. These details were described in our previous paper [8], where the characteristic features of the AC susceptibility were also summarised. With our experimental setup we are not able to determine exactly the absolute value of the measured susceptibility. Because of this we have normalised χ' to $-1/4\pi$ at low temperature (85 K) for all measured frequencies. It is justified because χ' is practically independent of T for lower temperatures in the case of small enough amplitude of the AC field H_{AC} and in zero external DC field H_{DC}. The amplitude H_{AC} has been calculated from the geometrical dimensions of the coils and was also measured by the other small coil in the middle of one of the bridge coils. These both methods have given the same results for H_{AC} with an accuracy better then 5% for all used frequencies. The phase angle of the susceptibility was adjusted at 85 K using a crushed electrotechnical ferrite as reference sample not introducing losses.

$\chi'(T)$ strongly depends on the history of the sample [6]. It is known, that the Meissner effect (FC, field cooled magnetisation) is much lower then the ZFC (zero field cooled) magnetisation. Because of these facts all presented results for the superimposed DC field were made under the same conditions in the field cooling procedure and we have assumed that the external DC field is equal to the internal DC field.

3. Results and discussion

Figure 1 presents some of the typical curves of $\chi'(T)$ and $\chi''(T)$ obtained for frequencies from 1.2 kHz to 75 kHz for the H_{AC} amplitude of 0.01 Oe. It is justified that the temperature T_J of the χ'' maximum determines the critical intergrains Josephson supercurrent at this temperature [6,8,18] through the relation

$$J_c(\mathrm{A/cm^2})=\frac{10}{4\pi}H_{AC}(\mathrm{Oe})/r(\mathrm{cm}),$$

where r is the radius of the sample. On the other hand, the intergrain supercurrent determines the interaction between the grains in Hamiltonian (1). Field and time (frequency) scaling of the critical Josephson current corresponds to field and time scaling of the interaction energy in (1). Because of this field and frequency dependence of the characteristic temperature $-T_J(H_{AC},f)$ can be treated as the field dependence of the Josephson interaction energy for the appearent time-scale $1/f$.

As was described previously [8] for H_{AC} lower than

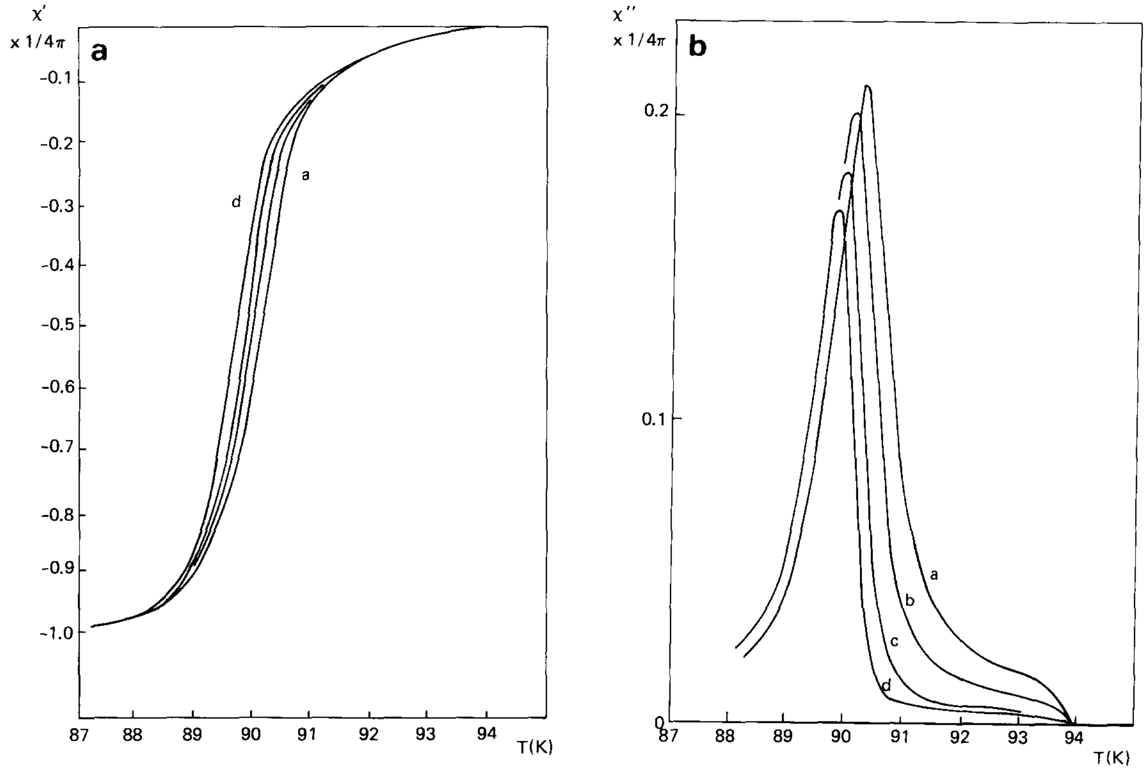

Fig. 1. Temperature dependence of the complex susceptibility close to the superconducting transition temperature $T_c=93.7$ K for the frequencies of the AC field of 75, 19.2, 4.8, and 1.2 in kHz (curves from a) to d). (a) real part, χ', (b) imaginary part, χ''. χ' was normalised to $-1/4\pi$ at $T=85$ K for all measured frequencies and χ'' is in the same units as χ'. $H_{AC}=0.01$ Oe.

about 0.05 Oe one is not able to see any difference in the $\chi(T)$ traces for different H_{AC} amplitudes. This fact was helpful in our measurements because of some uncertainty in determining the H_{AC} field insight the coils. But we have checked, that exponents connected with the critical slowing down of the glass transition do not depend in our measurements on H_{AC} amplitude for $H_{AC}=0.01$ Oe and $H_{AC}=0.1$ Oe. In fig. 2 we present a log–log plot of the glass transition temperature T_J obtained in $H_{AC}=0.01$ Oe. For practical reasons other measurements were done with $H_{AC}=0.1$ Oe.

In this figure we introduced the glass transition temperature T_g which has been determined in analogy to the case of spin-glasses from the fitting of the relation

$$f=f_0(T_J-T_g)^{\nu z}$$

to the experimental data of $T_J(f)$ for the frequencies from 300 Hz to 80 kHz. The straight line corresponds to $\nu z=3$, with $T_g=90.0$ K. For $H_{AC}=0.1$ Oe one obtains $T_g=89.75$ K and this value of T_g will also be determined by different scaling presented in fig. 4.

The field dependence of T_J for T above 87 K is shown in fig. 3 for the DC fields from 0 up to 100 Oe and for the different frequencies from 1.2 kHz to 75 kHz.

In the previous work [8] similar results for the same sample were obtained for the frequency of 7 kHz and the line $T_J(H_{DC})$ there was interpreted as the line of the second critical field for the bulk Josephson superconductivity: for $T<T_J(H_{DC})$ the whole sample is superconducting because of locking of the grain phases and for $T>T_J(H_{DC})$ only localised superconductivity exists. A similar critical line has been shown in the work of Barbara et al. [13], where susceptibility measurements were done both

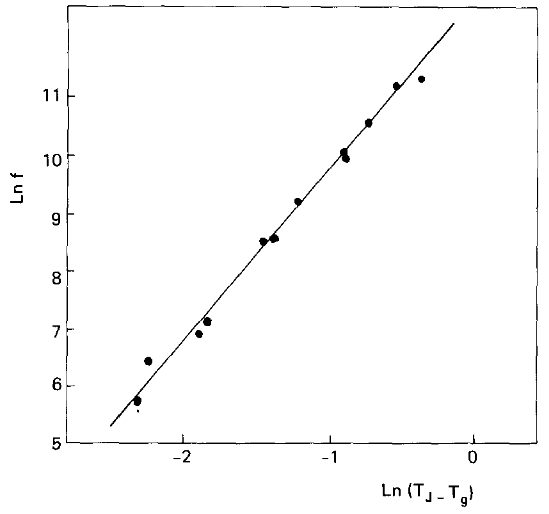

Fig. 2. Frequency dependence of temperature T_J of the maxima in the $\chi''(T)$ measured in the AC field 0.01 Oe for the frequencies from 300 Hz to 80 kHz: $\ln f$ - $\ln(T_J - T_g)$ plot. The strait solid line is: $\ln f(\mathrm{Hz}) = 12.8 + 3.0 \ln(T_J - J_g)$ with $T_g = 90.0$ K.

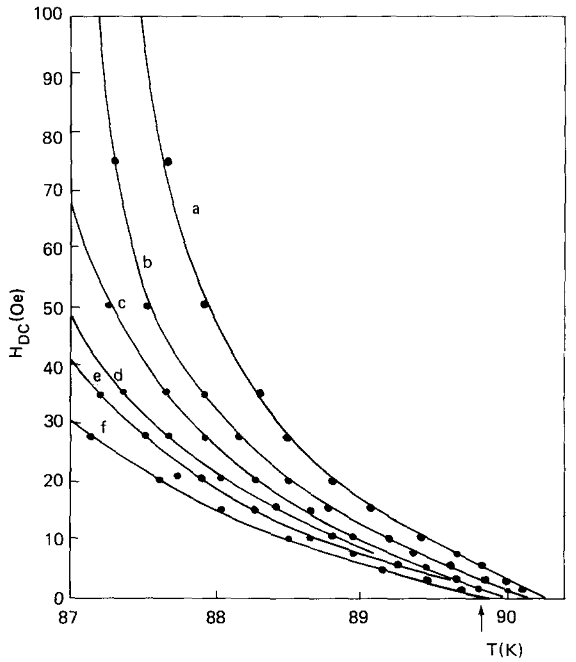

Fig. 3. A part of the measured data for the DC field dependence of the temperatures of $\chi''(T)$ maxima for the frequencies: (a) 75 kHz, (b) 38 kHz, (c) 19.2 kHz, (d) 9.6 kHz, (e) 4.8 kHz, (f) 1.2 kHz. The lines are only to guide the eyes. Measurements were done with $H_{AC} = 0.1$ Oe ($T_g = 89.75$ K for this amplitude and T_g is marked by the arrow on the temperature axis).

for the ceramic and single-crystal samples. There exists also large similarity to the transport results [14] of the field dependence of the critical temperature for the appearance of a very small critical current. Frequency and field scaling of T_J can be understood as a conventional scaling of the freezing temperature of the spin-glass transition T_F. We are obtaining [17] for the relaxation time:

$$\tau = \tau_0 t^{-\nu z} [g(H^2/t^\phi) + 1]^{-z} \quad (2)$$

where τ_0 is a characteristic relaxation time, t is the reduced temperature, $t = (T - T_g)/T_g$ with T_g the equilibrium spin-glass transition temperature, ν and z are the critical exponents defined by the temperature dependence of the spin-glass Edwards–Anderson correlation length $\xi_{EA} \propto t^{-\nu}$ and by the scaling of the relaxation time with ξ_{EA}: $\tau \propto \xi_{EA}^z$. ϕ determines the field scaling of χ_{EA}.

The asymptotic behaviour of $g(x)$ is known:

$$\lim_{x \to 0} g(x) = x \quad \text{and} \quad \lim_{x \to \infty} g(x) = x^{\nu/\phi} .$$

For $H = 0$ one obtains for the frequency scaling of the temperature T_J ($f = 1/\tau$):

$$f = f_0 (T_J - T_g)^{\nu z} .$$

Results in fig. 1 can be fitted by this formula with $\nu z = 3 \pm 0.5$ and with $T_g = 90.0$ K for $H_{AC} = 0.01$ Oe. At $T = T_g$ ($t = 0$) one obtains from (2):

$$\ln f = \ln f_0 + 2\nu z/\phi \ln H .$$

This equality does not hold at $t \neq 0$ and the fig. 4 obtained from the extrapolated data of fig. 3 illustrates this. At T_g we have from the slope of $\ln f$ vs. $\ln H$ a value of $2\nu z/\phi = 3/2$. Because of these results we calculate ϕ to be about 4. This exponent determines the field dependence of T_J, $T_J(0) - T_J(H) \propto H^{2/\phi}$ and one expects ϕ to be close to 3. But it is known for HTCS that the measured spin-glass-like temperature is not always fitted to $\phi = 3$ (Giovannella et al. [7] obtains values for $2/\phi$ from 0.55 to 0.71 from the quasistatic ZFC results for different fields) and in spin-glasses values from 3.3 to 5 are reported [19]. Figure 5 is a further illustration of the scaling of T_J with f and H_{DC}. After putting τ equal to a constant (2) can be written:

$$[t_J(0)/t_J(H)]^\nu = g(H^2/t_J(H)^\phi) + 1 , \quad (3)$$

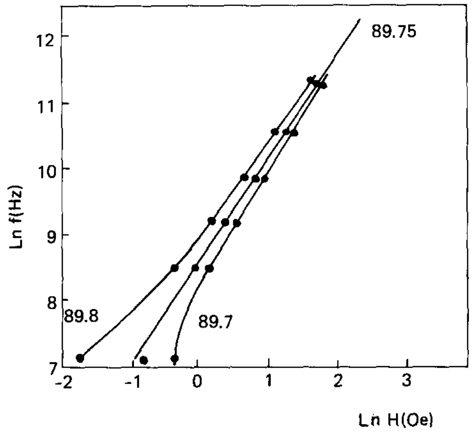

Fig. 4. $\ln f - \ln H_{DC}$ plot close to $T_g = 89.75$ K. Data of H_{DC} were obtained from fig. 3 by putting the straight vertical lines at $T = 89.7$ K, 89.75 K, and 89.8 K. Linear dependence of $\ln f$ on $\ln H_{DC}$ should be obtained for T_g. The straight solid line for 89.75 K has a slope equal to 3/2.

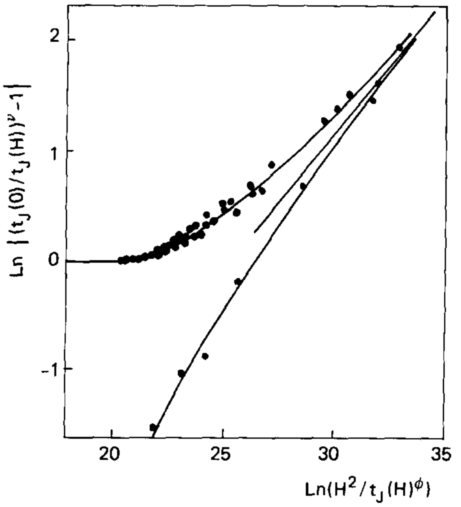

Fig. 5. Universal spin-glass-like scaling of the data of fig. 3 according to eq. (3) and similarly as Svedlindh et al. [7] are scaling T_J for the true spin-glass. It was assumed, that critical exponents $\nu = 1$ and $\Phi = 4$ and that $T_g = 89.75$ K. The solid lines are only to guide the eyes. The lower branch corresponds to $T > T_g$ and the upper one to $T < T_g$. The assymptotic behaviour at t_J decreasing to 0 should have the slope $\nu/\Phi = 1/4$ as the straight solid line in the figure has. This scaling is only an illustration of the spin-glass-like properties of HTCS because the self-consistency of the data with our experimental accuracy is not very sensitive to the choise of ν and Φ.

where

$$t_J(H) = (T_J(H) - T_g)/T_g .$$

After the assumption of $\nu = 1$ [7], which is justified by the results of the computer simulations of spin-glasses we are plotting $\ln t_J(0)/t_J(H) - 1$ on the vertical scale and $\ln (H^2/t_J(H)^{\Phi})$ on the horizontal scale of fig. 5 where all the data of fig. 3 were gathered.

4. Conclusions

The present investigation represents results of frequency dependent AC susceptibility, and the first dynamic scaling analysis of the spin-glass-like properties of the HTCS.

The obtained values of the critical exponent νz are 2–3 times smaller than for the true spin-glasses, but the field scaling exponent ϕ is close to the experimentally and theoretically determined one in spin-glasses. In spite of these problems the presented analysis should be treated as an argument for the common origin of the nonequilibrium properties of the magnetisation of HTCS and of the properties of spin-glasses. Together with the previous AC measurements of $\chi(T)$ [8] they give an indication for the Josephson junctions as the microscopic mechanism of these effects, at most as an important part of them. Further measurements of the AC susceptibility are needed to relate it more closely to the DC magnetisation and to the transport critical current results. The better theoretical background of the spin-glass-like properties of the disordered granular superconductors would be useful.

Acknowledgements

The author is grateful to A. Pajączkowska for the samples, to H. Szymczak, M. Baran and J. Piechota for helpful discussions and to A. Majhofer for the most early suggestions concerning the nature of Josephson junctions. The work has been supported by the Institute of Physics, Polish Academy of Sciences under the contract RPBP 01.9.

References

[1] W.Y. Shin, C. Ebner and D. Stroud, Phys. Rev. B30 (1984) 134.
[2] S.R. Shenoy, Physica B 152 (1988) 72.
[3] V.L. Aksenov and S.A. Sergeenkov, Physica C 156 (1988) 235.
[4] C. Rossel, Y. Maeno and I. Morgenstern, Phys. Rev. Lett. 62 (1989) 681.
[5] A.P. Malozemoff, T.K. Worthington, Y. Yeshurun and F. Holtzberg, Phys. Rev. B38 (1988) 7203.
[6] J. Piechota, Z. Kozioł, A. Pajączkowska and H. Szymczak, submitted to Phys. Status Solidi.
[7] C. Giovannella, C. Chappert and P. Beauvillain, Europhys. Lett. 5 (1988) 535.
[8] Z. Kozioł J. Piechota and H. Szymczak, submitted to J. de Physique.
[9] K.A. Müller, M. Takashige and J.G. Bednorz, Phys. Rev. Lett. 58 (1987) 1143.
[10] Y. Yeshurun and A.P. Malozemoff, Phys. Rev. Lett. 60 (1988) 2202.
[11] J. Mannhart, P. Chaudhari, D. Dimos, C.C. Tsuei and T.R. McGuire, Phys. Rev. Lett. 61 (1988) 2476.
[12] K.W. Blazey, A.M. Porits and J.G. Bednorz, Sol. State Commun. 65 (1988) 1153.
[13] B. Barbara, A.F. Khoder, M. Couach and J.Y. Henry, Europhys. Lett. 6 (1988) 621.
[14] M.A. Dubson, S.T. Herbert, J.J. Calabrese, D.C. Harris, B.P. Patton and J.C. Garland, Phys. Rev. Lett. 60 (1988) 1061. See also discussion in Phys. Rev. Lett. 61 (1988) 1259.
[15] A.P. Malozemoff, L. Krusin-Elbaum, D.C. Cronemeyer, Y. Yeshurun and F. Holtzberg, Phys. Rev. B38 (1988) 6490.
[16] A. Wiśniewski and Z. Kozioł, to be published.
[17] P. Svedlindh, L. Lundgren, P. Nordblad and H.S. Chen, Europhys. Lett. 3 (1987) 243.
[18] F. Gomory and P. Lobotka, Sol. State Commun. 66 (1988) 645.
[19] H. Bouchiat, J. de Physique 47 (1986) 71.