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Laboratori Nazionali di Frascati

LNF-90/027(P)
17 Aprile 1990

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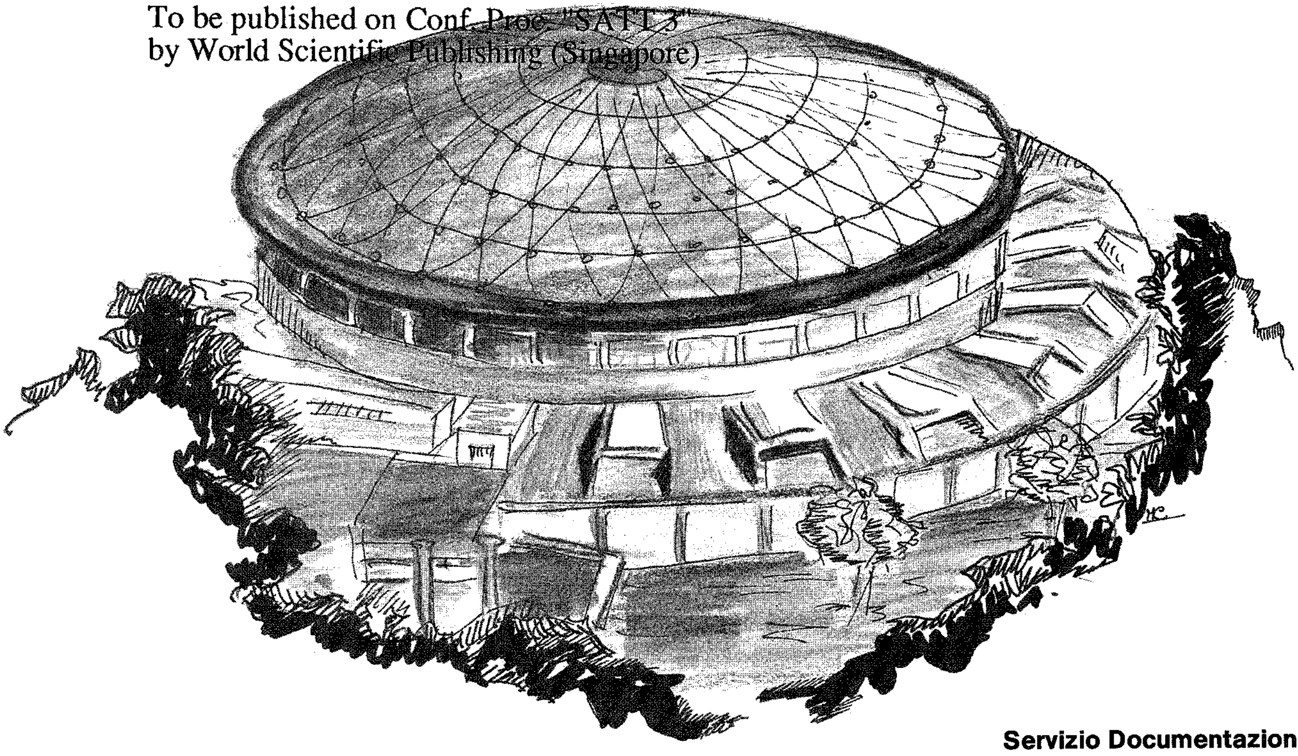
Presented at the
Superconduttività ad Alta Temperatura di Transizione
SATT 3, Genova 12-14 Febbraio 1990

To be published on Conf. Proc. "SATT 3"
by World Scientific Publishing (Singapore)



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FREQUENCY AND FIELD DEPENDENCE OF THE A.C. MAGNETIC SUSCEPTIBILITY OF YBCO PELLETS FABRICATED BY A CITRATE PYROLYSIS AND OZONE ANNEALING

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ABSTRACT

It is well known that low field diamagnetic properties of sintered high T_c superconducting samples are dominated by weak Josephson couplings between grains. Taking into account the effect of shielding currents on the junctions, a simple phenomenological model predicts the frequency dependence of the low field magnetic susceptibility. In this paper we report preliminary results on such measurements performed on synterized samples fabricated by the citrate pyrolysis method. In the magnetic field range between 1 Gauss and 20 Gauss, the temperature dependence of both the real and the imaginary part of the magnetic susceptibility has been studied at some frequency between 10 Hz and 120 Hz. The experimental data are in agreement with the qualitative expected behaviour.

1. INTRODUCTION

High T_C superconductors, in case of a weakly coupled granular structure, undergo a fast reduction of the critical currents and a deterioration of the diamagnetic properties in presence of small magnetic fields. In particular these granular systems show a lower critical field H_{GC1} dependent on the coupling between the grains, and an upper critical field H_{GC2} above which the grains seem to be decoupled from the superconducting point of view [1].

In order to explain this behaviour, it can be easily shown that, for any non simply connected superconducting system, any shielding state is a metastable state[2]. This result is still valid for these granular superconducting systems, which contain a large number of holes or non superconducting regions surrounded by superconducting loops joined by Josephson junctions between grain boundaries. These shielding metastable states decay to stable states that, in case of granular systems at small fields, consist of a complete shielding on the superconducting grains and a complete magnetic field filling of the holes. The decay, from metastable states into a stable state, is obtained by flux penetration through the weakest junction of the superconducting loops. The probability of flux entrap, usually negligible, rapidly increases as the shielding current approaches the maximum Josephson current. Summarizing, for small applied magnetic fields, the shielding currents on the superconducting loops are small and the probability of flux entrap will be negligible. In such case the decay time τ_D from the metastable state into the stable state goes to infinity and the metastable state seems stable. Otherwise if the applied magnetic field is high enough, the shielding currents are close to the maximum Josephson current and the probability of decay from the metastable state into the stable state increases enormously. In this way a time dependent magnetization can be obtained, where τ_D is the decay time from the magnetization curve, corresponding to the complete shielding, to the curve corresponding to the shielding generated by the decoupled grains. From the previous arguments follow that $\tau_D(H)$ goes practically to infinity as the applied magnetic field goes to zero and $\tau_D(H)$ goes to zero as the applied field goes to H_{GC2} , so that H_{GC1} will be determined by the condition that $\tau_D(H_{GC1})$ is of the same order of magnitude of the measuring time τ_M.

The apparent reversible or irreversible behaviour is related to the ratio between τ_M and $\tau_D(H)$. This effect can be experimentally analyzed from static magnetization decay measurements (see A. Siri et al.: this conference [3]), or by the frequency and magnetic field dependence of the a.c. magnetic susceptibility. Indeed for low enough a.c. magnetic fields, if the measurement period τ is such that $\tau < \tau_D(H)$, then, as the frequency increases, a decreasing of hysteretic effects in the magnetization curve is expected. It corresponds to a decreasing of the imaginary part and an increase of the real part of the susceptibility. On the contrary for higher a.c. magnetic fields, at small enough H(t) the condition $\tau < \tau_D(H)$ is still valid, while at higher fields H(t) we have $\tau > \tau_D(H)$. In these cases the hysteretic effects increase as the frequency increases, so that the imaginary part increases with the frequency[2].

2. EXPERIMENTAL RESULTS

Measurements of the frequency and field dependence of the a.c. magnetic susceptibility have been done on YBCO pellets fabricated by using the citrate pyrolysis method and ozone annealing. This method has shown high stability against humidity and both good diamagnetic and mechanical properties. The key points of the method are:

1) a better homogeneous mixing of the components in the correct stoichiometry due to the use of nitrate solutions instead of powders as starting materials,

2) the use of an high oxidizing atmosphere in all thermal processes.

Details of the fabrication method are reported elsewhere [4,5]. Diamagnetic properties of such samples have been investigated in the past by torque measurements [5], and a.c. susceptibility measurements [6,7].

The experimental apparatus, used in this paper for the measurements of a.c. susceptibility at different frequency for various a.c. magnetic field is made of three coaxial coils. The external one generates an optional d.c. magnetic field, the intermediate coil produces the a.c. magnetic field, while the inner one is the pick-up coil. In spite of the higher sensibility of a bridge configuration, the two coils system permits to avoid the frequency dependent balance; moreover the magnitude of the signals does not require high sensibility. The samples have a cylindrical shape with a diameter of 18 mm and an height of 4 mm. In these preliminary measures the sample demagnetization factors have been neglected.

The Fig. 1 shows the temperature dependence of the real and imaginary part of the a.c. susceptibility using an a.c. magnetic field with 10 Gauss amplitude and frequencies respectively of 10, 20, 60, 120 Hz. In our case, the good coupling between grains and the geometrical sample shape determine the presence of a single peak of χ'' generated by macroscopic transport currents which masks the grain peak [1]. In this figure it is evident the shift in temperature of the whole χ'' curve as the frequency increases, corresponding to a sharper diamagnetic transition shown by $\chi'(T)$. These measurements have been performed for several amplitudes of the a.c. magnetic field. In Fig. 2 the temperature value T_p of the χ'' peak as function of both frequency and magnetic field is reported. The temperature value T_p of the χ'' peak has been chosen as reference point for measuring the amount of shift of the entire χ'' curve. We observed similar behaviour for each amplitudes of the applied a.c. magnetic field from 1 up to 20 Gauss.

Further measurements were performed superimposing a d.c. magnetic field up to 100 Gauss to a 1 Gauss a.c. magnetic field. This measurement has been obtained by feeding the external coil, kept at the liquid nitrogen temperature, with the d.c. current and applying the a.c. signal to the intermediate coil as done previously. For a 100 Gauss applied d.c. magnetic field, the Fig. 3 again shows the temperature shift of the entire χ'' curve and a sharper χ' transition at higher frequencies. In this case the phenomenon of the temperature shift of the χ'' peak measured respectively at 120 Hz and 1000 Hz is remarkable.

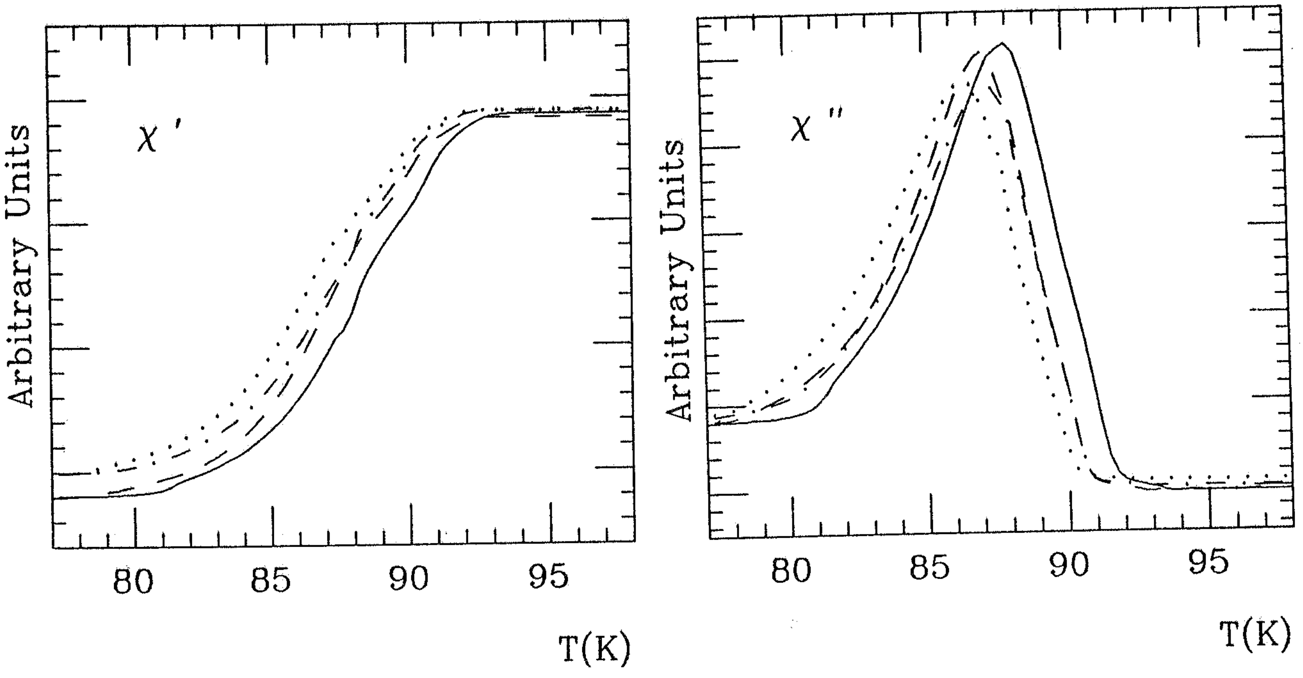

FIG.1 - Temperature dependence of χ' and c" for 10 Gauss a.χ. magnetic field amplitude at different frequencies: ... 10 Hz, _ _ 20 Hz, _ . _ 60 Hz, ___ 120 Hz.

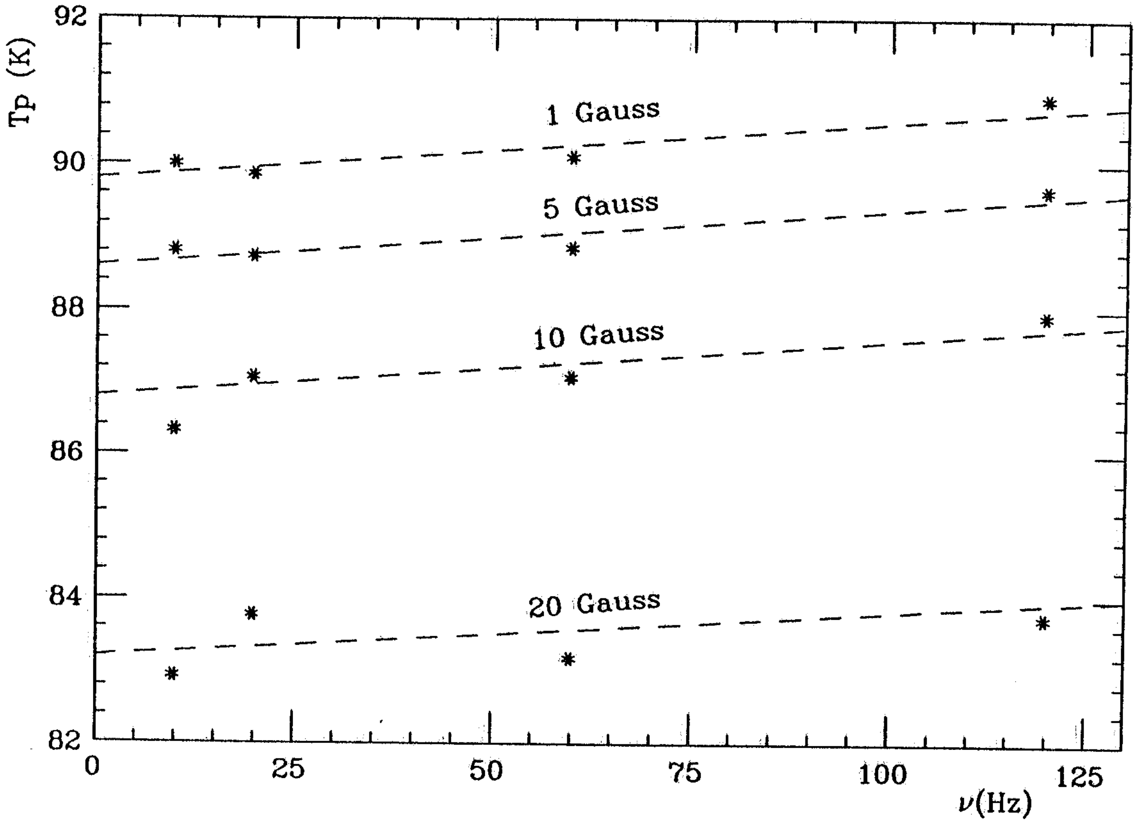

FIG. 2. - Temperature dependence of the χ'' peak for different values of the applied magnetic field as function of the frequency, the dashed line represents only a guide line.

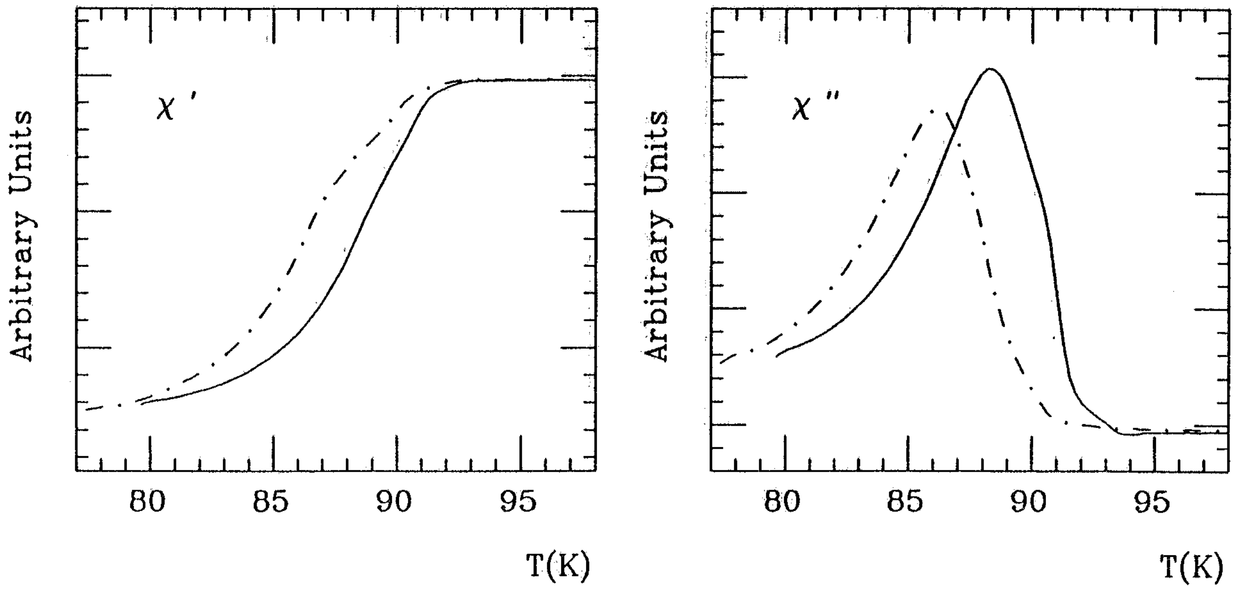

FIG. 3. - Temperature dependence of χ' and χ'' measured with 1 Gauss a.c. magnetic field plus 100 Gauss d.c. at two different frequencies: _ . _ 120 Hz, ___ 1000 Hz.

3. DISCUSSION AND CONCLUSIONS

In spite of amplitude and frequency limits of the a.c. magnetic field and neglecting the demagnetization factors (which cause an enlargement of the inductive transition) the experimental data show a clear displacement at higher temperatures of the χ'' curves when the frequency increases. This behaviour also appears in the measures made with the presence of d.c. magnetic fields. This behaviour is in agreement with the expectation of having a χ reduction at low magnetic field, and a χ'' increase at high magnetic field when the frequency increases. In fact, since all typical magnetic fields go to zero as the temperature goes to T_c, any magnetic field, which is small respect to a typical critical field at low temperature, becomes large as the temperature gets near T_c. In this way, in accordance with the qualitative previsions of the model [2,8], the shift at higher temperatures of the χ'' peak related to the frequency increases corresponds to:

a) a χ'' increase with the frequency for a given temperature higher than the temperature value of the χ'' peak,

b) a χ'' decrease with the frequency for temperatures lower than the temperature value of the χ'' peak.

Indeed these two conditions correspond respectively to the low and high field case.

More careful measurements and further developments of the model are needed for quantitative comparisons.

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