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# Frequency Dependence of the Higher Harmonics Susceptibilities of Hydrogen Loaded and Unloaded Melted YBCO Samples

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## Abstract

We measured the first and higher components of ac susceptibilities of melted YBCO samples before and after hydrogenation by  $\mu$ s pulsed electrolysis. First component was measured versus temperature at fixed frequency while higher harmonics were measured versus frequencies at fixed temperature near critical temperature. All measurements were performed at fixed amplitude of ac magnetic field and zero dc magnetic field. The frequency and temperature behavior of the harmonic components before and after hydrogenation gave us information on possible dynamic losses in comparison to the flux pinning mechanism.

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#### **1. INTRODUCTION**

The measurements of the higher harmonic components  $(\chi'_n + i \chi''_n)$  of  $\chi$ , in the response of high temperature (HTSC) superconductors is explained in the framework of the critical state models [1], but these models do not predict their frequency dependence, while some measurements are instead function of this parameter [2,3,4], probably because the  $\chi_n$  are affected by non–linear dynamic losses. In this contest it is possible to study the effects of the hydrogenation on superconducting properties. For example, hydrogen change the magnetic state of the samples, it can break the grain and worsen the quality of material, but in some case it can enter inside the grains improving the superconducting properties [5]. In order to study this subject, we present measurements and discussions on the behavior of  $\chi''_{1-3-5}$  in function of the temperature and the applied ac magnetic field frequency analyzing YBCO melted samples behavior before and after hydrogenation.

### 2. SET-UP AND PROCEDURE

The ac magnetic susceptibilities were measured using a two–coils coaxial susceptometer. The external coil generates an ac magnetic field and the inner one is used as a pick–up coil. In order to detect the real part  $\chi'$  and the imaginary part  $\chi''$ , we acquired the in–phase and the out–of–phase voltage signals at the pick–up coil by a lock–in amplifier (EG&G 5302). The local earth magnetic field was shielded by  $\mu$ –metal screen. The temperature was measured by two platinum thermometers (PT100) with a temperature controller (DR91C, Lake Shore Cryotronics).

We measured  $\chi_1$  versus temperature at frequency of 107Hz, while the measurements versus frequency was done at fixed temperature near the peak temperature  $T_P$  of the  $\chi''_1$ . The amplitude of ac magnetic field was kept constant at  $H_{ac}$ =7G. The used thermal cycle procedure can be summarized as:

- a) Start–up from room temperature. The samples was cooled down to T=79K with a decreasing rate of 5 K/min with H<sub>ac</sub>=0 (ZFC) and H<sub>dc</sub>=0;
- b) Sample thermalization for one hour at 79 K;
- c) Start measurements. During the measurements (fixed temperature) versus frequency, the temperature variation was better than 0.1K.

The parameters used are following reported:

- 1) The on-set temperature  $T_{on}$  is defined as the temperature corresponding to the beginning of the deviation of  $\chi'$  and  $\chi''$  from the normal magnetic state;
- 2) The  $\chi$ " peak temperature T<sub>p</sub> is defined as the temperature corresponding to the maximum value of it.

In the hydrogen loading procedure [6], the YBCO sample (cylinder  $\Phi$ =4mm, d=5mm, 153µmol) was used as cathode in a solution of LiOH (0.1M). The anode was a grounded coil platinum wire. The electrolytic parameters are:

1) Unipolar voltage pulse with rectangular shape;

2) Voltage peak pulse was settled at 50V, corresponding to about 25A of electrolytic peak current, because the impedance of electrolytic system was about  $2\Omega$ .

- 3) Pulse time width was settled at  $1\mu$ s;
- 4) Repetition rate of pulse was settled at 5KHz;
- 5) The electrolysis time duration was about 120s.

The electric charge passed through the electrodes was 15C. Hence,  $39\mu$ mol of hydrogen was produced on the YBCO surface and the loading ratio H/YBCO was about 0.25 (electrolysis efficiency of 100%).

#### 3. RESULTS AND DISCUSSION

The temperature dependence of  $\chi''_1$  before and after hydrogen loading is shown in Figure 1. The variation of the behavior is related to the change of the YBCO superconducting properties due to hydrogenation (H-YBCO). This effect can be generated only by the hydrogen presence inside the grains. We see the same Ton before and after the loading, but a little increasing of the  $T_p$  is visible and the peak amplitude also, i.e. there is an increasing of the magnetization loop area. Using the Bean model prediction, this evidence is corresponding to a little but significant increasing of the critical current. Therefore the behavior of  $\chi''_1$  is determined by the contemporary presence of hysteretic and flux motion losses, so we need information on higher harmonic components. For this reasons we measured the 3<sup>rd</sup> and 5<sup>th</sup> harmonic components of the susceptibility in function of the ac magnetic field frequencies near the  $T_p$  of the  $\chi''_1$ . These harmonics (imaginary part) are shown in Figure 2 and Figure 3. In  $\chi_3^{"}$  and  $\chi_5^{"}$  behaviors (similar in  $\chi_{3,5}$  real parts) when frequency increases there are oscillations at 30,50,80Hz. The oscillations in higher harmonics are an evidence that the penetrated ac magnetic field  $H_p$  into the sample [7] is connected with the applied ac magnetic field frequency, hence we should take in account the diffusion effects due to the non-linear dynamic losses [8]. These frequencies furnished quantitative information on dynamic losses. Knowing the characteristic time  $\tau = 1/f$ , it is possible to compute the resistivity  $\rho$  and the magnetic diffusivity D in the superconducting state. From the experimental data, the calculated creep resistivity is  $30n\Omega cm$ , this value is less than flux flow processes and magnetic diffusivity is about 2.4 cm<sup>2</sup>/s [8,9].



**Fig.**  $1 - \chi''_1$  vs temperature, (f=107Hz, H<sub>ac</sub>=7Gauss).



Figures 2 and 3, show that the  $\chi_{3-5}^{"}$  have regular behavior near 107 Hz, in this region the higher harmonics are not dependent on the frequency, hence in this case, it is possible to correlate the amplitude of  $\chi_{1}^{"}$  at T<sub>p</sub> with critical state model. In conclusion, we can say that, the hydrogen loadings produce an increasing of the critical current, moreover do not change the resistivity  $\rho$  and the magnetic diffusivity D at this stoichiometric ratio of H/YBCO=0.25.

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