The Influence of Granularity on Dissipation in High Temperature Superconductor

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Abstract— We have investigated AC losses in high temperature superconductors. Granular YBCO samples were prepared by the conventional solid state synthesis process. Individual grains are anisotropic and these polycrystalline samples have randomly oriented c axes. Intrinsic and hysteretic losses were investigated using a Quantum Design PPMS magnetometer. AC susceptibility was measured as a function of magnetic field, frequency and temperature and field history. Three types of temperature dependent loss peaks, T_p's, were observed. The three peak types (in χ'') are attributed to intrinsic (London theory), intergranular Josephson junction, and intragranular pinning loss mechanisms, respectively. They are observed to change systematically as temperature and magnetic field varies. We suggest physical models for the H, T and ω dependence of the loss peaks.

I. INTRODUCTION

The measurement of AC susceptibility, χ is a useful method to identify power losses in superconductor samples [1]-[6]. The AC susceptibility of superconductors is a function of temperature (T), magnetic field (H), frequency (ω) and state (as state we mean the magnetic field history of the material). It can be written as:

$$\chi_{ac} = f(T, H, \omega, \text{state}) = \chi' + i\,\chi'' \qquad (1)$$

where χ' is the real part of χ_{ac} and χ'' is the imaginary part of χ_{ac}.

The imaginary part of χ_{ac}, i.e. χ'', is associated with losses. In a single phase superconductor we expect the following loss peaks in $\chi''(T)$:

(1) The ***intrinsic peak***: represents losses associated with London field penetration, i.e. losses due to the screening of magnetic fields by the superconducting surface currents [7]. This loss peak can be explained in terms of the two-fluid model [7], where the dissipation occurs through AC excitation of normal electrons. The intrinsic loss peak is expected to vary slowly with temperature, magnetic field, and frequency.

(2) The ***intergranular peak***: is due to losses associated with flux penetrating the grain boundaries. This peak can also be called the coupling peak and can be explained by considering the weak links or Josephson junction (JJ) coupling of adjacent superconducting grains. This peak is expected to be observed at low magnetic fields, because of the weak JJ coupling in polycrystalline superconductor [8]. The behavior of this peak as a function of magnetic field is expected to be similar to the behavior of the J_c (critical current density) in polycrystalline materials, and as such also dependent on H, T and the DC magnetic field history of the material (i.e field cooled (FC), zero-field cooled (ZFC) or remanent (R) states) [9]-[11].

(3) The ***intragranular peak***: is due to losses associated with depinning Abrikosov vortices. This peak appears for magnetic fields exceeding H_{c1} ($H > H_{c1}$). It can be explained by the Bean or critical state models [12]-[14].

We have measured AC susceptibility in granular $YBa_2Cu_3O_{7-\delta}$ samples prepared by the solid state synthesis process. AC susceptibility measurements were made at different DC magnetic fields, frequencies and temperatures. The effects of field history was also explored.

II. EXPERIMENT DETAILS

Polycrystalline $YBa_2Cu_3O_{7-\delta}$ samples were prepared by mixing and grinding powders of Y_2O_3, $BaCO_3$ and CuO and calcining for 4 hours at 900°C. The powder was then reground and cold-pressed into pellets (3.2 cm in diameter and 3 mm high). The pellets were then placed in a furnace at 970°C and sintered for 16 hours in air. At the end of the 16 hours, the the samples were furnace-cooled to room temperature, at a rate of ~130°C/hr.

We have performed the AC susceptibility measurements using a Quantum Design Physical Property Measurement System (PPMS) magnetometer which has a sensitivity of 2×10^{-8} emu at 10 kHz and temperature stability of 0.1 K at 300 K and of 0.01 K at 5 K. The sample used for all measurements was an irregularly shaped piece with approximate dimensions of 6x3x2 mm³.

All the samples were mounted in straws and centered in the secondary coil of a mutual inductance coil set using an AC magnetic field amplitude of 10 Oe, a frequency of 5 kHz and temperature of 70 K. This temperature was chosen to guarantee that the sample was in its superconducting state, therefore supplying a more reliable signal for the centering procedure. When the sample became superconducting both the real (χ') and the imaginary (χ'') components were measured with a Digital Signal Processor (DSP) chip rather than a lock-in

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amplifier, which improves signal to noise performance over analog filters [15].

In this work, we chose to use an AC magnetic field amplitude of 10 Oe rms for all the measurements. We performed 3 types of experiments:

(1) χ' and χ'' were measured as a function of frequency as the sample temperature was slowly raised from low temperature to above T_C. This was done for frequencies varying from 10 Hz to 10 kHz, and both in a static DC field (100 Oe, 500 Oe and 10 kOe) and without a DC magnetic field being applied;

(2) χ' and χ'' were measured as a function of DC magnetic field from low temperature to above T_C. Here the sample was cooled to 5 K in a zero DC magnetic field and then measured in a constant DC magnetic field as the temperature was raised from low temperature to above T_C (Zero Field Cooling, ZFC) and then cooled back to low temperature (Field Cooling, FC). This was done for DC magnetic fields varying from 1 Oe to 50 kOe at a frequency of 10 Hz,

(3) χ' and χ'' were measured as a function of temperature as a DC magnetic field was applied above T_C and turned off at 5 K, before starting to measure the ac susceptibility (remanent, R, field measurement). This was done for a bulk sample with DC magnetic field of 10 kOe and frequency of 10 Hz.

III. RESULTS AND DISCUSSION

Typical χ' and χ'' measurements in zero DC magnetic field are shown in Fig. 1 for a bulk sample. In $\chi''(T)$ we can identify two peaks that reflect power losses in the material. We can see one peak near T_C (the intrinsic peak) and another at lower temperature (the coupling peak). It is apparent that the inflection points observed in the real part of the susceptibility ($\chi'(T)$), roughly correspond to the position of peaks in $\chi''(T)$. This reflects Kramers-Kronig relations between χ' and χ''. We can also see that the T_C of this sample is near 90 K.

In Fig. 2 we show examples of $\chi(T)'$ for three different values of the DC magnetic field. We observe three different field regimes in which the behavior of $\chi(T)''$ changes:

(1) **low magnetic field region ($H < H_{c1}$)**: in this

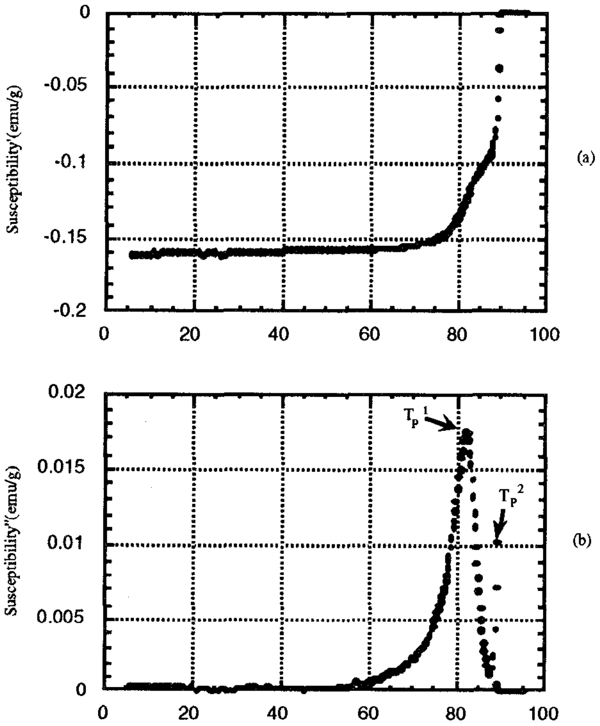

Fig. 1. Typical χ' and χ'' measurements for $YBa_2Cu_3O_{7-\delta}$. The measurement was performed in zero DC magnetic field being applied with an AC amplitude of 10 Oe and frequency of 10 Hz.

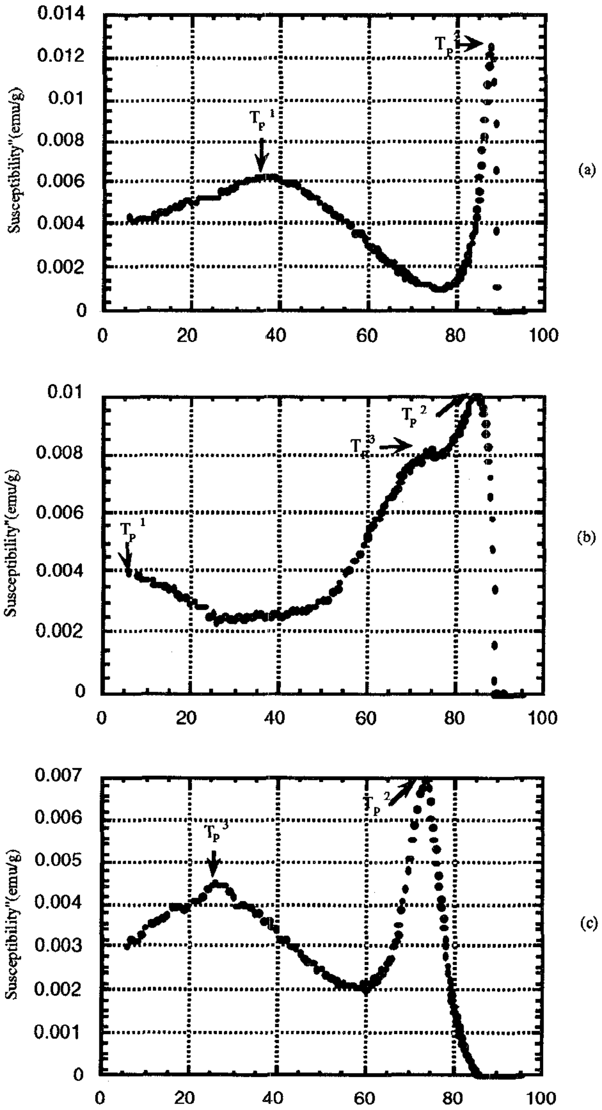

Fig. 2. Examples of χ'' measurements for the three different regions of the magnetic field: (a) χ'' measurement for DC magnetic field of 100 Oe (the low magnetic field region); (b) χ'' measurement for DC magnetic field of 500 Oe (the intermediate magnetic field region) and (c) χ'' measurement for DC magnetic field of 5 T (the high magnetic field region). All three ZFC measurements were performed at 10 Hz with an AC field amplitude of 10 Oe.

region we can observe only the intrinsic, and the intergranular JJ peaks, because no magnetic flux penetrates the grains there is no intragranular loss peak. Note that because of single crystal anisotropy and randomly oriented c-axes in the polycrystalline materials, $\mathbf{H}_{C1}$ is not precisely defined.

(2) **intermediate magnetic field region ($\mathbf{H > H_{C1}}$):** we can identify three loss peaks for $\chi''(T)$. In this region as magnetic flux lines penetrate the grains the intragranular loss peak becomes evident. The intergranular JJ loss peak shifts rapidly to lower temperatures as field is increased. The intrinsic peak is much more slowly varying with applied DC field than the intra- and inter-granular peaks.

(3) **in the high magnetic field region ($\mathbf{H >> H_{C1}}$):** the magnetic field destroys the JJ coupling between the grains and the coupling peak is no longer observed. This is similar to the behavior that we see in ground superconducting powder where only the intrinsic and the pinning peaks are observed. The position of the pinning peak, T_P^3, is also strongly field dependent consistent with the field dependence of the critical state penetration.

Table I summarizes the DC field dependence of the three loss peaks at fixed AC frequency (10 Hz) and amplitude (10 Oe) for both the ZFC and FC states, respectively. For both ZFC and FC measurements we observe that the three peaks in $\chi''(T)$ are shifted to lower temperatures as a function of increasing DC magnetic field. The intrinsic peak, however, varies much more slowly (reflecting $T_C(H)$) with the DC field than the other two peaks. We believe that the field dependence of the intergranular peak, T_P^1, reflects the strong field dependence of the intergranular critical current density which is dominated by weak link effects. It is concluded that the intergranular JJ coupling which gives rise to this effect is strongly field dependent. The field dependence of the intragranular peak, T_P^3, reflects the field dependence of the intragranular critical current density and is therefore a measure of pinning within individual grains.

TABLE I
FIELD DEPENDENCE OF THE AC LOSS PEAKS T_P^1, T_P^2, AND T_P^3, AS A FUNCTION OF APPLIED DC FIELD IN FC AND ZFC STATES FOR A POLYCRYSTALLINE Y123 SAMPLE.

	ZFC measurements			FC measurements		
H	T_P^1(K)	T_P^2(K)	T_P^3(K)	T_P^1(K)	T_P^2(K)	T_P^3(K)
1 Oe	82.0	89.0	-	82.2	89.0	-
10 Oe	77.3	88.8	-	77.5	88.8	-
50 Oe	57.0	88.2	-	63.5	88.3	-
100 Oe	38.0	87.5	-	55.0	87.6	-
300 Oe	10.0	86.3	75.5	37.0	86.2	70.5
500 Oe	6.0	84.6	73.0	32.1	85.0	69.9
0.1 T	~6	@	@	12.1	@	69.4
0.5 T	< 5	85.3	59.0	9.0	85.2	64.5
1 T	< 5	83.4	47.8	< 5	83.4	57.0
2 T	< 5	80.3	33.1	< 5	80.5	37.9
5 T	< 5	73.5	26.0	< 5	73.4	26.0

@ designates that for this value of DC magnetic field there is overlaping of the peaks T_P^2 and T_P^3.

We also observe that the peak widths for $\chi''(T)$ are markedly different. The intrinsic peak, T_P^2 is quite sharp even in a strong DC magnetic field. The hysteretic peaks, T_P^1 and T_P^3, are much broader. The width of the intergranular peak, T_P^1, is interpreted to reflect the fact that in our polycrystalline samples we have a distribution of JJ couplings between adjacent grains. The width of T_P^3 is interpreted to reflect the random distribution of c-axes in the samples and therefore pinning potentials. It is well known that single crystalline $YBa_2Cu_3O_{7-\delta}$ is strongly anisotropic (J_C along the c axis is greater than J_C perpendicular to the c axis [8]).

We also observed that the peaks in $\chi''(T)$ for the ZFC measurements do not coincide with the respective peaks for the FC measurements. The AC response in these materials is therefore demonstrated to be a function of field history. Table I shows the values of T_P^1 (intergranular peak), T_P^2 (intrinsic peak) and T_P^3 (intrgranular peak) for both ZFC and FC measurements. This field history dependence of the loss peaks is indicative of hysteretic effects. In the case of the intergranular peak, T_P^1, the history dependence is consistent with hysteresis in the intergranular current density $J_C(H)$ which is routinely observed in granular samples [6]-[9]. In the case of the intragranular peak, T_P^1, the history dependence is consistent with hysteresis in the intragranular current density, as predicted by the Bean and critical state models [10]-[12].

Fig. 3(a) illustrates the frequency dependence of both T_P^1 (the coupling peak) and T_P^2 (the intrinsic peak of χ''). The frequency dependence of the intragranular pinning peak will be discussed in future work. As the AC drive frequency was increased, the intrinsic peak shifted only slightly. To the accuracy of our measurements it appears to be invariant with frequency. On the other hand, the intergranular coupling loss peak varied systematically with frequency showing shifts to higher frequency at higher temperatures. The shift was more dramatic when we superimposed a DC magnetic field on the AC magnetic field.

Fig. 3(b) illustrates and inverted Arrhenius plot of the inverse of the peak position $1/T_P^1$ (the coupling peak) as a function of the log(frequency). This show the coupling peak to be thermally activated, with the activation energy barrier strongly field dependent. This thermally activated response also depends on the field history [16]. It is similar to the behavior reported by Nikolo and Goldfarb [3]. Given the previus discussion, however, it seems suspect to attribute this to flux creep. On the other hand, given the parallels between the field and temperature dependence of the intergranular loss peak, it seems more appropriate to explain the thermally activated response with relaxation in a coupled Josephson junction network. The details of such a description await further theoretical developments. However, we argue that these observationsadd to the body of evidence suggesting polycrystalline ceramic HTSC materials to brhave as weak link networks.

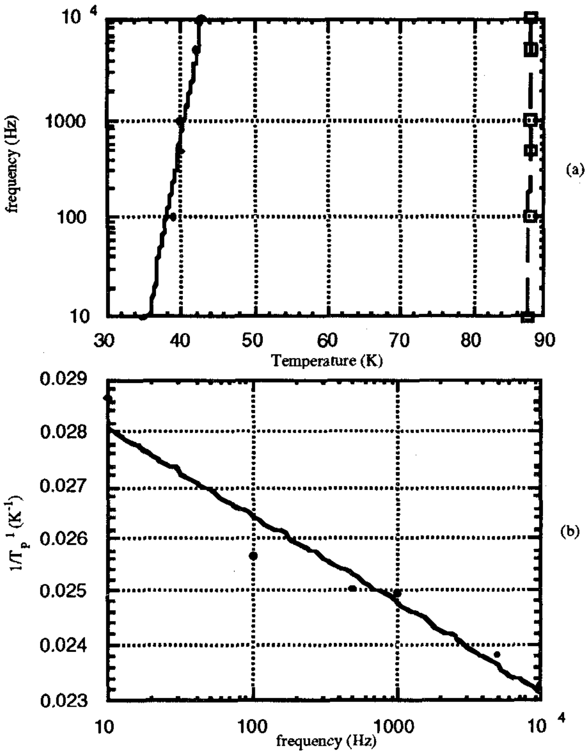

Fig. 3. (a) Closed circles show the frequency dependence of the intrinsic peak, T_P^1 and the open squares show the dependence of T_P^2 with frequency. (b) $1/T_P^1$ as a function of frequency showing activated behavior. These measurements were performed in a 100 Oe DC field, 10 Oe AC amplitude and frequncy of 10 Hz.

IV. CONCLUSION

We have observed the field, field history and frequency dependence of the intrinsic, intergranular and intragranular AC loss peaks for a polycrystalline $YBa_2Cu_3O_{7-\delta}$ sample. The intrinsic peak is slowly varying as a function of field and frequency, while the inter- and intra-granular loss peaks show a stronger field and frequency dependence. The inter- and intra-granular loss peaks are broader indicative of distributed losses. The field history dependences of the inter- and intra-granular loss peaks are attributed to hysteretic effects. The results confirm the importance of both flux pinning and intergranular (Josephson junction) coupling in the loss peaks in polycrystalline HTSC materials. The latter is evident only in granular materials. The loss peak is shown also to be thermally activated consistent with relaxation in a weak link network.

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