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Magnetic Characterization of Differently Processed YBaCuO-Ceramics

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Abstract

Measurements of the fundamental ac-susceptibility provide a sensitive tool to study the shielding behavior of ceramic high-T_c superconducting material. In this work, ac-susceptibility data for differently prepared YBaCuO-samples are presented. From the data, information can be extracted about an effective grain size, which is not necessarily identical to the optically determined particle size. It is demonstrated that the data allow an estimation of the bulk quality which depends strongly on the preparation conditions.

1. Introduction

It is well established both experimentally and theoretically that the shielding behavior of a HTSC ceramic is determined by the intergranular contribution of the Josephson network as well as by the shielding of the individual grains [1-3]. At low temperatures and fields, the current flows via superconducting weak links, thus the sample is macroscopically shielded, provided the grains are in good contact with one another. In an ac-susceptibility experiment, this behavior results in diamagnetic shielding ($\chi' = -1$) and a vanishing imaginary part $\chi''(T)$. The imaginary part is a measure for magnetic ac-losses in the sample. As a result of increasing the temperature and/or the external magnetic field, magnetic flux penetrates the sample above $H_{c1J}(T)$, the Josephson lower critical field. In this case, one observes magnetic losses, i.e. a non-vanishing imaginary part of the ac-susceptibility. If, in addition to the ac amplitude, an external dc field is applied, the induced signal corresponds to the response of the partially decoupled weak link system on the exciting ac amplitude. Thus, from a measurement at fixed H_{dc}, the fraction of the shielded volume can be extracted. By comparison to the results for a corresponding powder sample information can be obtained on the distribution of the strength of coupling between weak links as a function of H_{dc} [4]. If the network is completely decoupled, only the grains contribute to the shielding signal of the sample. Two cases must be considered:

- The average diameter of the grains is large compared to the magnetic penetration depth $\lambda_0 = \lambda(T = 0)$. In this case, intragranular losses become observable, resulting in a second maximum of $\chi''(T)$ at temperatures close to T_c.
- If the grain size is of the same order as λ_0, the magnetic field varies smoothly over the grain cross section. A small field independent real part ($\chi' < 0$) at temperatures close to T_c and zero losses are characteristic for such a behavior. If $\chi'(T)$ is measured as a function of temperature for a powdered sample, a fit to the data according to the empirical two-fluid expression allows the calculation of an effective grain size, providing λ_0 is known [4]. In the case of very small grains ($d \ll \lambda_0$) the magnetic field penetrates the grains almost completely. This magnetic transparency results in an absence of the intragranular contribution to the ac-susceptibility.

2. Experimental

The ceramic samples were prepared by a tape casting technique [5,6] starting from powder with the composition $YBa_2Cu_3O_{7-x}$. The powder itself was prepared by a special reaction spray process, described in [4]. Two powders with different grain sizes were used for the preparation of the ceramic samples. The mean grain diameter of the powder was determined by light scattering and was evaluated to about 3 µm and 5 µm, respectively. Two of the sintered samples were obtained by recycling, i.e. grinding of already sintered material and then repeating the sintering process. Some samples were prepared using additives such as magnetite, teflon and silver to study their influence on the transport and magnetic properties. The ceramics were characterized by x-ray diffraction, electrical resistivity (T_c, j_c(77 K)) and

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SEM-analysis. The ac-susceptibility experiments were performed with a computer-controlled vector-lock-in susceptometer, which is specially optimized for small samples [7,8]. The sample is mounted on a sapphire holder in the center of a pair of Helmholtz coils, which allow to apply ac amplitudes in the range of 0.01 Oe up to 50 Oe rms with frequencies of $31\,Hz < f < 3\,kHz$. Additionally, a dc field up to 120 Oe can be superposed in the same coils. A pair of oppositely wound coils is used to pick up the signal. As all coils are at constant temperature during the measurement, the apparatus allows a proper phase setting and minimizes thermal drifts. The data were obtained by measuring the real (χ') and imaginary (χ'') part of the ac-susceptibility as a function of temperature for different ac amplitudes or at a small fixed ac amplitude, typically 0.1 Oe, for different superposed dc fields.

3. Results and Discussion

The susceptibility data can be divided into four characteristic groups, which can be distinguished clearly with regard to their intragranular behavior. An essential feature is the grain size, which depends e.g. on the sintering conditions and on additives to the powder. In the following, each case will be discussed separately. The samples are denoted as F1 to F4.

In figure 1, the real and imaginary parts of the ac-susceptibility are plotted vs. temperature for the tape F1. Curves for various ac amplitudes are labeled in the figure. The data show the typical behavior of a YBaCuO-ceramic with a grain diameter large compared to the penetration depth of the magnetic field. For an ac-amplitude not too small, e.g. $H_{ac} = 20$ Oe, two maxima at different temperatures can be clearly distinguished in the imaginary part which are attributed to the weak link network and to the grains, respectively, as discussed above. The decoupling temperature of the network, $T_{c,J}$ and the critical temperature of the grains, $T_{c,g}$, are marked in the figure. The assumption of a weak link network for temperatures below $T_{c,J}$ is supported by the strong field dependence of both real and imaginary part. The limiting curve, where the grains are completely decoupled, is given for $H_{ac} = 20$ Oe (see figure 1). The appearance of a second maximum in $\chi''(T)$ for sufficiently large amplitudes ($H_{ac} = 20$ Oe) and temperatures close to T_c indicates large grains, i.e. a diameter large compared to λ_0. For small ac amplitudes however, e.g. $H_{ac} = 0.1$ Oe, the intragranular loss maximum is not visible. This low loss together with the sharp transition in $\chi'(T_{c,J} < T < T_{c,g})$ which does not depend significantly on the ac amplitude up to $H_{ac} = 20$ Oe, points towards a flux pinning dominated dissipation mechanism within the grains and in particular to an absence of intragranular weak links.

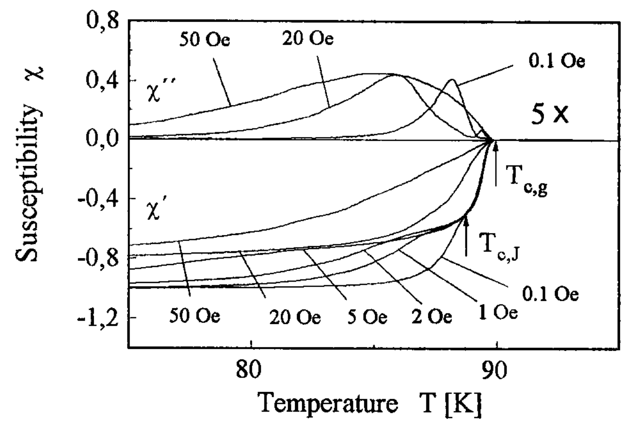

Figure 1. Real and imaginary parts of the ac-susceptibility vs. temperature for the tape F1. Curves for different ac amplitudes H_{ac} are labeled in the figure. Note that the imaginary part is upscaled by a factor 5.

From the weak dependence of χ' on the amplitude H_{ac} at temperatures below 80 K, one concludes on a high shielding quality of this ceramic sample. This correlates with the fact that x-ray diffraction measurements performed on this sample showed a pure orthorhombic phase. The portion of the non-superconducting phase Y_2BaCuO_5 is estimated to be below 5 %. This is also in agreement with the high critical temperature of 92.2 K, which is the highest of all ceramics investigated in this work.

It is interesting to mention that another sample, which was prepared under the same conditions, but with the addition of 0.75 weight percent teflon, shows a very similar behavior of χ_{ac} as F1.

For the preparation of sample F2, the above mentioned recycling technique was used, starting from the finer grained of the two powders. Figure 2 shows the results. Here, the real and imaginary parts are given as a function of temperature for different dc fields superposed to a small ac amplitude of $H_{ac} = 0.1$ Oe. The figure shows the typical behavior of a ceramic, where the grain size is of the order of λ_0. It is evident that there is a region near the critical temperature of the grains, where the real part is field independent and the imaginary part is zero. To prove this observation, measurements were performed at different ac amplitudes without an applied dc field. For amplitudes $H_{ac} \geq 10$ Oe, the absolute value of the real part is suppressed gradually for $T_{c,J} \leq T \leq T_{c,g}$, together with a very low magnetic loss component. This behavior is probably a result of the non linearity at large ac amplitudes.

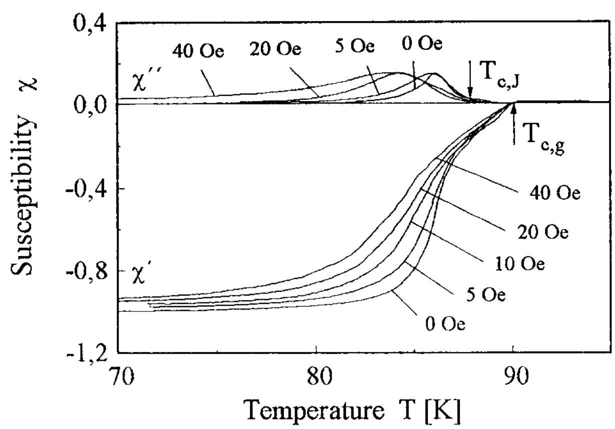

Figure 2. Real and imaginary part of the ac-susceptibility for the sample F2 as a function of the temperature. Different dc fields H_{dc} are labeled. The ac amplitude is fixed at a value $H_{ac} = 0.1$ Oe.

The interpretation concerning the grain size is supported by the low value of the mean grain diameter of the powder used to prepare this ceramic which has an average magnitude of 3.2 μm. This value is clearly below that for sample F1. The interpretation of the ac data even allows the conclusion of an 'effective' grain diameter in the sintered material which is smaller than the value obtained from light scattering data. This means that the particles can be conceived as an agglomeration of even smaller sub-grains.

Sample F3 was prepared following the ordinary sintering process, but now with the addition of 2-5 w.-% grinded Fe_3O_4 (magnetite) to the starting powder. An influence on the magnetic properties above T_c could not be detected within the apparative resolution. In figure 3, the real and imaginary parts are plotted vs. temperature for different ac amplitudes labeled in the figure. A strong field dependence of both χ'(T) and χ''(T) can be recognized, which again indicates that the shielding behavior is determined by weak links. At 77 K, the real part is suppressed to half of its value by an ac amplitude of only 0.5 Oe. Another characteristic feature of the sample is the lack of an intragranular contribution to the ac-susceptibility. This points towards very small grains ($d << \lambda$) which form a Josephson network, but are magnetically transparent even for smallest fields. In the case of sample F3, the addition of Fe_3O_4 possibly leads to an embedding of the material into the grains.

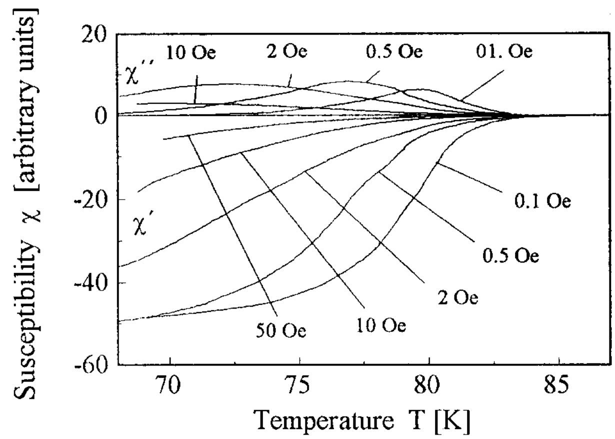

Figure 3. Series of real and imaginary parts as a function of temperature for the sample F3. Different ac amplitudes are marked in the figure.

The grains thus consist of a sub-network, leading to an effecive grain size, clearly below λ_0.

Sample F4 was prepared by applying the recycling procedure and starting from the same fine powder as F2. The mean grain diameter of the powder is 3.1 μm and thus is of the same magnitude as in sample F2. However, x-ray diffraction shows stronger peaks from the non-superconducting tetragonal phase and a weak (001)-texture. In figure 4, the susceptibility data are presented. In this case, the dc field is varied in the range of 0 Oe to 80 Oe at constant ac amplitude of $H_{ac} = 0.1$ Oe. The real and imaginary parts are plotted as a function of temperature. A striking feature is the existence of two loss peaks at different temperatures (for example for $H_{dc} = 5$ Oe) together with a field-independent real part close to T_c, which again indicates an effective particle size comparable to λ_0. For temperatures lower than $T_{c,J} \approx 86$ K, the real part shows a strong dependence on the applied dc field. This behavior can also be attributed to weak links, which are decoupled even at low fields. The reason for a further loss maximum in χ''(T) at low temperatures might be a second, possibly oxygen-deficient phase, which becomes superconducting below about 74 K.

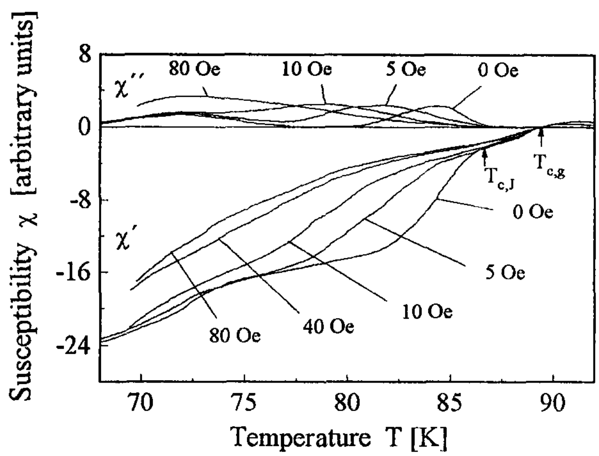

Figure 4: Ac-susceptibility data for sample F4. Real and imaginary parts are plotted vs. temperature for different dc fields, superposed to an amplitude of $H_{ac} = 0.1$ Oe.

The observed increase of the shielding for $T \leq 74$ K can only be explained by assuming that the second phase is located at the sample surface. The superconducting transition of this phase leads to additional coupling of grains and therefore to an improvement of the shielding. A possible reason can be oxygen out-diffusion from the sample surface during the sintering process.

The ability of the ceramic material to shield alternating fields is of interest for applications. Furthermore, a certain interrelation between this ability and the critical current density j_c as determined by transport measurements can be found. The ceramic F1, for example, exhibits the highest $j_c(77\ K)$ of all samples investigated in this work: j_c=200 A/cm^2. The shielding of an ac amplitude of 5 Oe amounts to 95 % at 77 K. Sample F3, which was prepared by the addition of Fe_3O_4, however, shows only a shielding of 75 % as response to an even smaller ac amplitude of 0.1 Oe. In fact, the critical current density was found to be only j_c=70 A/cm^2. This correlation can be understood if the Josephson network is considered to be responsible for carrying the shielding currents as well as the transport current. It should be mentioned, however, that both experimental methods can easily result in different values because of a possible lateral inhomogeneity of the samples.

4. Conclusions

In ac-susceptibility experiments performed on HTSC ceramics, the contributions of the Josephson network and of the grains to the shielding can be clearly distinguished. In the present work it is demonstrated that an effective grain size can be deduced from measurements on YBaCuO-ceramics which were prepared by a special tape cast technique. The extracted effective grain size must not necessarily correspond to the particle size as observed by optical experiments (light scattering on the powder) or REM-images of the ceramics. It rather depends strongly on the processing parameters such as additives and sintering conditions. The admixture of Fe_3O_4 to the powder results in a drastic reduction of the grain diameter and also leads to weak intergranular coupling. As a consequence, the resistively determined critical current density is lowered. The addition of teflon (0.75 w. %), however, shows no significant influence on the shielding as well as on the transport properties.

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