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Role of grains and weak links in the nonlinear magnetic response of Y-Ba-Cu-O

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We present a study of the field dependence of the remanent nonlinear magnetic response induced in sintered Y-Ba-Cu-O samples by momentary application of a dc magnetic field H. The results show that, contrary to theoretical predictions, the remanent nonlinear response depends strongly on H. We attribute this behavior to weak links subjected to internal fields induced by flux trapped within the grains. To support this view, we present an extensive comparison between ac and dc data in the same samples. This comparison clarifies the role of grains and of weak links in various field regimes, and demonstrates the relationship between the remanent nonlinear response and the trapped flux.

The magnetic properties of Y-Ba-Cu-O have been widely investigated using both ac and dc techniques.[1] However, in many cases, it is unclear whether the measured properties are characteristic of weak links or of the bulk material. Identification of the contributions of the weak links and grains to the measured properties has been the goal of many studies.[2] Goldfarb *et al.*[3] have reported two peaks in measurements of the out-of-phase susceptibility versus temperature in sintered Y-Ba-Cu-O. They associated the lower peak with intergranular and the upper peak with intragranular material in their samples. Shinde *et al.*[4] have confirmed the existence of two components in the ac response and studied their relative strength by comparing ac and dc data. Muller[5] has analyzed the complex susceptibility data of sintered Y-Ba-Cu-O on the basis of the critical state model, assuming different intergranular and intragranular pinning forces. The predictions of this model for the field and temperature dependence of the complex susceptibility were found to be in good agreement with the experimental data.

In this paper, we examine the intergranular and intragranular contributions to the *nonlinear* magnetic response of sintered Y-Ba-Cu-O. The nonlinear magnetic behavior of high-temperature superconductors has recently been the topic of several theoretical and experimental studies. Experimentally, the nonlinear behavior is indicated by the appearance of harmonic components in the magnetic response of the material to sinusoidal fields.[6-11] The origin of the nonlinear response is still controversial. Jeffries *et al.*[6] related the nonlinear magnetic behavior of high-temperature superconductors to the weak links. According to their model, the magnetic nonlinearity results from the nonlinear relationship between the Josephson current in supercurrent loops with weak links and the magnetic flux enclosed by the loops. Xenikos and Lemberger[11] observed nonlinear magnetic behavior in Y-Ba-Cu-O single crystals and interpreted it as a consequence of the nonlinear magnetoresistance of crystals near T_c due to flux creep. Ji *et al.*[9] have extended the Bean critical-state model[12] and attributed the harmonic generation to the hysteretic, nonlinear relationship between the magnetization and the external field due to flux pinning. In a recent work, Ishida and Goldfarb[10] studied the field and temperature dependence of the harmonic signals in Y-Ba-Cu-O ceramics, and showed good agreement between the predictions of the model of Ji *et al.* and the experimental data.

The magnetic fields used in most of the experiments described above were rather small, of order 10 Oe. Thus, only the intergrain contribution to the nonlinear response was examined. Moreover, in these studies the amplitude of the ac field was of the same order as the maximum dc field, therefore the ac field cannot be considered as a small perturbation to the dc field. In the present work we use a relatively small ac field ($h_{\mathrm{ac}} \simeq 50$ mOe) and extend the measurements to higher dc fields, up to 2 kOe. This choice of fields enables us to distinguish clearly between intergrain and intragrain contributions to the nonlinear response. In addition, we report new measurements of the remanent nonlinear response induced by momentary application of a dc magnetic field. In this experiment, the dc field is applied for a few seconds (typically 20 s), and the remanent nonlinear response is measured after turning off this field.[13] The field dependence of the remanent nonlinear response is not predicted by any one of the above models. We outline an explanation of these results within the framework of the Bean model by taking into account the field induced in the weak links by flux which is trapped within the grains.[13,14] This explanation is supported by an extensive study, in the same samples, of the magnetic response to dc fields, and of the remanent magnetization measured after turning off this dc field. We find a close relationship between the field dependencies of the remanent magnetization and the remanent third-harmonic component in the ac response. In addition, we apply the Bean model for a system composed of grains and weak links[5] and reproduce the main features which are observed experimentally.

Two Y-Ba-Cu-O ceramic samples were used in these experiments. Sample No. 1 was made of micrograins of Y-Ba-Cu-O powder, 3–10 μm in size, which were produced by crushing a ceramic sample sintered at 940 °C. The powder was pressed at 10 tons/cm^2 to form a cylinder (diameter, 5 mm; height, 5 mm). This cylindrical pellet was sintered at 900 °C, $\simeq$40 °C below the usual sintering temperature, in order to weaken the connectivity between grains. Sample 2 was produced in the same firing process, but the powder was coated in an electroless process with silver (6% by weight). For details of preparation see Ref. 15. Ag in Y-Ba-Cu-O ceramics enhances critical currents and connectivity between grains.[15,16] Thus, this particular preparation technique yields samples in which the effect of weak links can be controlled. In order to emphasize the effect of the weak links we choose in the present article to focus on sample 1. Sample 2 serves here as a reference to a "good" sample which exhibits more familiar magnetic behavior.

The nonlinear magnetic response was characterized by measuring the amplitude of the third-harmonic component in the response of the material to a small sinusoidal field. The measuring circuit consisted of a primary coil coaxial with a pair of balanced coils, one containing the sample. The primary coil induced in the secondary coils a sinusoidal field of amplitude 50 mOe at a frequency of 20 kHz. The response of the material was monitored by measuring the power spectrum of the off-balance voltage induced in the coil pair using an HP3585 spectrum analyzer. dc magnetic measurements were performed using a commercial SHE superconducting-quantum-interference-device magnetometer with the field oriented along the cylinder axis. All dc and ac measurements have been performed after cooling the sample in zero field to the measurement temperature $T=77$ K. At this temperature the field H is increased in steps, but after each step the field is reduced to zero. For each field H, data is recorded with the field on, and then the remanent value is measured when the field is switched off.

The squares in Fig. 1 show the amplitude V_3 of the third-harmonic component in the ac response of sample 1 at 77 K, as a function of the applied dc field. The field-induced third-harmonic signal exhibits two peaks: a narrow peak around 100 Oe and a broad peak around 1 kOe. The remanent third-harmonic response (triangles in Fig. 1) increases slowly with the field and reaches a saturation value at a field of about 250 Oe. For a better understanding of these data, additional measurements of the dc magnetization and the remanent magnetization were performed on the same sample. The results of these measurements are illustrated in Fig. 2. In the low-field limit, up to a field of about 20 Oe, the magnetization M (squares in Fig. 2) increases rapidly with the field with an average slope of roughly 30% of the full Meissner effect. Above 20 Oe there is a clear crossover to a linear dependence of the magnetization on the field with a moderate slope. Above approximately 80 Oe the magnetization behaves in a nonlinear fashion; it decreases to a minimum value around 200 Oe, and increases slowly thereafter. The remanent magnetization (triangles in Fig. 2) exhibits a two-step behavior. It sharply increases with the field in the range 0–20 Oe, and becomes independent of the field in the range 20–80 Oe. Above 80 Oe the remanent magnetization slowly rises to a saturation value.

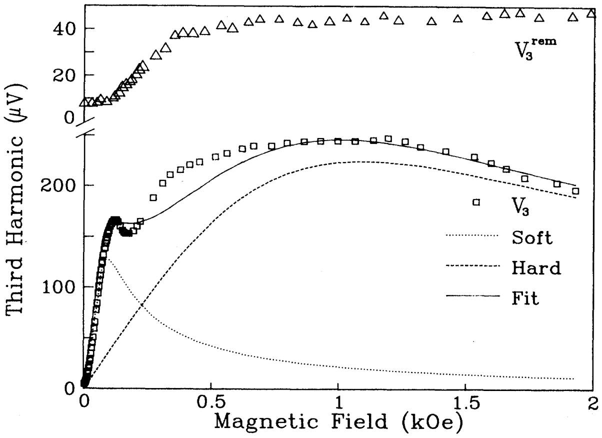

FIG. 1. Amplitude of the third-harmonic component (squares) and the remanent third-harmonic signal (triangles) in the ac response of sintered Y-Ba-Cu-O (sample 1) at 77 K as a function of the applied dc field. The solid line is a theoretical fit, calculated as superposition of contributions from weak links (dotted line) and grains (dashed line).

The dc magnetization data clearly demonstrate the granular nature of the sample. The initial change of the magnetization up to $H=20$ Oe indicates a partial shielding of the sample by both the weak links and the grains. In this range the shielding of the grains is complete while flux lines partially penetrate the weak-link network. Thus, the increase of the remanent magnetization in this range indicates flux trapping within weak links. Above 20 Oe the dc field penetrates the entire weak-link network

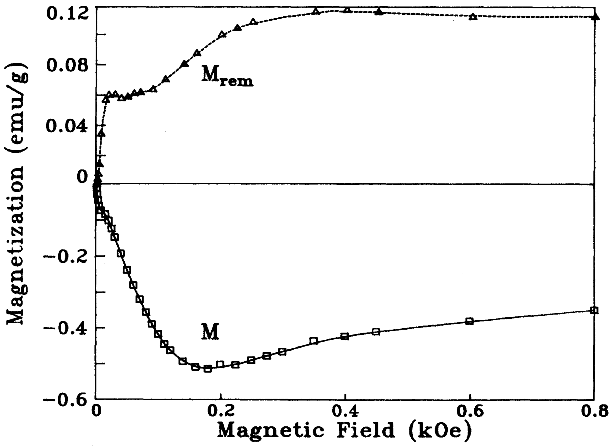

FIG. 2. Field dependence of dc magnetization (squares) and remanent magnetization (triangles) of sintered Y-Ba-Cu-O (sample 1) at 77 K.

while the grains are still shielded up to 80 Oe. As expected, there is no increase in the remanent magnetization in this range. Above 80 Oe, flux lines start to penetrate the grains, and the remanent magnetization increases as a result of flux trapping within grains. At a field of about 250 Oe, corresponding to penetration of the dc field throughout all the grains, the remanent magnetization reaches a saturation value.

In the interpretation of the ac data we apply the results of the critical-state model to both components of the system, namely the weak links and the grains. According to this model, when the ac field partially penetrates a homogeneous sample (a slab or a cylinder), the amplitude of the third-harmonic response V_3 is inversely proportional to the critical current density J_c. When the sample is fully penetrated by the ac field, V_3 is directly proportional to J_c. In a granular sample, one must consider both the intergrain and the intragrain current densities J_{c1} and J_{cg}, respectively. It is clear that below 80 Oe the third-harmonic response is dominated by the weak links, since in this range the grains are completely shielded. Thus, the initial sharp increase of V_3 with the field reflects a strong decrease of J_{c1} with the field. This decrease of J_{c1} allows the ac field to penetrate deeper into the weak links. The narrow peak at 80 Oe corresponds to full penetration of the ac field throughout the entire weak-link network. Above this field, the contribution of the weak-links to V_3 starts to drop while the contribution of the grains to V_3 starts to increase. The slow rise of V_3 with the field above 200 Oe reflects slow decrease of J_{cg} with the field. The broad peak around 1 kOe corresponds to penetration of the ac field throughout all the grains. Above this field, V_3 decreases at the same rate as J_{cg}.

The remanent third-harmonic response, measured after momentary application of the dc field, is generated only by the weak links because at zero dc field the small ac field cannot penetrate the grains. The grains, however, affect this remanent response indirectly; the flux trapped within the grains create an internal bias field in the weak links. As a result, the intergrain critical current decreases and consequently V_3 increases. This interpretation is consistent with the experimental observation that the remanent magnetization and the remanent third-harmonic response saturate at the same dc field.

The explanation outlined in the previous paragraph invokes a superposition of two contributions, namely from the grains and from the weak links, to the magnetic response. Such an approach was successfully applied by Muller[5] who was able to quantitatively fit the observed temperature dependence of the linear susceptibility in Y-Ba-Cu-O samples. In the following, we show that this approach may indeed reproduce the *qualitative* features of the field dependence of the nonlinear response described in Fig. 1, but a quantitative fit still awaits further improvements in the model.

Our theoretical calculations are based on the equations derived by Ji *et al.*[9] for the magnetization $M(t)$ of a slab as a function of field $H(t)=H_{dc}+H_{ac}\sin(\omega t)$, using the Bean model and a simplified Kim model for the critical current density. These equations include a *single* adjustable parameter, namely the full penetration field H_p which is proportional to the square root of the product of the pinning force and the thickness of the slab. In our special case where $H_{ac} \ll H_{dc}$, we derived a simplified form of these equations in terms of a parameter $\gamma \equiv H_p^2/(H_{ac}H_{dc})$. For $\gamma \leq 2$:

$$M_{\pm}(t)/H_{ac} = \mp \gamma/4 \pm 1 - \sin(\omega t) \mp [\sin(\omega t) \mp 1]^2/2\gamma\,, \quad \pm\sin(\omega t) > 1-\gamma\,,$$

$$M_{\pm}(t)/H_{ac} = \pm \gamma/4, \quad \pm\sin(\omega t) < 1-\gamma\,,$$

and for $\gamma > 2$:

$$M_{\pm}(t)/H_{ac} = -H_{dc}/H_{ac} + 2H_{dc}^2/H_p^2 \pm 1/\gamma - \sin(\omega t) \mp [\sin(\omega t) \mp 1]^2/\gamma\,,$$

where the plus and minus signs correspond to decreasing and increasing field, respectively. Using these equations, the third-harmonic susceptibility $|\chi_3|$ may be calculated analytically:

$$|\chi_3| = (\gamma/15\pi)(5\gamma^2 - 22\gamma + 25)^{1/2}, \quad \gamma \leq 2\,,$$

$$|\chi_3| = 4/15\pi\gamma, \quad \gamma > 2\,.$$

Calculations of $|\chi_3|$ from these equations yield the same results as the numerical calculations described by Ishida and Goldfarb.[10] If we consider the sample as an effective medium characterized by a single effective field H_p, as in Ref. 10, the calculations yield theoretical curves that cannot be fitted to the experimental data described in Fig. 1 over the entire field range. A reasonable fit could be obtained only by a superposition of two curves corresponding to a small and large H_p. The best fit, which is described by the solid line in Fig. 1, was obtained by superposition of the response of a "soft" component with $H_p = 2.3$ Oe and the response of a Gaussian distribution of "hard" components with a mean value of $H_p = 7.6$ Oe and a width of 2.4 Oe. The volume fraction of the hard component was taken as 0.67. The calculated contributions from the soft and the hard components are indicated in the lower part

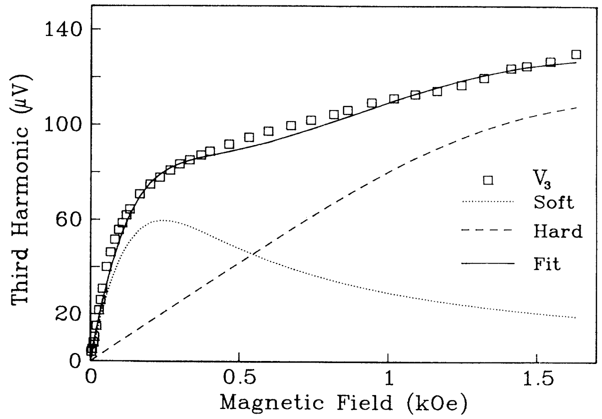

FIG. 3. Field dependence of the third-harmonic component in the ac response of Y-Ba-Cu-O sample doped with 6% Ag (sample 2). The solid line is a theoretical fit, calculated as superposition of contributions of weak links (dotted line) and grains (dashed line).

of Fig. 1 by the dotted and dashed lines, respectively. The distribution of the H_p values for the hard component is consistent with the fact that the grains in the sample are of different size and orientation. When H_p of the soft component is increased, the low-field peak due to the weak links is merged with the broad peak of the grains, as illustrated in Fig. 3. The squares in this figure describe experimental data obtained in sample 2. The solid line in Fig. 3 is a theoretical fit obtained by a superposition of the response of a soft component with $H_p = 3.5$ Oe (width 1.5 Oe) and the response of a hard component with $H_p = 10.5$ Oe (width 2.5 Oe). The contributions of the soft and hard components are illustrated in Fig. 3 by dotted and dashed lines, respectively. The fact that the soft component in this sample has a larger H_p is consistent with the better connectivity and larger J_{c1} in this sample.[15,16] The theoretical fits of Figs. 1 and 3 nicely reproduce the qualitative features of $V_3(H)$ in both samples. However, the dc data of Fig. 2 imply that full penetration is obtained at fields $\simeq 20$ Oe and $\simeq 200$ Oe for the weak links and the grains, respectively. Thus, the dc data imply larger values for the two fields. This apparent discrepancy is still not understood.

In conclusion, the experimental data clearly show that both the weak links and the grains contribute to the nonlinear magnetic response. The contribution of the weak links dominates at low fields while the contribution of the grains dominates at high fields. Consequently, the sample cannot be considered as an effective medium over the entire field range. The remanent nonlinear response is entirely dominated by the weak links. The grains affect this response indirectly; the flux trapped within the grain create an internal field in the weak links which affect the intergrain critical current and the nonlinear response.

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