Magnetization and harmonic response of $YBa_2Cu_3O_{7-\delta}$:Ag composites

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(Received 29 April 1991; revised manuscript received 20 September 1991)

The magnetic response and the behavior of the harmonic components of a bulk sample of the superconducting composite system $YBa_2Cu_3O_{7-\delta}$:$Ag_{0.1}$ are studied in the presence of a varying sinusoidal magnetic field. It is observed that as the amplitude of the field increases, the wave form of the magnetization M changes from a nearly sinusoidal, to a square-wave-like, then to a two-peak structure, consistent with the prediction of critical-state models. The crossover from the Bean to Anderson-Kim regime is found in the amplitude dependence of the third- and fifth-harmonic response at a fixed temperature. The third-harmonic response shows a peak below T_c as the sample is cooled and the peak shifts to lower temperature and broadens with increasing amplitude of the exciting field. The second-harmonic response, which is zero in the absence of a dc field, passes through a maximum as the dc field is increased. The results of these measurements are in agreement with predictions of the modified critical-state model that incorporates the field dependence of the critical current.

I. INTRODUCTION

Among the number of unusual behaviors of high-T_c superconductors, the nonlinear electromagnetic behavior at low amplitude of the exciting field is one of the interesting features. The harmonic response of type-II superconductors in the presence of a sinusoidal magnetic field was extensively studied by Bean[1] using the critical-state model. The model assumes the existence of a limiting macroscopic superconducting current density J_c that flows transverse to the local magnetic field. When the field dependence of J_c is ignored, only the odd harmonic component of the magnetic induction $B(t)$ is finite, even in the presence of a nonzero dc magnetic field.[1,2] In low-T_c superconducting materials the field dependence of the critical current can be ignored for small fields. On the contrary, a strong field dependence of J_c for a small magnetic field has been observed in bulk materials of high-T_c superconductors. The low-field nonlinear magnetic behavior is related to the existence of weak links across the superconducting grain boundaries. The magnetic field penetrates easily within these intergranular weak links. The responses of these links to the applied harmonic field are the main sources of the nonlinearities of low-field magnetic behaviors of the bulk samples. A modified critical-state model, which incorporates the field dependence of J_c, has recently been used to explain the nonlinear behavior—in particular harmonic generation in ceramic superconductors.[2–5]

Alternatively, other theoretical models like a suitably averaged collection of quantized loops containing weak links,[6] a superconductor glass,[7] and a loop model with phase slip[8] are also considered to explain the harmonic responses of high-T_c superconductors. The nonlinear magnetic response of a sintered ceramic sample $YBa_2Cu_3O_{7-\delta}$ in the presence of a small harmonic field has been investigated at length by Jeffries and coworkers.[6] Other measurements on harmonic generation in the same material have been reported by a number of researchers.[2–5,7,9–13] The harmonic response in $TlBa_2Ca_3Cu_4O_y$ has been studied experimentally and the results are interpreted in terms of the modified critical-state model.[14]

In this paper, we report the measurement of the magnetization and the harmonic component of the magnetic induction of a sintered superconductor composite, $YBa_2Cu_3O_{7-\delta}$:$Ag_{0.1}$. By measuring the field dependence of the magnetization of the sample in its superconducting state at 77 K, we have observed the crossover behavior from the Bean regime to the Anderson-Kim regime as predicted by the critical-state model.[2] The dependence of the harmonic response on the amplitude of the ac exciting field, dc biasing field, and temperature has been studied at a frequency of 1 kHz. The experimental data are found to be in good agreement with the theoretical behavior obtained from the critical-state model.[2]

II. EXPERIMENTAL

The material used in this work was a polycrystalline composite of $YBa_2Cu_3O_{7-\delta}$:$Ag_{0.1}$. The bulk sample of cylindrical geometry was placed inside a signal coil which was in series opposition with another identical coil. These coils were placed coaxially within a long solenoid, which creates the ac exciting field. A pair of coils placed over the solenoid was used to eliminate the effect of the earth magnetic field. The dc biasing field was produced by another coaxial solenoid. The magnetization measurement was performed at a frequency of 72 Hz and at varying field amplitudes from 400 mOe to 20 Oe. The signal from the signal coil was integrated by an integrator (Walker Scientific Inc., Model MF 3D) whose output was thus proportional to the magnetization of the sample.

The integrator output was measured and recorded with the help of a storage oscilloscope. For the measurement of the harmonic response a small secondary coil of thin copper wire (44 Swg) with 100 turns was wrapped directly around the middle of the sample, and the measurements were carried out at a higher frequency (1 kHz) of the exciting field. The signal from this secondary was passed through a notch filter which rejects ~ 60 dB of a 1 kHz component and was detected by an amplifier tuned at third (or fifth) harmonics. The second-harmonic component was measured with a lock-in amplifier. The sample was zero-field cooled at 77 K and then the amplitude V_{nf} of the nth harmonic component ($n=2,3$, and 5) of the signal was measured as a function of H_{ac}, the amplitude of the exciting ac field. The temperature dependence of the third-harmonic component and the dc field dependence of the second-harmonic component were also studied.

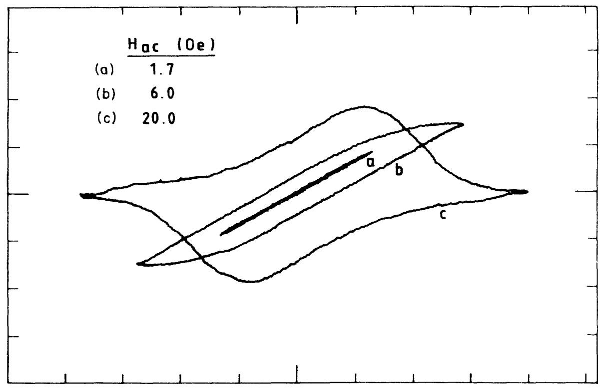

FIG. 2. Hysteresis loops recorded by the storage oscilloscope of the same sample at 77 K for three applied ac fields (1.7, 6, and 20 Oe).

III. MAGNETIZATION

The measured magnetization ($4\pi M$) of the sample for three different amplitudes of the exciting field $H_{ac}=1.7$, 6, and 20 Oe is plotted against time in Fig. 1 and the corresponding hysteresis $4\pi M$ vs H behavior is shown in Fig. 2. The exciting fields are sinusoidal in all the three cases. For a small amplitude of the excitation, $H_{ac}=1.7$ Oe, the magnetization is nearly sinusoidal [Fig. 1(a)] in nature and a small hysteresis is observed [Fig. 2(a)]. Around this field amplitude, the flux starts to penetrate the intergranular region. With the increase of H_{ac}, the magnetization deviates from the sinusoidal character and for $H_{ac}=6$ Oe, the magnetization takes a nearly square wave character [Fig. 1(b)] and a normal hysteresis loop [Fig. 2(b)] appears. We note that such a behavior of $4\pi M(t)$ is observed only within a small field interval around $H_{ac}=6$ Oe. Further distortion in the magnetization arises as H_{ac} increases. Figure 1(c) depicts the behavior of $4\pi M$ at $H_{ac}=20$ Oe. Sharp peaks appear at the points where the field is near zero and a small hump is found at the points where the amplitude of the field is maximum. The corresponding hysteresis behavior shows a two-peak structure [Fig. 2(c)]. These behaviors of $4\pi M$ vs t (or $4\pi M$ vs H) can be qualitatively understood in the light of the modified critical-state model.[2-4] The sintered sample of a high-T_c superconductor can be viewed as agglomerates of superconducting grains connected by weak links. The magnetic behavior of the bulk sample at a low magnetic field is mainly due to the penetration of the flux line into weak links and is assumed to be described by the critical-state model[2-4] with the critical current density $J_c=\alpha/(|H_i|+H_0)$, where H_i is the local magnetic field, α is the pinning force per unit volume, and H_0 is the field parameter which limits J_c in the limit $H_i \to 0$. For $H_i \ll H_0$, it reduces to the Bean model and in the other limit $H_i \gg H_0$, $J_c \sim \alpha/|H_i|$, which is the Anderson-Kim model[15]. Using $J_c=\alpha/|H_i|$, Ji *et al.*[2] derived the equations for the magnetization of a sample in the presence of the field $H=H_{dc}+H_{ac}\cos\omega t$. To analyze the present data on the magnetization, expressions for $4\pi M$ with $H_{dc}=0$ are reproduced. For a zero-field cooled sample of slab geometry,[2]

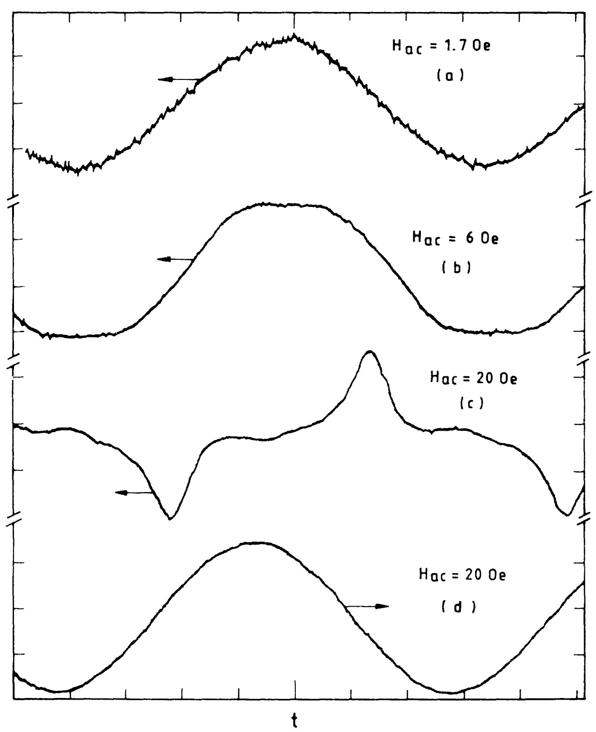

FIG. 1. Recorder plot (curves a, b, and c of the magnetization (in arbitrary units) against time for different values of the ac fields. Curve d shows a similar plot of the ac field, $H=H_{ac}\cos\omega t$ with $H_{ac}=20$ Oe. The zero-field cooled sample $YBa_2Cu_3O_{7-\delta}$:$Ag_{0.1}$ was at 77 K.

$$4\pi m = \frac{4\pi M}{H^*} = -h \pm \tfrac{2}{3} h_{ac}^3 \left\{ 2\left[\left(1 \pm \frac{h}{h_{ac}} \left| \frac{h}{h_{ac}} \right| \right) \Big/ 2 \right]^{3/2} - \left| \frac{h}{h_{ac}} \right|^3 \right\}, \quad \text{for } h_{ac} < 1 , \tag{1}$$

where $h_{ac} = H_{ac}/H^*, h = H/H^*$, and $H^* = (4\pi\alpha D)^{1/2}$, D being the width of the slab.

The plus and minus signs correspond to the decreasing and the increasing H, respectively. The minimum field H^* to penetrate the whole sample depends on the pinning force per unit volume and the width of the sample. For the other limit $h_{ac} > 1$ and for the decreasing H,

$$4\pi m = -h + \tfrac{2}{3} \left\{ 2 \left| \frac{h^2 \operatorname{sgn}(h) + h_{ac}^2}{2} \right|^{3/2} - |h|^3 - |1 - h_{ac}^2|^{3/2} \right\} ,$$

$$\text{for } h_{ac} > h > |h_{ac}^2 - 2|^{1/2} \operatorname{sgn}(h_{ac}^2 - 2) , \tag{2}$$

$$4\pi m = -h + \tfrac{2}{3} [|h^2 \operatorname{sgn}(h) + 1|^{3/2} - |h|^3 ,$$

$$\text{for } |h_{ac}^2 - 2|^{1/2} \operatorname{sgn}(h_{ac}^2 - 2) > h > -h_{ac} . \tag{3}$$

The behaviors of the reduced magnetization obtained from Eqs. (1)–(3) are shown in Figs. 3 and 4 for typical values of $h_{ac} = 2, 0.7$, and 0.1 (curves for the increasing field direction are obtained by symmetry). We note that for a low field the induced magnetization is small and the global M-H behaviors appear to be identical in the two limiting models. However, the harmonic response in the low-field regime in the two models differs which is discussed in Sec. IV.

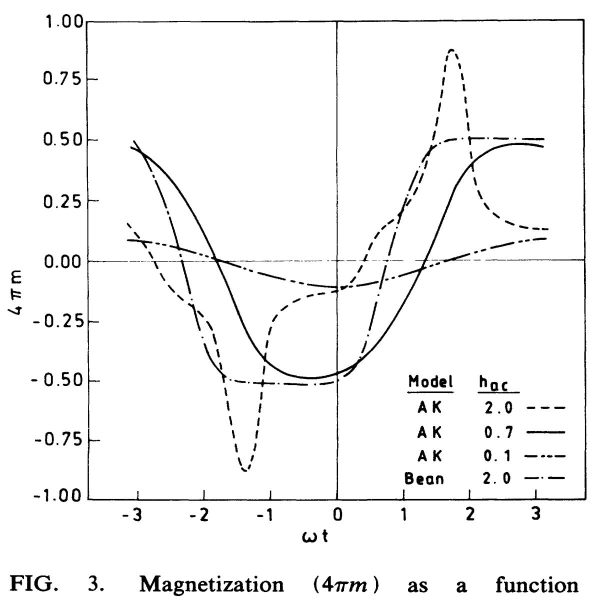

FIG. 3. Magnetization ($4\pi m$) as a function of $\omega t(-\pi < \omega t < \pi)$ predicted by the Bean and the Anderson-Kim (AK) model. $4\pi m$ and h_{ac} are normalized with H^*.

Within the low-field region the magnetization is nearly sinusoidal and the hysteresis is small. For the intermediate field $H_{ac} = 0.7H^*$, the magnetization, as obtained from Eq. (1), tends towards a square-like character, which is closer to the characteristic behavior of M in the Bean model.[2] The magnetization is nearly flat and close to the maximum of the ac field; it exhibits a normal hysteresis (Fig. 4). For a higher field $H_{ac} = 2H^*$, two peaks appear in the Anderson-Kim model, close to the region where the field sweeps through zero. The two-peak structure is also reflected in the hysteresis as shown in Fig. 4. The parameter H^* for this sample was estimated to be around 10 Oe from another set of measurements[16] and using the Bean model. So the calculated $4\pi M$-t (or, H) behaviors correspond to the experimental conditions. It is evident that the theoretical results (Figs. 3 and 4) are in qualitative agreement with the experimental ones (Figs. 1 and 2). For comparison, the magnetization for the field $H_{ac} = 2H^*$ as obtained from the Bean model[2] is also given in Fig. 3 (dash-dot line). The response $4\pi M(t)$ is square wave in nature. Therefore the experimental results clearly demonstrate the validity of the Anderson-Kim model for the excitation $H_{ac} > H^*$. Although all the theoretical curves are for the sample with a slab geometry, the results will differ quantitatively and not qualitatively for cylindrical geometry as noted by Ji *et al.*[2]

IV. FIELD DEPENDENCE OF THE THIRD- AND THE FIFTH-HARMONIC RESPONSE

The amplitude dependence of the third-harmonic signal V_{3f} and the fifth-harmonic signal V_{5f} is shown as a function of H_{ac} from 400 mOe to 20 Oe in Fig. 5. The upper limit of H_{ac} was set up by the power from an audio

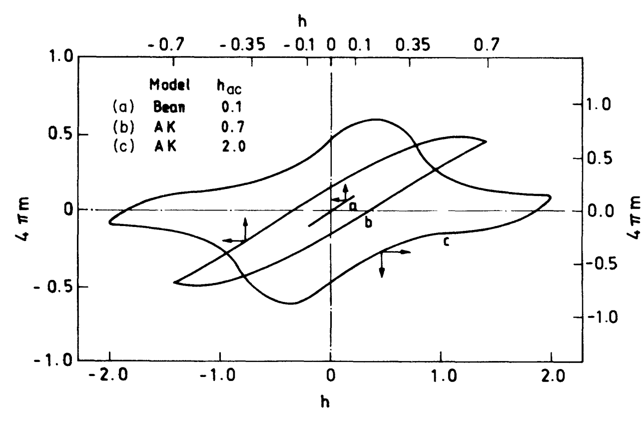

FIG. 4. Hysteresis loops as predicted by the Bean and the Anderson-Kim models. $4\pi m$ and h_{ac} are normalized with H^*.

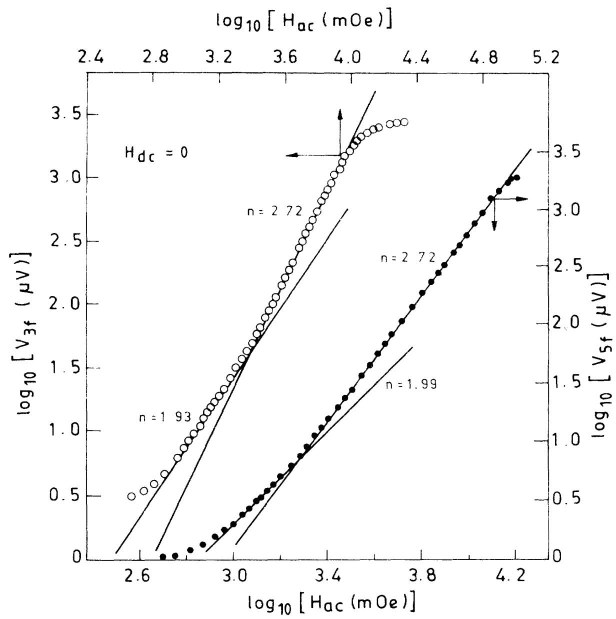

FIG. 5. Log-log plot of the amplitude of the third- and the fifth- harmonic component of the measured signal at 77 K with H_{ac}. The sample was zero-field cooled. The numbers are the values of the slope (n) in the lower- and the higher-field region.

power amplifier and the lower limit by the noise in the detection system. The solid lines are least-squares fitted lines. For fields $H_{ac} < 2.5$ Oe the data are well described by the relation $V_{3f} \propto H_{ac}^n$, $n = 1.93$, which is close to the prediction of the Bean model ($n = 2$). At higher fields, $n = 2.72$, which is close to a cubic dependence on H_{ac} corresponding to the Anderson-Kim model. For a field above 12 Oe, the dependence on H_{ac} becomes weaker. A similar behavior is exhibited by V_{5f} also. Thus a crossover from the Bean regime to the Anderson-Kim-like regime is observed for both the third- and the fifth-harmonic response.

This kind of behavior of V_{3f} was observed earlier in bulk samples of $YBa_2Cu_3O_{7-\delta}$ (Ref. 2) and $TlBa_2Ca_3Cu_4O_y$ (Ref. 14). The crossover from $n = 2$ to $n = 3$ occurs at a field $H_{ac} \simeq H_0$, the field parameter[14] which limits J_c. So for $H_{ac} \ll H_0$ the critical current J_c is essentially independent of the internal field and therefore the electromagnetic response can be described by the Bean model. For this sample, H_0 comes out to be nearly 3 Oe. We note that the situation with the lowest field $H_{ac} = 1.7$ Oe $< H_0$ corresponds to the Bean regime whereas the other two situations ($H_{ac} = 6$ Oe and 20 Oe) lie in the Anderson-Kim regime.

V. TEMPERATURE DEPENDENCE OF THE THIRD-HARMONIC AMPLITUDE

We measured the temperature variation of the third-harmonic signal from the temperature above the transition temperature up to 77 K which keeping the ac field amplitude constant and $H_{dc} = 0$. The third-harmonic response function, which is proportional to V_{3f}/H_{ac}^n (where $n = 1.93$ and 2.72 for a low and a high field, respectively), is plotted against temperature in Fig. 6.

For a low amplitude of H_{ac}, a very steep increase in the response function is observed and a peak appears below T_c. The response falls off less sharply as the sample is cooled below the peak temperature. When the amplitude of H_{ac} increases, the peak height of the response decreases and the peak appears at a lower temperature. For a large field amplitude (the Anderson-Kim limit) the peak is broadened. This feature can be understood within the general framework of the critical-state model. In this the temperature dependence of the third-harmonic signal is solely determined by H^*, which increases monotonically as the sample is cooled below T_c. The critical current J_c is also proportional to H^*.

Within the Bean model (small H_{ac}), the third-harmonic response is proportional to H^* and H^{*-1} when $H^* < H_{ac}$ and $H^* > H_{ac}$,[2] respectively. When T is close to T_c, J_c is small and hence the third-harmonic response ($V_{3f}/H_{ac}^{1.93}$) increases linearly with H^* as the temperature passes through T_c. When $T < T_c$, H^* becomes larger than H_{ac} and hence the third-harmonic response is small ($\sim 1/H^*$). The combination of these behaviors produces a peak below T_c. Below the peak temperature the third-harmonic response for the two lower fields coincides and is consistent with the field dependence of the signal in the Bean model. Similarly the response at a higher field at low temperatures follows the results of the Anderson-Kim model. Near the peak temperature the field dependence of the signal is complicated due to the crossover from one regime ($H^* < H_{ac}$) to another ($H^* > H_{ac}$).

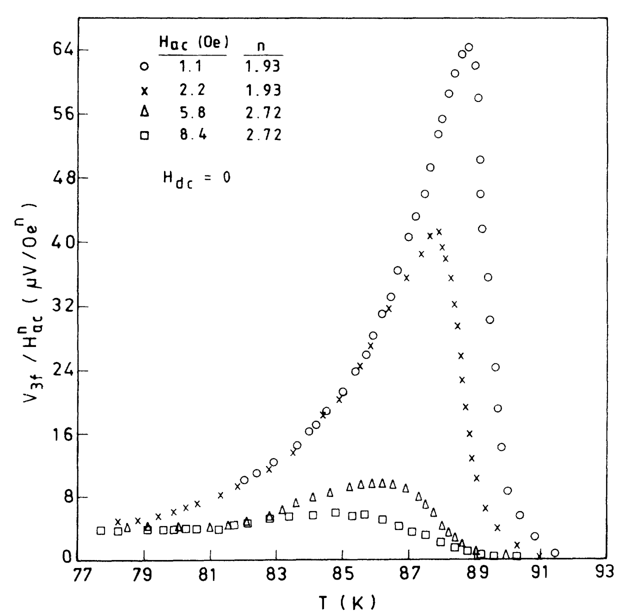

FIG. 6. Temperature variation of the third-harmonic response function $V_{3f}/(H_{ac})^n$ of the sample. The values of n are taken from Fig. 5.

VI. HARMONIC AMPLITUDE IN THE PRESENCE OF A dc FIELD

The results of the second-harmonic response V_{2f} measured at 77 K are presented in Fig. 7. Three different values of H_{ac}:1,9, and 15 Oe, are given for the three regions of the $4\pi M$ vs t behavior (Fig. 7). The second-harmonic signal is zero in the absence of any dc field for all amplitudes of the ac field. The signal increases very sharply for all the three values of H_{ac} when H_{dc} is small. As H_{dc} increases the signal passes through a peak which gets sharper with increasing H_{ac}. For $H_{ac}=15$ Oe, the peak is closer to a kink. For a high dc field the signal decreases but the relative decrease is slow for the small H_{ac}. The data for $H_{ac}=15$ Oe closely resemble the theoretical result for $H_{ac} \gg H^*$ in the Anderson-Kim model.[2]

These behaviors of the second-harmonic response can be qualitatively understood considering the $4\pi M$-H behavior. The hysteresis loop is symmetrical about $H_{dc}=0$ and the even harmonic response is zero due to this symmetry. As the finite H_{dc} breaks this symmetry, the even harmonic component appears in the presence of a dc field. The amplitude of even harmonic depends on the degree of asymmetry produced by H_{dc} and hence it increases with H_{dc}. When H_{dc} is large, J_c is predominantly determined by H_{dc} and thus within the entire subloop of magnetic induction, J_c is approximately constant. Within this field region the Bean-model results are expected and hence the even harmonic component disappears. The response is symmetrical about $H_{dc}=0$ when the measurement is taken, starting from a demagnetized state. However, a large hysteresis is observed when the field is reversed from a maximum applied field. This will be reported elsewhere.

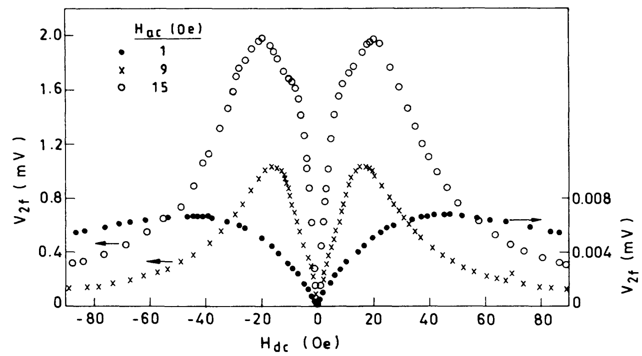

FIG. 7. dc field dependence of the second-harmonic signal amplitude (V_{2f}) taken for the three different values of the ac exciting fields (1, 9, and 15 Oe) at 77 K. The sample was zero-field cooled.

VII. CONCLUSIONS

We have studied the magnetization and its harmonic response of a sintered superconducting composite, $YBa_cCu_3O_{7-\delta}$:$Ag_{0.1}$, within the excitation field regime where intergranular characteristics are predominant in determining the nonlinear response of the system. Experimental results on magnetization and hysteresis loops have been compared with the theoretical ones derived from the critical-state model.[2] The theoretical curves are in good agreement with time- or field-dependent features of the magnetization. The experimental results demonstrate the transition of the field dependence of the intergranular critical current from H_i^0 (Bean's regime) to the $|H_i|^{-1}$ dependence (Anderson-Kim regime). The field and temperature dependence of the harmonic response is also in accord with the prediction of the critical-state model. These results further strengthen the view that intergranular superconductivity has all the features of a type-II superconductor with reduced critical current and critical field.

ACKNOWLEDGMENTS

The authors acknowledge the financial support by the Department of Science and Technology, Government of India and the partial support for D. S. by the Council of Scientific and Industrial Research, Government of India. We are thankful to K. L. Chopra and D. Bhattacharya for their interest in this work. The technical assistance of S. Mazumder is gratefully acknowledged.

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