Journal of Applied Physics

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Citation: J. Appl. Phys. 69, 6738 (1991); doi: 10.1063/1.348901
View online: http://dx.doi.org/10.1063/1.348901
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v69/i9
Published by the American Institute of Physics.

Characterization of Y-Ba-Cu-O thin films using their nonlinear magnetic response

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(Received 17 December 1990; accepted for publication 22 January 1991)

Measurement of the third harmonic component in the ac magnetic response was used to investigate the multiphase nature of Y-Ba-Cu-O thin films grown epitaxially on single-crystal wafers of $SrTiO_3$. Films prepared by electron beam evaporation and by activated reactive evaporation techniques exhibit several peaks in the harmonic amplitude versus temperature, indicating multiple phase transitions. The film prepared by laser ablation technique exhibits a sharp single peak, indicating a single phase transition. These results, corroborated by x-ray diffraction and x-ray photoelectron spectroscopy, demonstrate the usefulness of third harmonic measurements in characterizing thin-film samples.

High-temperature superconductors (HTSC) in the form of thin films have attracted special attention because of their potential applications in electronic devices. A wide variety of techniques have been successfully employed in fabricating HTSC thin films, e.g., plasma spray, sputter deposition, electron-beam evaporation, activated reactive evaporation, and laser ablation. The quality of the films is commonly tested by measuring resistivity, ac susceptibility, and critical current density versus temperature. However, these techniques are not sensitive to the homogeneity of the thin film. In this communication we describe a novel magnetic technique for characterizing HTSC thin films. This technique has the advantage of the existing magnetic technique, i.e., it does not require contacts, and in addition it can provide clear information about the presence of a number of superconducting transitions in a given sample. Moreover, we propose this technique as a method for local measurement of superconducting properties in thin films, allowing the detection and mapping of inhomogeneities in these properties. Critical properties such as the transition temperature, T_c, and the critical current density, J_c, may vary across a film as a result of variations in composition and stoichiometry. It is important to detect such variations in the fabrication process of the films into electronic devices.

The method is based on measuring harmonic components in the response of the material to sinusoidal magnetic fields. The global response of the material is measured using the standard ac susceptibility system.[1] This system consists of a primary coil coaxial with a pair of balanced coils, one containing the sample. The material's response is monitored by measuring the power spectrum of the off-balance voltage induced in the coil pair. Our measurements were performed with an ac field of amplitude 50 mG and frequency 20 kHz applied perpendicular to the surface of the films.

Harmonic generation in the ac magnetic response of HTSC has been the topic of several theoretical and experimental studies.[2–6] A recent model by Ji *et al.*[2] extends the Bean critical state model and attributes the harmonic generation to the hysteretic, nonlinear relationship between the magnetization and the external field due to flux pinning. The magnetization equations derived by Ji *et al.* include a single adjustable parameter, namely the full penetration field H_p which is proportional to the square root of the pinning force α. Thus, the temperature dependence of the harmonic components comes from that of α. The solid curve in Fig. 1 shows the prediction of this model for the third harmonic response V_3 of a single phase sample in a form of a slab assuming $\alpha \propto [1 - (T/T_c)^2]^2$ and $T_c = 90$ K.[7] In this case, V_3 exhibits a single peak near T_c. The dashed curve in Fig. 1 shows the third harmonic response when a second superconducting phase, with a lower T_c, is added to the system. In the calculation of this curve we assumed that the volume fractions of the two phases are equal and that the low-T_c phase is not screened by the high-T_c phase. In this case two peaks are obtained near the transition points of the two phases. Thus, each phase reveals itself by a peak in V_3 near its transition temperature. It is important to note that resistivity measurements in such a system often exhibit a single transition corre-

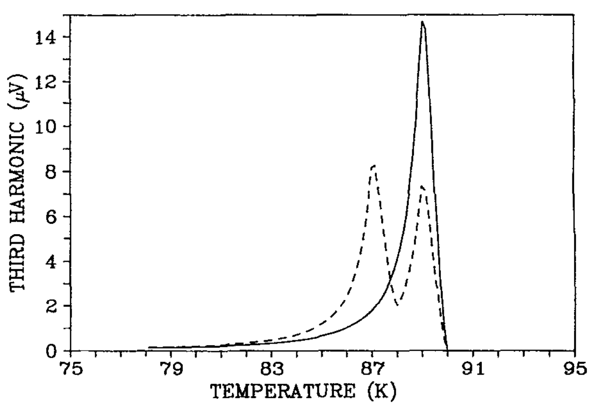

FIG. 1. Calculated third harmonic susceptibility in single phase (solid line) and two-phase (dashed line) superconducting samples.

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0021-8979/91/096738-03$03.00

sponding to the high-T_c phase. The low-T_c phase reveals itself only if it is not shorted by the high-T_c phase. It should also be noted that the behavior of the out-of-phase component χ'' of the linear susceptibility is similar to that of V_3. However, the measurement of χ'' involves a phase adjustment procedure which is not always straightforward.

Thin film samples of Y-Ba-Cu-O, prepared by various techniques, were obtained from different sources. The films studied were all deposited upon polished (100) $SrTiO_3$ single-crystal wafers. One method employed electron beam evaporation of Cu, Y, and BaF_2 source materials.[8] Oxygen was directed at the rotating sample during deposition. The sample was annealed outside the deposition chamber, in flowing oxygen, including a period of exposure to moist oxygen. The 2500 Å film (sample A) was confirmed to be epitaxial by Rutherford backscattering. The second thin-film sample (sample B), a 1200 Å single crystal with its ⟨100⟩ axis normal to the (100) surface of the $SrTiO_3$, was grown by activated reactive evaporation.[9] Y and Ba metals were individually evaporated, onto a heated substrate, using electron guns, and Cu metal from an alumina crucible heated by a tungsten wire. Their rates were adjusted to provide the proper stoichiometry. A rf generated oxygen plasma was maintained between the evaporation sources and the substrate by locally introducing oxygen. The crystalline nature of the film was determined by x-ray diffraction. The third sample (sample C) was grown by the laser ablation technique.[10] Frequency doubled Nd:YAG laser pulses of 1.7 J/cm^2 and 10 ns duration were used to deposit the film on a $SrTiO_3$ substrate held at 725 $\pm$ 5 °C in 0.2 Torr of O_2 ambient. X-ray diffraction patterns showed that the 3600 Å film grew epitaxially with the c-axis perpendicular to the substrate.

The upper curve of Fig. 2 shows the temperature dependence of the ac magnetic susceptibility in the thin-film sample A. These data indicate a superconducting transition at about 91 K, consistent with the results of resistivity measurements. However, a close examination of the magnetic susceptibility data indicates traces of additional transitions around 79 and 87 K. The power of the third-harmonic technique in detecting these transitions is clearly demonstrated in the lower curve of Fig. 2, where the various transitions produce large and sharp peaks in the response signal. The multiphase nature of this sample has been confirmed by x-ray diffraction[11] and x-ray photoelectron spectroscopy.[12] Measurements of the third harmonic response in sample B (Fig. 3) indicate two transition temperatures around 81 and 82.5 K. This corroborates the multiphase nature of the sample determined by XPS.[12] Sample C, prepared by the laser ablation technique, exhibits a single peak in the measurements of the third harmonic signal versus temperature, as illustrated in Fig. 4. This sharp peak indicates a single phase transition near 91 K. The various phases in samples A and B are probably Y-Ba-Cu-O phases with different oxygen content. A similar staircaselike structure in the oxygen content has been recently reported for Y-Ba-Cu-O single crystals.[13]

The magnetic measurements described above can be performed locally on the surface of a thin-film sample.[14] For local measurement of the response, a combined read/write magnetic head is held in close proximity to the surface of the film. A sinusoidal field is produced at the gap of the write head, and as a result a voltage is induced across the gap of the read head. The spectrum of this voltage is measured while the read/write head is scanned across the surface of the film. If one wishes only to map superconducting and nonsuperconducting regions, it is not necessary to analyze the harmonic signals. In this case the phase relationship between the input and the output signals at the driving frequency could be used. In superconducting regions the output fundamental signal will be 180° out of phase with the field input signal. In nonsuperconducting regions the output fundamental signal will be small and in phase with the input. Measurement of the harmonic signals permits the mapping of

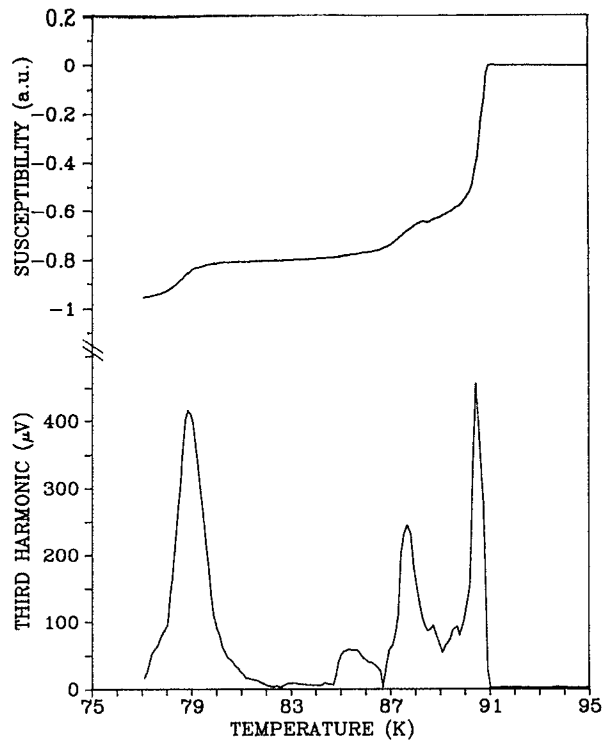

FIG. 2. Measured linear (upper curve) and third harmonic (lower curve) susceptibilities in Y-Ba-Cu-O thin-film sample prepared by electron-beam evaporation technique.

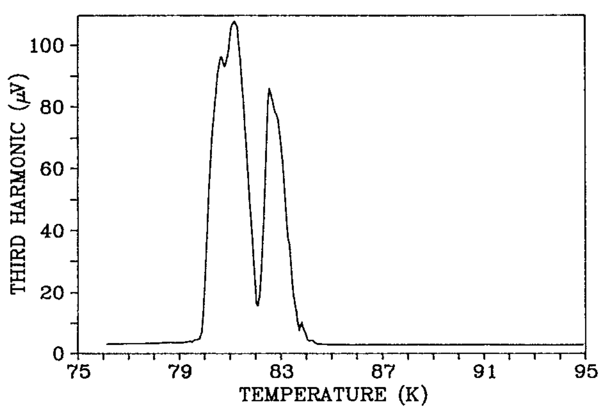

FIG. 3. Measured third harmonic susceptibility in Y-Ba-Cu-O thin-film sample prepared by activated reactive evaporation method.

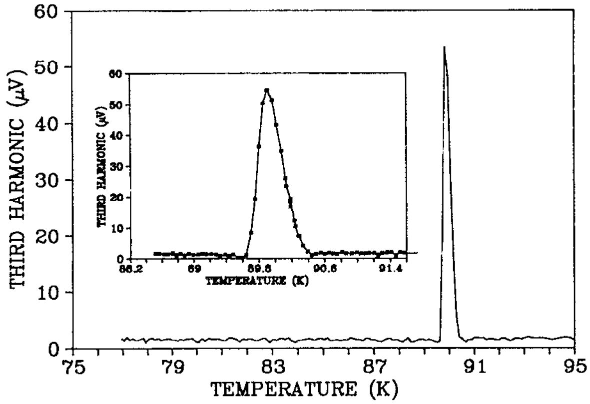

FIG. 4. Measured third harmonic susceptibility in Y-Ba-Cu-O thin-film sample prepared by laser ablation technique.

other critical properties such as the critical current density. This can be accomplished by measuring the amplitude V_3 of the third harmonic signal. According to the Bean Model, for small ac fields, V_3 is inversely proportional to the critical current density. Accordingly, a mapping of the inverse of the third harmonic amplitude indicates the variations of the critical current density across the sample. Preliminary experiments using a magnetic head of a computer disk drive have indicated the feasiblity of this concept.[14,15]

In summary, measurement of harmonic components in the ac magnetic response of HTSC films provides a sensitive and nondestructive tool for the characterization of these films. This measurement provides clear information about the presence of a number of superconducting transitions in a given sample. Moreover, this technique offers a principle for local characterization of thin films. A system based on this principle can detect and map inhomogeneities in superconducting properties across the surface of a film resulting, for example, from variations in composition or stoichiometry.

The authors thank Dr. B. Dam, Dr. T. Terashima, and Dr. G. Koren for providing the thin-film samples. Thanks are also extended to Dr. J. Ladell and Dr. M. H. Frommer for sharing the results of their XRD and XPS measurements on the samples, and to Professor Y. Yeshurun for many helpful discussions. The work at Bar-Ilan was partially supported by the National Council for Research and Development through Grant No. 3074-1-89.

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