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Harmonic generation studies in laser ablated YBCO thin film grown on ⟨100⟩ MgO

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Abstract. The generation of harmonics in a laser ablated YBCO film deposited on a ⟨100⟩ MgO substrate is reported. Higher odd harmonics appeared when the film was subjected to an ac field. The presence of a dc field induces only the second harmonic with a small value of slope of $V_2 - H_{dc}$ curve ($\delta V_2/\delta H_{dc}$) compared to bulk YBCO. The variation of the amplitude of third harmonic (V_3) with H_{ac} and temperature was studied. These results are explained in terms of a critical state model. The observation of only a small amplitude of second harmonic (V_2) with a small $\delta V_2/\delta H_{dc}$ is explained in terms of a special kind of clean grain boundary present in YBCO laser ablated films on ⟨100⟩ MgO.

Keywords. Harmonic generation; thin film YBCO; laser ablation.

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1. Introduction

Magnetic harmonic generation in bulk high-T_c superconductors has been observed by many workers [1–10]. A small amount of this material, when subjected to an ac field ($H_{ac} > H_{cl}$) of frequency f, generates odd harmonics $(2n+1)f$. The addition of a dc field induces the generation of even harmonics. The amplitude of both families of harmonics are found to modulate with the variation of dc field. Even harmonics are odd functions of dc field whereas odd harmonics are even functions of dc field. Grain boundaries in bulk YBCO acting as pinning sites are responsible for the generation of harmonics.

Study of harmonic generation in films would be useful for understanding the quality and nature of grain boundaries. However, although there have been many reports on harmonic generation in bulk YBCO only a few are available on YBCO thin film [11–13]. Yamamoto *et al* [11] reported the observation of third harmonic in YBCO film prepared by evaporation technique. Shaulov *et al* [12] showed the use of third harmonic to investigate the multiphase nature of YBCO thin films and Revenaz and Dumas [13] reported the harmonic generation in YBCO thin films due to magnetically modulated microwave absorption.

In this paper, we report studies of harmonic generation in a laser ablated YBCO film on ⟨100⟩ MgO with and without a dc field. Variation of the third harmonic with ac field and temperature has also been studied.

Table 1. Relative levels of various observed higher harmonics in the YBCO film [$H_{ac} = 8{\cdot}3$ Oe $p^k - p^k$, $f = 10$ kHz, $H_{dc} = 10$ Oe]

Harmonics	Amplitudes
1	10 mv
2	3 μv
3	31 μv
4	0
5	10 μv
6	0
7	4 μv
8	0
9	2 μv

2. Experimental

Epitaxial YBCO films were deposited *in situ* on (100) MgO substrates by a laser ablation technique using the third harmonic (355 nm) of a Nd-YAG laser. Details of the deposition techniques are described elsewhere [14]. The films were highly oriented with the c-axis perpendicular to the substrates and had $J_c > 10^5$ A/cm^2 at 77 K. The film used in the present study had $T_c(R = 0) = 80$ K and thickness ≈ 200 nm.

To study the harmonic response two flat spiral copper coil of ten turns were used. The primary coil was glued on the back side of MgO substrate while the pick up coil was glued on top of the film. This configuration measures shielding. The field was applied perpendicular to the surface. An HP synthesizer (HP 3325A) was used to apply the ac signal and a dynamic signal analyzer (HP 3561 A) was used to observe the frequency spectrum of the signal in the pick up coil. A lock-in amplifier was used to record the variation of the second harmonic with dc field. A variable temperature cryostat was used in the study of the temperature variation of the third harmonic.

3. Results and discussion

Table 1 shows for a typical film the amplitudes of various harmonics induced in the detection coil, as recorded by the dynamic signal analyzer when both ac and dc field were present ($H_{ac} = 8{\cdot}3$ Oe, $f = 10$ kHz, $H_{dc} = 10$ Oe). With bulk YBCO the second harmonic appeared only when a dc field was present is a point to be noted.

In bulk YBCO the amplitude of even and odd harmonics had been found to modulate with the variation of H_{dc} showing maxima and minima [5, 7]. In these experiments on YBCO films different results were found: (i) Application of a dc field cause the appearance of the second but no higher even harmonics, and (ii) the variation of H_{dc} results in a very small modulation of the harmonics.

Figure 1 shows the variation of the amplitude of second harmonic (V_2) with H_{dc} for three different values of H_{ac} as recorded using the lock-in amplifier. This shows that slope of $V_2 - H_{dc}$ curve ($\delta V_2/\delta H_{dc}$) increases as H_{ac} is increased. This behaviour is the same as was observed for bulk YBCO [5]. However, $\delta V_2/\delta H_{dc}$ is two orders of

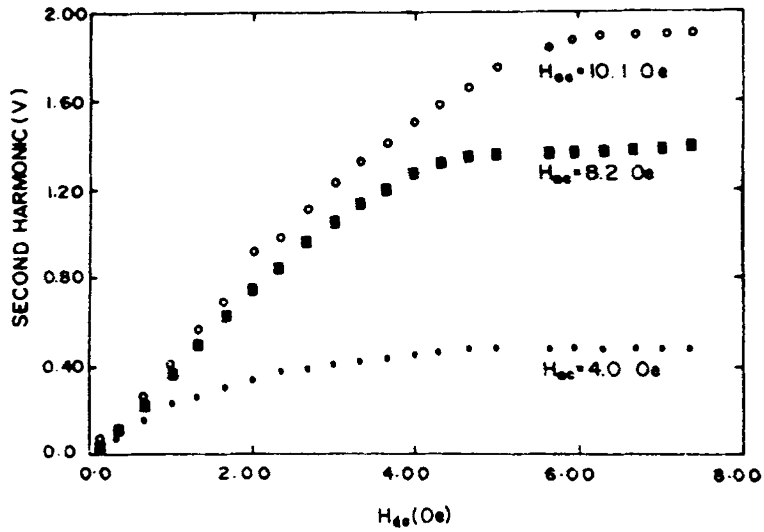

Figure 1. Variation of second harmonic with dc field at three different ac fields.

magnitude smaller than the bulk YBCO. The application of dc field also causes the variation in the third harmonic amplitude. The change in the amplitude of third harmonic when dc field is changed from 0 to 10 Oe is only 6% of the value at zero dc field. These changes in the third harmonic in film are much smaller than what has been observed in bulk YBCO.

The observed features of higher harmonics generation and very small change in harmonics amplitude due to application of dc field for the YBCO films can be understood in terms of a critical state model in which grain boundary weaklinks provide the pinning as flux sweeps in and out of the material, between the grains. The Kim model for the critical state, which assumes the dependence of J_c on local field explains the appearance of even harmonics on application of a dc field. However, if J_c is independent of the local field (Bean model) then even harmonics will not appear. Since the appearance of higher harmonic is a bulk effect, the amplitude will be proportional to the number of pinning sites. In laser ablated YBCO films on (100) MgO substrates, pinning sites are clean low angle grain boundaries. Such films have a high degree of crystallographic orientation, but in addition possesses a larger number of grain boundaries due to the lattice mismatch between YBCO and MgO basal planes [15]. These grain boundaries had been observed to have only some specific low angle orientations and appeared to be clean, (absence of any secondary phases at the interfaces) unlike the grain boundary usually found in bulk YBCO [15, 16]. It is therefore reasonable to assume that for most of the grain boundaries J_c will be independent of local field and only for a few grain boundaries J_c will depend on the local field. In such a case the application of a dc field will result in second harmonic with small $\delta V_2/\delta H_{dc}$ as observed experimentally. J_c for the film was 10^5 A/cm^2 which was about two order higher than bulk YBCO. The observed two order of magnitude lower value of $\delta V_2/\delta H_{dc}$ in film compare to that of bulk is due to larger J_c in the film.

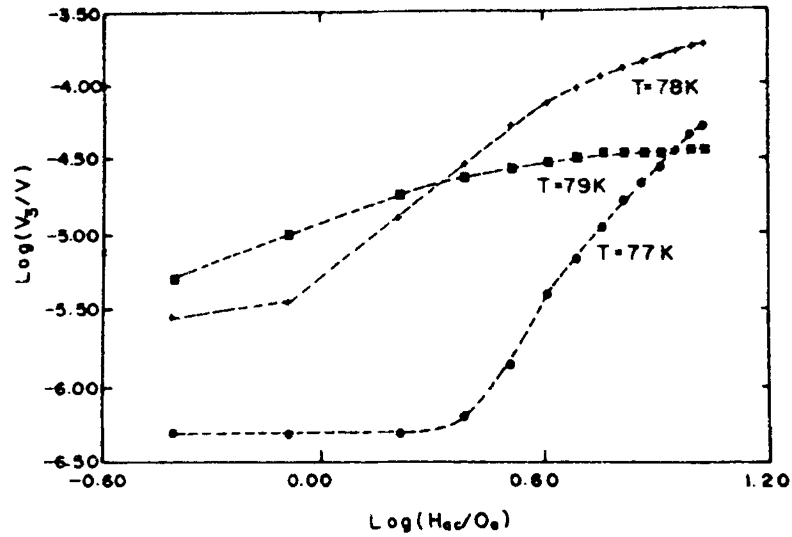

Figure 2. Variation of third harmonic with ac field ($H_{dc} = 0$).

Figure 2 shows the variations of the third harmonic with ac drive field ($H_{dc} = 0$) at three temperatures. At 77 K, V_3 was initially negligibly small, and for $H_{ac} > 2{\cdot}5$ Oe, it increased with no sign of saturation up to 11 Oe. In this region $V_3 \propto (H_{ac})^3$. At 78·2 K no substantial growth of V_3 was observed up to 0·82 Oe. At higher H_{ac} we found $V_3 \propto (H_{ac})^{1{\cdot}9}$ with a sign of saturation at 7 Oe. At 79 K growth of third harmonic was observed even from H_{ac} as small as 0·39 Oe and saturation was reached at $H_{ac} = 3{\cdot}5$ Oe.

These observations can also be understood in the framework of Bean's critical state model. The third harmonic will appear only when flux starts penetrating into the film. With the increase of temperature, J_c and H_{c1} decreases. Thus, the value of H_{ac} where the third harmonic starts growing will shift to lower values with the increase in temperature. The increase of H_{ac} increases penetration and so V_3 increases to a point of saturation when ac field approaches to the saturation field (H^*), which is given as [10]

$$H^* = KJ_c a \tag{1}$$

where K is a geometric constant, J_c is the critical current and a is the dimension perpendicular to the field. The saturation field depends on J_c, thus with the increase of temperature the saturation of the third harmonic is expected to occur at lower value of H_{ac}.

Figure 3 shows the variation of the third harmonic with temperature ($H_{ac} = 8{\cdot}3$ Oe, $f = 10$ kHz, $H_{dc} = 0$). At lower temperatures ($T < 74$ K) no harmonic generation was observed. Third harmonic appeared only after 75 K and then it continued to grow up to 78 K, after which it began to decay rapidly. At low temperatures, H_{c1} is large so that no penetration of flux occurs and therefore the third harmonic does not appear. The increase of temperature started penetration of flux in the film through the grain

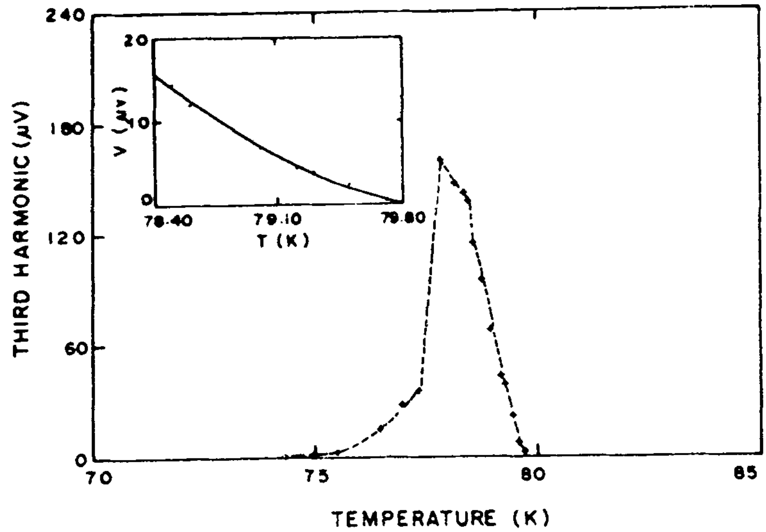

Figure 3. Variation of third harmonic with temperature ($H_{ac} = 8{\cdot}3$ Oe, $f = 10$ kHz, $H_{dc} = 0$). Inset shows fitting of decrease of V_3 to $(1 - T/T_c)^{3/2}$ near T_c.

boundaries and V_3 started growing and became maximum at the temperature when the flux fully penetrated the film. As the flux started penetrating into the film the pickup coil received signals from the applied field coil and also due to the circulating super currents on the film flowing through the various grain boundaries. The amplitude of V_3 is proportional to the pinning potential at the grain boundaries. As the temperature reaches near T_c, the pinning potential which is proportional to J_c decrease very fast. We have found that near T_c the variation of V_3 follows $(1 - T/T_c)^{3/2}$. For the YBCO laser ablated film, the decrease of J_c with T near T_c has been found [17] to follow $(1 - T/T_c)^{3/2}$. This supports the view that V_3 is proportional to the pinning potential and hence J_c of the film.

4. Conclusion

The generation of higher harmonics was observed in YBCO films deposited by laser ablation on ⟨100⟩ MgO substrate. Grain boundaries originating due to the lattice mismatch between YBCO and ⟨100⟩ MgO are responsible for harmonic generation.The presence of a dc field caused the appearance of small magnitude of second harmonic with a value of $\delta V_2/\delta H_{dc}$ which is small compared to what has been seen in bulk YBCO. The change in V_3 with dc field has also been found to be very small compared to the bulk YBCO. These features are due to the special nature of grain boundaries present in the YBCO films. These grain boundaries are clean with high J_c and very little dependence of J_c on H(in the probed range of 0 to 10 Oe). It has been demonstrated that the harmonic generation studies can give information about the quality of the grain boundaries in high-T_c superconductors and can be used as a non-contact technique for characterizing high-T_c thin films.

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