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Comparative study of the irreversibility line and of harmonic generation in field modulated microwave absorption on $YBa_2Cu_3O_7$ thin films

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Abstract

Magnetic properties and field modulated microwave absorption have been studied on epitaxial $YBa_2Cu_3O_7$ thin films prepared by MOCVD. Possible correlations between the irreversibility line and the properties of harmonics of the modulation field frequency in the reflected microwave power are discussed.

1. Introduction

The irreversible and non-linear properties of $YBa_2Cu_3O_7$ are of interest both for practical applications and for the study of their fundamental properties. Magnetic measurements and field modulated microwave absorption (1,2) should give complementary information on flux pinning mechanism on one hand and on flux dissipation on the other hand. The irreversibility line (3), which corresponds to the vanishing of the critical current below T_c in fields H_{irr} much smaller than the higher critical field H_{c2} , can be obtained from high field hysteresis loops at different temperatures (4,10,11).
Field modulated microwave absorption technique provides information on the influence of a magnetic field on microwave losses and on the vortex dynamics. In this method , the microwave power reflected from an EPR cavity loaded with the sample under study is proportional to its surface resistance . The reflected power is modulated by the application of a magnetic ac field $h = h_{ac} \sin\omega t$ and phase sensitively detected at the modulation frequency ω (2). Despite the fact that the dominant response is expected to be at twice the excitation frequency in zero dc field, since the surface resistance depends only on the magnitude of the magnetic field and not on its sign, most experimental studies deal with the signal detected at the excitation frequency. The response at 2ω has received little attention (5,6).
We report in this paper magnetic properties and field modulated microwave absorption studies as well as harmonic generation effects in the low frequency spectrum of the reflected microwave power on c-axis oriented epitaxial $YBa_2Cu_3O_7$ thin films prepared by MOCVD.
Possible correlations between the irreversibility line and properties of the harmonics of the modulation field frequency in the reflected microwave signal are discussed.

2. Experimental techniques

Epitaxial films of $YBa_2Cu_3O_7$have been prepared by MOCVD on single crystal (012) $LaAlO_3$ substrates , with thickness of 750Å for film A and 1300Å for film B. Typical size is $4x4mm^2$.The films are synthetized in a hot wall reactor with a total pressure of 6.66hPa and at temperature of about 760°C. In these conditions, the growth rate is of the order of 2.8 nm/mn. They have been characterized by x-ray diffraction and ac susceptibility. T_c was found to be 87K from ac susceptibility measurements.CuO precipitates are found on the film B surface, as observed by optical microscopy in contrast with film A.
The magnetic properties have been studied by means of a home-built high sensitivity (10^{-6} emu) vibrating sample magnetometer in fields ranging from 0.1mT to 6T. The magnetic field was perpendicular to the film plane. The microwave measurements have been performed by means of a 9.4 GHz Bruker ESP 300 spectrometer equipped with a rectangular TE_{102} cavity and continuous helium gas flow Oxford cryostat. The microwave magnetic field h_1 was parallel to the surface of the film placed at the center of the cavity where h_1 has a maximum. h_1 was perpendicular to the externaly applied colinear dc and ac fields. Zero field cooling was obtained by appropriate shielding with a μ-metal box; the residual field is of the order of 0.2 Oe. The Fourier analysis of the direct signal detected at the output of the microwave bridge was performed using a HP 70 000 spectrum analyzer.

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3. Experimental results

The low frequency Fourier analysis of the microwave power reflected by the cavity reveals generation of high order harmonics of the modulation field fundamental frequency, as shown in Fig. 1. Just below T_c, the fundamental is observed together with even harmonics of strong amplitude . These results are similar to that found on laser ablated epitaxial thin films (8). However , the temperature interval where these signals are found is larger in the present study. One should note that in all cases no modulated signal is found when the magnetic field is parallel to the plane of the film, i.e. perpendicular to the c-axis. In the results reported here, the angle between the modulation field and the plane of the film was 20 degrees.

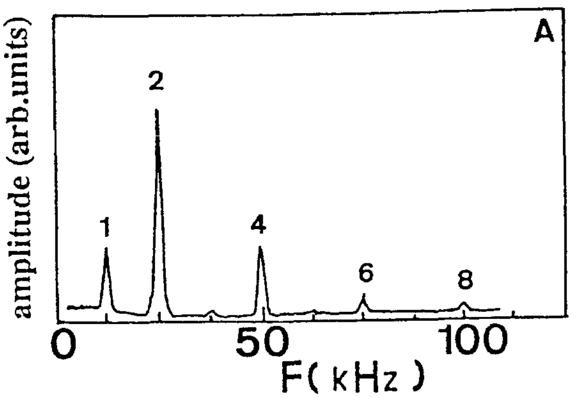

Fig.1. Typical Fourier spectrum of the field modulated microwave absorption showing the onset of harmonics below T_c. Modulation field amplitude 4 Oe; microwave power 100μW; excitation frequency $\omega/2\pi$ =12.5kHz. Film A.

One can define a parameter associated to the even harmonics $S = \sqrt{\Sigma|c2n|^2}$ where c2n are the individual amplitudes.

Fig. 2a shows the reflected power, the response at the frequency ω (L) and the even harmonics response (S), measured simultaneaously as a function of temperature for film A. Above T_c, only a base line is detected in the ω– signal. At T_c, this signal shows a sharp rise, marked also by a rapid change in the temperature dependence of the power reflected by the film placed in the cavity. At lower temperatures, the ω- signal returns to the base line. The even harmonics response S shows a maximum at $T=T_m$ as a function of temperature below T_c .This maximum is found at lower temperature than the maximum of L(T).

Similar results are shown for film B in Fig. 2b. In this case, the ω-signal is found down to lower temperature and does not show a maximum as a function of temperature. However, the 2ω-response is similar to that found on film A.

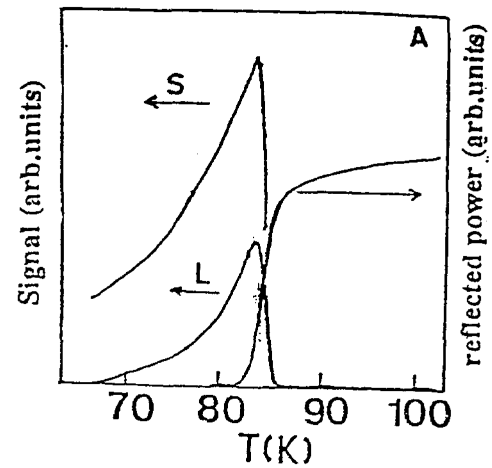

Fig 2.(a) Reflected power (right scale) and signals(left scale) at the frequency ω (L) and 2nω (S) as a function of temperature. Film A.Critical current from remanent magnetization : 2×10^6 A.cm^{-2} at 4.2K.

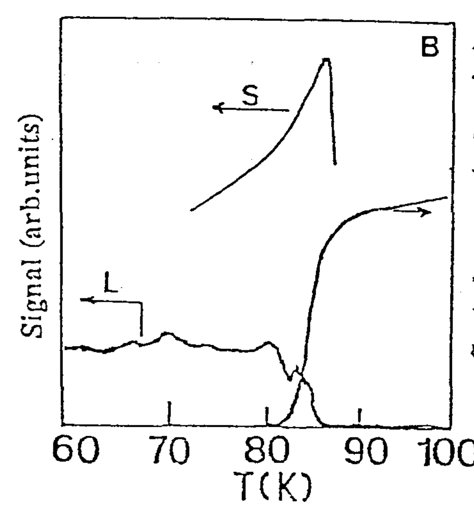

Fig.2(b). Same as in Fig.2(a) for film B. Critical current from remanent magnetization : 1.4×10^7 A.cm^{-2} at 4.2K.

The temperature dependence of harmonics amplitude is shown in Fig. 3 for film A. The amplitude of S shows a marked maximum at a temperature $T_m < T_c$. Also shown in this figure is the effect of a weak dc field H_0 coaxially superimposed on the ac field, on harmonics amplitude. While the reflected power remains unchanged under application of a weak field H_0, the amplitude of the fundamental decreases rapidly and the position of the maximum remains unchanged. The amplitude of S also

decreases under application of the dc field and the maximum at T_m broadens noticeably. The temperature interval where the response at 2ω is observed increases as the dc field amplitude increases. Fig.4 shows typical results for the variation of the S signal at different temperatures ($T_m<T<T_c$), as a function of the dc magnetic field. S decreases steeply in low fields, then shows a maximum and vanishes at a field H_i which is found to increase with decreasing temperature.
The irreversibility line close to Tc has been obtained on the same samples by dc magnetization studies. Fig. 5 shows typical hysteresis loops recorded for sample A in the temperature range $T_m<T<T_c$. From these data, the irreversibility lines shown in Fig.6, have been determined with the magnetization criterion of ≅ 10emu.cm^{-3} .In this temperature range, H_{irr} is found approximately linear with $T-T_c$.

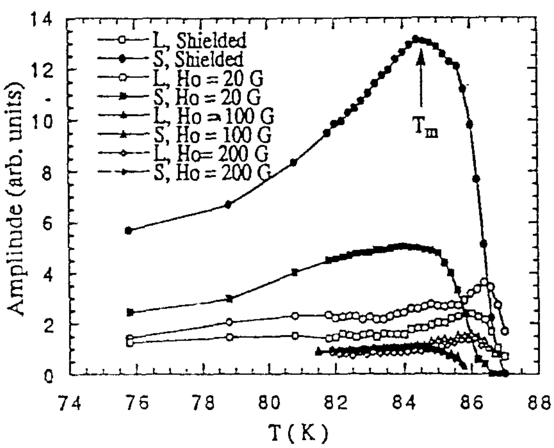

Fig. 3. Temperature dependence of even harmonics (S) and fundamental (L) amplitude for several values of the dc field H_0 .Film A.

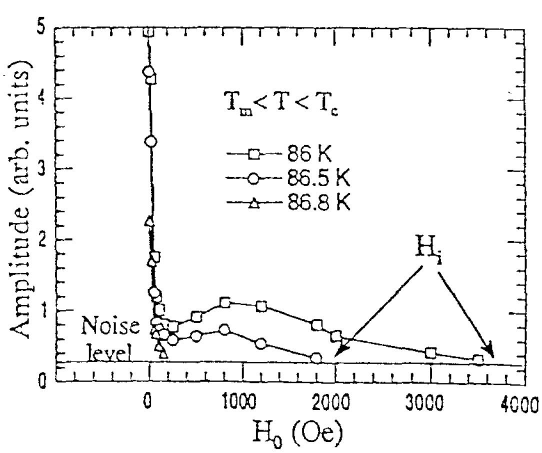

Fig.4. Amplitude of the even harmonics signal (S), as a function of dc field H_0 at different temperatures. ($T_m< T< T_c$); Film A.

As previously reported (10,11), the irreversibility line is displaced towards higher fields for thicker samples. For sample A, the field H_i corresponding to the vanishing of S vs H_0 is also shown. Except very close to T_c , H_i seems to be close to H_{irr} .

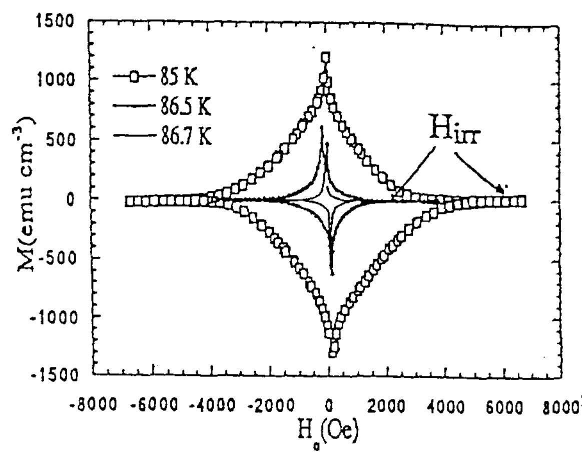

Fig.5.Typical hysteresis loops at $T<T_c$ ($T_m<T<T_c$); FilmA .

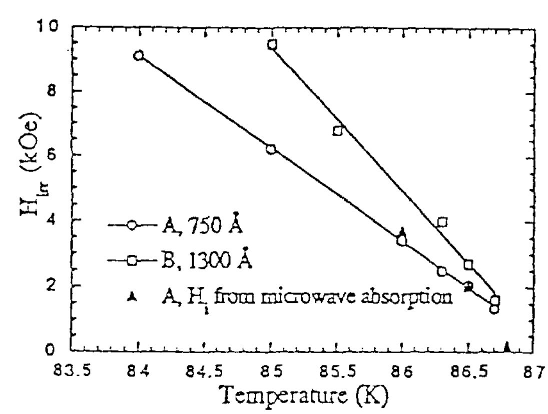

Fig.6 Irreversibility line obtained from hysteresis loops for films A (o) and B (□). The field H_i corresponding to the vanishing of the 2nω signal is also shown for film A,(▲).

4. Discussion

The dissipation phenomena observed in the field modulated microwave absorption experiments have to be attributed to the response of vortices to the modulation field and of the residual or applied dc fields. The non-linear response at 2ω is clearly predominant. In the Portis model (1) for microwave absorption, involving pinning and depinning of vortices during a modulation cycle, the microwave response is proportional to the number of weakly pinned vortices; this number is proportional to the absolute value of the field gradient. We have recently shown that this model can account for the response at 2ω (6).

Near T_m, vortices can be thermally depinned. We have proposed that a thermally activated flux flow mechanism is responsible for the signal at the excitation frequency. The maximum at T_m in the temperature dependence of even harmonics reflects a cross-over in the flux dynamics: diffusive flux motion is dominant above T_m and pinning at lower temperatures. Under the application of a dc field, the maximum at T_m broadens and the temperature interval where the harmonics are found becomes larger.This is due to a decrease of the height of the pinning barriers.

Fig.2 shows clearly a difference between the field modulated microwave response of films A and B. We suggest that the absence of a well defined peak for the ω-signal vs temperature might be due in the case of filmB, to the presence of CuO precipitates. These non-superconducting precipitates should not contribute to the modulated response , even though they might contribute to the residual surface resistance. However, they might be associated to film inhomogeneity and to regions in which vortex pinning could be weaker than in the rest of the sample, even at temperatures well below T_c. These regions would mainly contribute to the flux-flow response related to the ω-signal. On the other hand , the rest of the film would be responsible for the 2nω-response (signal S), which is similar to that found in film A. If this description is corroborated by more extensive studies, it will establish that the CuO precipitates have an important influence on the physical properties of $YBa_2Cu_3O_7$ films. Since the non-linear response contained in the 2nω signal is associated to weakly pinned vortices, it should vanish with increasing dc fields on the irreversibility line. This is consistent with the experimental data (Fig.5).

The irreversibility lines (Fig.6) obtained on MOCVD films are similar to what has been obtained on epitaxial films (11). Furthermore, the effect of thicknes, found on these films, is also found on MOCVD films. This corroborates that the thickness dependence of the irreversibility line is an intrinsic properties.

In conclusion, we have reported comparative studies of the irreversibility line and of field modulated microwave absorption on two films with different microstructural properties. This study corroborates that the non-linear microwave response related to the even harmonics found in the frequency spectrum is associated to weakly pinned vortices and therefore vanish on the irreversibility line. It also indicates that CuO precipitates may have a strong influence on the physical properties.

References

1 For magnetization properties of high-Tc superconductors, see for example: H.G.Schnack, R.Griessen, J.G.Lensink, C.J. van der Beek and P.H. Kes, Physica C, 197 (1992) 337; E.H. Brandt, Physica C, 195 (1992) 1.

2 A.M. Portis in Springer Series in Solid State Science, Vol. 90, J.G.Bednorz and K.A. Müller (eds), Springer Verlag, 1990, p. 278; H. Vichery and F.Beuneu, Physica C, 159 (1989) 823.

3 See for example : for thin films : R.H. Koch, V. Foglietti, W.J. Gallgher, G.Koren, A.Gupta and M.P.A. Fischer, Phys. Rev. Lett. 63 (1989) 1511.

4 C.Schlenker, J. Dumas, C.J. Liu and S.Revenaz in Chemistry of High Temperature Superconductors, C.N.R.Rao (ed), World Scientific 1992, p. 484.

5 M.Golosovsky, D.Davidov, E. Farber, T. Tsach and M.Schieber, Physica A , 168 (1990) 353.

6 B. Ravkin, M.Pozek, M.Paljevic and N.Brnicevic, Solid State Commun. ,70 (1989) 729.

7 O.Thomas, E.Mosang, J.Fick,F.Weiss, R.Madar, J.P. Sénateur, M.Ingold, P.Germi, M.Pernet, F.Labrize, L.Hubert, Physica C, 180 (1991) 42; O.Thomas, J. Hudner, M.Östling, E.Mossang, B. Chenevier, F.Weiss, D.Boursier and J.P. Senateur, this Conference.

8 S.Revenaz, J.Dumas and A. Gerber, Physica C, 180 (1991) 180.

9 S.Revenaz and J.Dumas, to be published.

10 C.J. Liu, C.Schlenker, J.Schubert,and B. Stritzker, Physica C, 180 (1991) 148 and to be published.

11 C.J. Liu, S.K. Agarwal, R. Buder, J. Schubert and C. Schlenker in "High-Tc Superconductor Thin Films", ed. L. Correra, Elsevier Science Pub. (1992) p. 177.