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Non-resonant microwave absorption in high-T_c thin films*

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Magnetic-field-dependent non-resonant microwave absorption in thin film samples of various high-T_c superconductors is reported. Complex types of signals were observed as the temperature was lowered from T_c to ~10 K. Possible correlation between the thin film quality and the occurrence of the signals is suggested.

Rapid developments in the field of high-T_c superconductivity [1] have made the application of thin superconducting films in microelectronics feasible. Possible applications range from high speed chip interconnections to active devices. Much of the excitement comes from the fact that for the first time the superconductivity occurs at temperatures where it is practical to operate semiconductor devices e.g. microwave detectors, gallium arsenide field effect transistors, etc. Therefore, preparation of high quality thin films of high-T_c superconductors at sufficiently low processing temperatures and on technological substrates as well as their characterization are of obvious importance. In particular, the study of their microwave properties becomes essential for many applications. The phenomenon of the low-field non-resonant microwave absorption [2], detected by a lock-in technique commonly used in electron spin resonance (ESR) spectrometers, is one of the most interesting microwave features of the high-T_c superconductors. This phenomenon is the basis of a technique that is now widely used to establish the presence of a superconducting phase, the onset of superconductivity, and some other important properties of high-T_c superconductors. Since the technique had been successfully employed to the investigation of both ceramic and single crystal samples of high-T_c superconductors, it seemed to us quite natural to utilize it for thin film samples as well. Moreover, it has been shown recently [3] how field modulated signals by the lock-in technique can be related to the absorption in unmodulated fields. This result has lifted much of the previous confusion about the lock-in detected signals and has aided in their interpretation. Here, we present preliminary results of our low-field microwave absorption measurements on thin films of high-T_c superconductors Re–Ba–Cu–O (Re=Y, Gd, Eu, Nd, Dy, Sm) for a wide range of temperatures and show that useful information relevant to the sample preparation could be obtained.

The high-T_c superconducting thin films were prepared by magnetron sputtering from the corresponding sintered target onto different substrates [Al_2O_3, ZrO_2, and Si (100)]. For the thin film deposition and processing, we used a two-step-procedure [4]. The disc-like target of 40 mm diameter was prepared by established solid-state reaction technique [5]. As sputtering gas, we used a mixture of ~20 Pa oxygen and ~40 Pa argon. The discharge was run at 130 V and 0.5 A. The substrates were mounted at a distance from 40 to 60 mm from the target surface on

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a steel stripe. The films were deposited at substrate temperatures from 740 to 820°C. The sputtering time was approximately 30 min and the film thickness was about one micron. After deposition, the films were cooled to 400°C, held at this temperature up to 30–60 min at 1 bar oxygen pressure and then cooled in the deposition chamber to room temperature.

The superconducting transition temperatures were determined by a standard DC four-probe method (Al stripes connected to the films by silver paste) and the non-resonant microwave absorption method [2]. The measuring currents through the films ranged from 20 to 200 μA. The characterization of samples by X-ray diffraction has shown that the prepared thin films have a highly textured growth with the *c*-axis predominantly perpendicular to the substrate surface. Gd–Ba–Cu–O thin films that were sputtered directly on a (100) silicon substrate without any buffer layer were polycrystalline.

Microwave absorption measurements were made using a Brüker ER-200D ESR spectrometer operating at X-band with a TE_{102} microwave cavity. To facilitate the measurements at and beyond zero field, the magnet was biased by an external constant current power supply connected to the rapid-scan coils of the ESR spectrometer. The samples were mounted on a holder in an Oxford Instruments ESR-10 helium flow cryostat inside the microwave cavity. The temperature could be varied from 300 down to 2.4 K. The temperature was independently measured by a thermocouple positioned in the vicinity of the sample (only ~0.2 mm distant). The measurements were done for two different positions of the samples with respect to the scanning DC magnetic field: B_o perpendicular to the substrate (usually the (*a*, *b*) plane of the thin films) and B_o parallel to the substrate.

Microwave signals detected below T_c in our thin film samples have shown a greater complexity than those observed earlier in ceramics [2,3] and similar to those of single crystals [6]. Modulated signals recorded in Sm–Ba–Cu–O high-T_c thin film at different temperatures for DC magnetic field perpendicular to the substrate ($B_o \perp (a, b)$) are shown in fig. 1. In the absence of a hysteresis, a modulation and lock-in detection yields the first derivative of the unmodulated microwave absorption curve [3]. The inspection of the signals in fig. 1. shows that when the temperature of the thin film is reduced below 85 K,

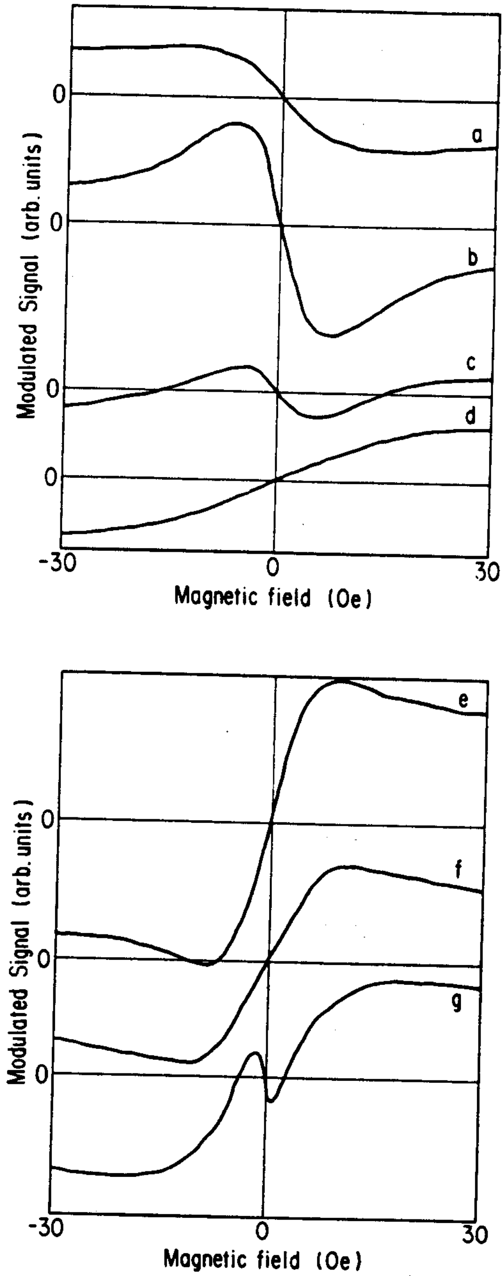

Fig. 1. Modulated signals detected in the high-T_c thin film Sm–Ba–Cu–O deposited on a ZrO_2 substrate for $B_o \perp (a, b)$ at different temperatures: (a) 85 K, (b) 84 K, (c) 83 K, (d) 82 K, (e) 76 K, (f) 72 K, (g) 64 K.

one observes first a signal with a maximum at $B_0=0$ (henceforth called Max 1), then a signal with a minimum at $B_o=0$ (henceforth Min), and, at still lower temperatures, a superimposed narrower and weaker second signal with a maximum at $B_o=0$ (henceforth Max 2). Similar signals were observed also in Eu–Ba–Cu–O thin film. Figure 2 shows the temperature dependence of the peak-to-peak amplitudes of the modulated signals denoted above as Max 1, Min and Max 2 in the Eu–Ba–Cu–O sample for DC magnetic field perpendicular to the substrate, i.e. $B_o \perp (a, b)$.

In order to show that the observed absorption maxima, inferred from the modulated microwave signals, are real and not instrumental, we have monitored directly the response of the spectrometer's preamplifier to DC magnetic field sweep. Figure 3 shows the change of the microwave absorption with temperature for a DC magnetic field scan from -30 Oe in Eu–Ba–Cu–O high-T_c thin film deposited on a ZrO_2 substrate. Here, negative fields are reversed in direction with respect to positive fields. The applied DC field is parallel to the sample substrate ($B_o \parallel (a, b)$). There is no qualitative difference in the preamplifier responses taken for B_0 perpendicular to the substrate; they are only narrower.

It follows immediately from an inspection of fig. 3 that the microwave response in an Eu–Ba–Cu–O sample develops at $T\sim 85$ K as a broad absorption maximum centered at $B_o=0$. The absorption peak

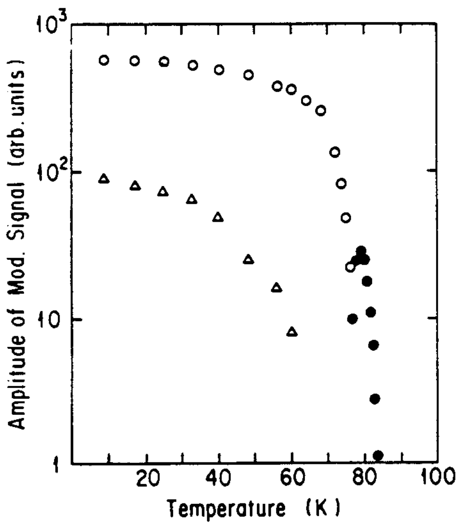

Fig. 2. Temperature dependence of the peak-to-peak amplitudes of the modulated signals, Max 1 (●), Min (○) and Max 2 (△), in the high-T_c thin film of Eu–Ba–Cu–O deposited on a ZrO_2 substrate for $B_o \perp (a, b)$.

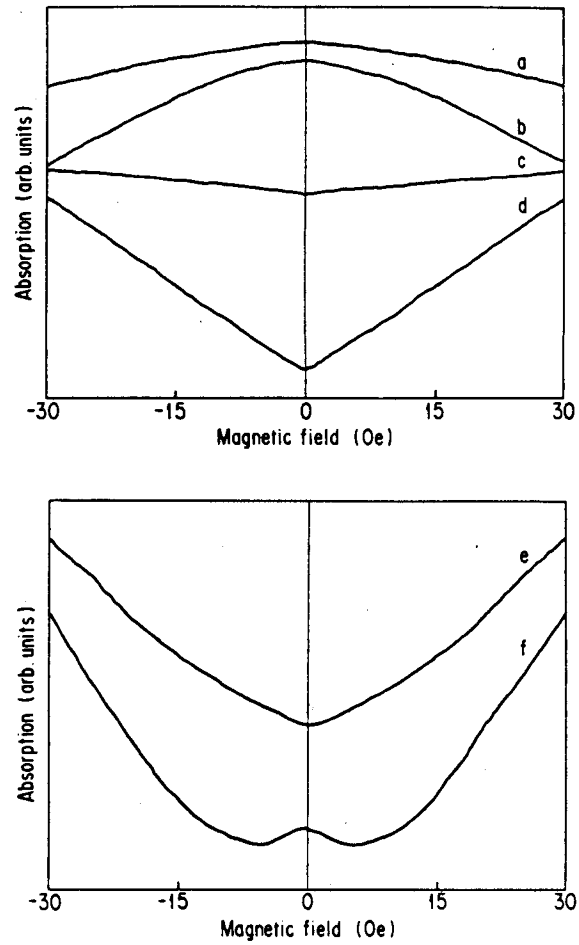

Fig. 3. Response of the ESR spectrometer's preamplifier to a DC magnetic field sweep with $B_o \parallel (a, b)$ for high-T_c thin film of Eu–Ba–Cu–O deposited on a ZrO_2 substrate recorded at different temperatures (negative fields are applied opposite positive fields): (a) 82 K, receiver gain (RG) = 10; (b) 79 K, RG = 5; (c) 76 K, RG = 5; (d) 74 K, RG = 5; (e) 64 K, RG = 1, 22; (f) 40 K, RG = 1.

first increases with decreasing temperatures and afterwards gradually diminishes. Then the absorption minimum starts to develop and increases precipitously for temperatures a little below the transition. The temperature $T_{co}=81$ K, at which this sample reaches its zero-resistivity state. With further decrease of temperature the signal flattens considerably. In contrast to the observation made on very good quality single crystals of $YBa_2Cu_3O_7$ [6], the absorption minimum in thin films persists to the lowest temperature measured. Below 60 K a narrow and weak (in comparison with the main signal) absorption maximum (Max 2) clearly develops superimposed on the broad minimum. The maximum grows with decreasing temperatures down to ~20 K, where it levels off.

One might consider the evolution of the signal from fig. 3(e) to 3(f) also as a splitting from a single minimum to a double minimum signal so that the maximum at $B_o=0$ is only apparent. However, the fact that the signal with a maximum at $B_0=0$ has been observed alone at higher temperatures leads us to believe that the evolution observed below 60 K is due to the onset of a microscopically independent second signal with a maximum at $B_0=0$, which originates in a fraction of the sample with a lower T_c.

The above experiment has confirmed once again that signals obtained by the experimentally much more convenient method of field modulation and lock-in detection do not contain modulation artifacts, but rather yield reliable data even in complex absorption cases.

Different experimental observations were made in some other thin film samples. Figure 4 shows the temperature dependence of the peak-to-peak amplitudes of the modulated signals in (a) Nd–Ba–Cu–O high-T_c thin film deposited on an Al_2O_3 substrate that exhibits a weak Max 1 signal and the main signal: Min (b) Dy–Ba–Cu–O high-T_c thin film deposited on a ZrO_2 substrate that exhibits the main signal min only.

The above-mentioned modulated signals were recorded for the DC magnetic field perpendicular to the substrate of the samples. Again, there is no qualitative difference in the signals when the magnetic field is parallel to the substrate; detected modulated signals are broader only.

From our study on a number of samples, it follows that Max type absorption signals appear only in samples of lower quality. Their microscopic origin might be common to the recently observed negative magnetoresistance in small superconducting loops [7].

We have also investigated the influence of both the modulation amplitude (MA) and the microwave power on the modulated signal in a Dy–Ba–Cu–O high-T_c thin film. Results (see fig. 5) clearly demonstrate that the sweep dependent component can be reduced by increasing either the modulation am-

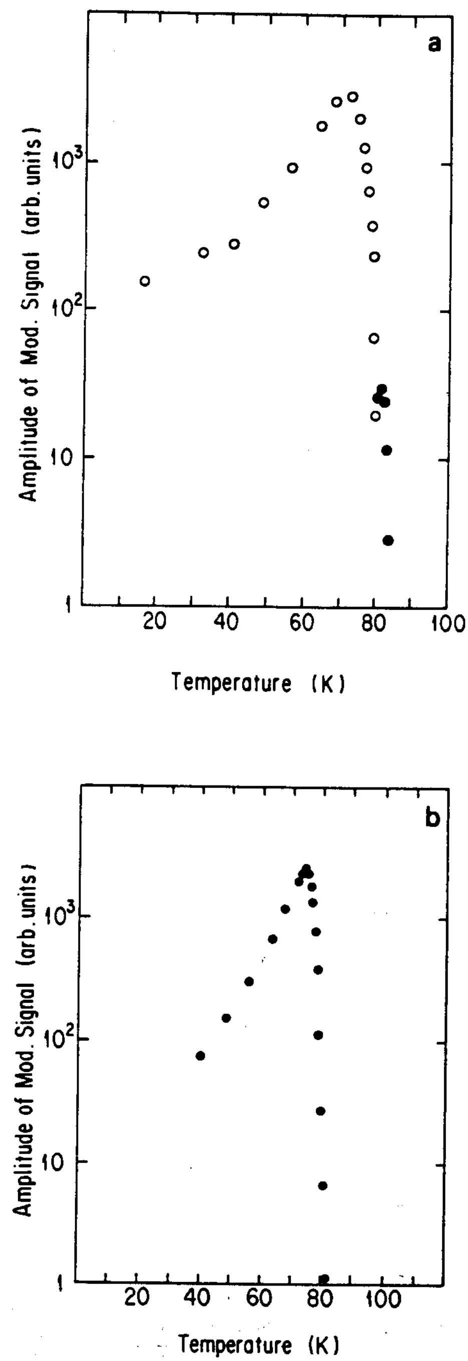

Fig. 4. Temperature dependence of the peak-to-peak amplitudes of the modulated signals for $B_o \perp (a, b)$ in (a) Nd–Ba–Cu–O high-T_c thin film deposited on a Al_2O_3 substrate Max 1 (•) and Min. (o) are present; (b) Dy–Ba–Cu–O high-T_c thin film deposited on a ZrO_2 substrate (the Min. signal present only).

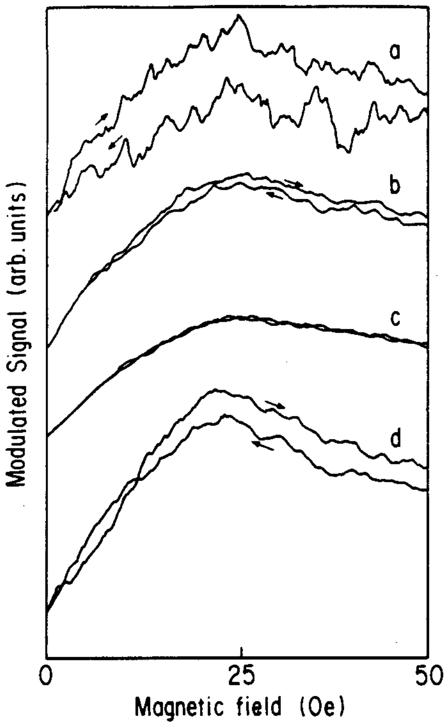

Fig. 5. The modulated signal (Min) in the high-T_c thin film of Dy-Ba-Cu-O for different modulation amplitudes and microwave powers at 59 K for $B_0 \perp (a, b)$: (a) MA=4 G, P=8 mW; (b) MA=16 G, P=8 mW; (c) MA=40 G, P=8 mW; (d) MA=4 G, P=80 mW.

plitude or the microwave power, as shown by Pozek et al. [8] for ceramic $YBa_2Cu_3O_7$. Hence, the main signal: Min shows the characteristic features of the microwave absorption in a granular superconductor with a dense weak link network.

Regarding the signal widths, we found that the modulated signals in high-T_c thin films are generally broader for the DC magnetic field parallel to their substrates in comparison with the perpendicular geometry. This is conceivable if the (a, b) planes are parallel to the substrates, because when the magnetic field is perpendicular to the (a, b) planes, the flux is more readily forced into the sample as shown by our microwave absorption measurements on a single grain of $YBa_2Cu_3O_7$ [10]. Considering the X-ray diffraction data on the films investigated, we come to the conclusion that the angular dependence of the modulated signals can give us further valuable information on the degree of the c-axis orientation of the high-T_c thin films.

Taking into account the results of our non-resonant microwave absorption measurements, the X-ray diffraction results, and the results of the transport measurements on the high-T_c thin films under investigation, it is possible to conclude that good quality films exhibit only the main signal corresponding to the absorption minimum and a weak, or zero, Max 1 signal. The higher the superconducting quality of the film, the narrower is the temperature interval below T_c in which this signal is observed (cf. fig. 4). An even more peaked temperature dependence was reported for thin films on a $LiNbO_3$ substrate [9]. In thin films of lesser quality one may observe one or two Max types signals. In our experience, a Max 2 signal at lower temperatures was observable only in those samples which had a relatively strong Max 1 signal at higher temperatures.

We may conclude that the method of the low-field non-resonant absorption of microwaves proves to be very useful for the characterization of high-T_c thin films and can serve as an aid in their preparation and quality improvements. For applications in high speed electronic circuits it is important to avoid high frequency losses and their fluctuations. Therefore, acceptable thin films should have vanishingly small absorption signals at the intended operating temperature.

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