Magnetic field modulation effects on the microwave transmission through superconducting thin films of Y-Ba-Cu-O

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We have measured the magnitude and phase of the microwave transmission through thin films of Y-Ba-Cu-O on the MgO substrate in the presence of a dc magnetic field H_0 and an oscillating magnetic field. The oscillating field modulates the microwave transmission in the superconducting state with the generation of many high-order even harmonics for $H_0=0$. For $H_0\neq0$ there is a symmetry-breaking and additional odd-harmonic generation. This strong nonlinear behavior may be explained by a model which assumes an array of Josephson junctions and vortex motion in the presence of a magnetic field. An estimate of the Josephson-junction area is obtained from the field dependence of the amplitude of the second harmonic.

The experimental study of electrodynamic properties of high-T_c superconductors provides useful information necessary for a basic understanding of the superconducting mechanism of these materials. This includes (i) microwave absorption in low magnetic fields,[1] (ii) nonlinear radio-frequency response.[2,3] The recent model of Xia and Stroud[4] accounts for both these classes of experiments. This model assumes superconducting grains weakly coupled into closed loops. The simultaneous observation of both phenomena in one experiment can prove the validity of this model. The measurement of microwave transmission through thin films in ac and dc magnetic fields is ideally suited for this purpose. Such measurements[5,6] may yield direct information on the complex conductivity $\sigma=\sigma_1-j\sigma_2$.

We report here a new class of measurements of the amplitude and phase of microwave transmission through thin films of the Y-Ba-Cu-O superconductor in the presence of a dc magnetic field ($H_0<500$ G) and an ac magnetic field $H_1\sin(2\pi ft)$($H_1<20$ G; $10<f<10^4$ Hz).

The experiments were carried out on thin films (~1 μm thick) on MgO substrate with a thickness of 1-2 mm. The substrates were prepared by the cleavage of large MgO single crystals originating from the Dead Sea. The superconducting films were prepared by the spray pyrolisis method[7] in our laboratory, or were provided by Dr. G. Koren of the Technion, Haifa. At least six different films (all exhibiting the superconducting transition temperature $T_c=80$–85 K) were studied with consistent results. These films retain their superconducting microwave properties for at least a year after preparation.

The transmission measurements were carried out at X-band frequency (9.5 GHz). The experimental setup is shown in Fig. 1. The film-substrate combination is held in a frame across the waveguide flanges or across a small iris in the center of the waveguide. Special precautions were taken to eliminate any microwave leakage except through the film. The incident power is ~100 mW. The transmitted power is picked up electrically by a coaxial cable and measured with a crystal detector. The phase shift of the transmitted wave through the sample is determined by balancing the transmitted microwave with a reference arm using a precision attenuator and a reference phase shifter for a null readout. The sample is situated in a vacuum can immersed in liquid helium or nitrogen. The ac magnetic field perpendicular to the film (parallel to the axis of the waveguide) is produced by a small coil around the vacuum can. The dc magnetic field is parallel or perpendicular to the film and is produced by independent coils (Fig. 1). The modulated microwave transmission is analyzed using a spectrum analyzer or a lock-in amplifier.

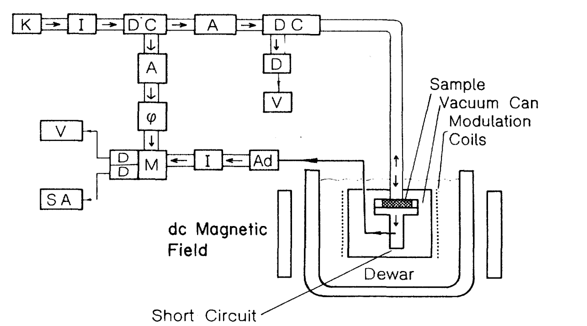

FIG. 1. Schematic description of the experimental system for the determination of the transmission (both magnitude and phase) and reflection. K—Klystron; I—Isolator; DC—directional coupler; A—attenuator; V—digital voltmeter; φ—phase shifter; Ad—adapter; M—mixer; SA—spectral analyzer.

Although the microwave properties were measured down to liquid-helium temperatures, we emphasize here our results in the vicinity of the superconducting transition temperature.

The experimental results can be summarized as follows:

(i) The temperature dependence of the intensity and phase shift of the transmitted wave in the presence and in the absence of the dc magnetic field is shown in Fig. 2. It is readily apparent that the microwave transmission properties are almost temperature independent in the normal state, in the range 85–100 K, but change dramatically upon transition to the superconducting state. Particularly, the transmission sharply decreases, the reflection increases, and a negative phase shift appears upon transition to the superconducting state. The transmission goes through a minimum at about $T=65$ K [Fig. 2(a)].

(ii) Applying a dc magnetic field does not affect the microwave properties in the normal state but has a significant effect in the superconducting state. Particularly, a dc magnetic field enhances the microwave transmission just below the transition temperature in the range 80–65 K [Fig. 2(a)] but decreases the transmission for $T<65$ K. The inset of Fig. 2(a) shows the dependence of the transmission on the magnetic field H_0 at different temperatures. Note that the transmitted power versus H_0 exhibits a clear minimum at zero field above 65 K, but a clear maximum below 65 K. At $T=65$ K the transmission is almost independent of H_0. The phase shift always decreases when a magnetic field is applied [Fig. 2(b)].

(iii) Applying an audio-frequency magnetic field, $H_1 \sin 2\pi f t$ has no effect on the transmitted power in the normal state (Fig. 3). However, an ac magnetic field modulates the transmitted power in the superconducting state and generates even higher harmonics ($2f$, $4f$, $6f$, etc.) of considerable intensity. Figure 3 exhibits the low-frequency power spectrum of the transmitted wave in a small oscillating magnetic field. Figure 4 exhibits the temperature dependence of the amplitude of the second harmonic. As can clearly be seen, the intensity of the second harmonic dramatically increases below T_c and exhibits a clear maximum. It almost vanishes around $T=60$ K [corresponding to the field-independent transmission, see inset of Fig. 2(a)]. Although not shown here, the amplitude of the second harmonic increases again at lower temperatures, changes phase, and goes to saturation. These features are consistent with the field dependence of

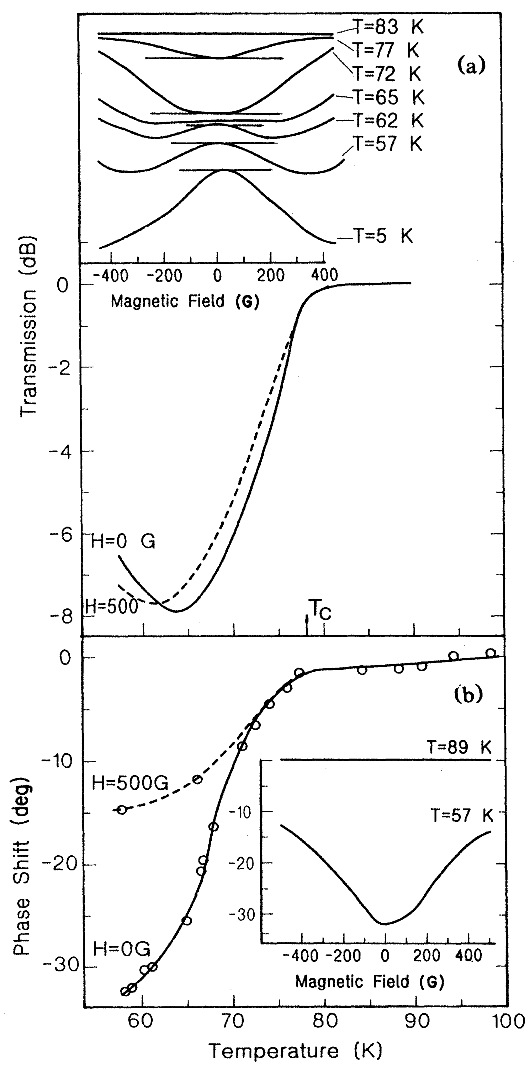

FIG. 2. (a) Transmission amplitude vs temperature at $H_0=0$ and 500 G. Inset: The transmission vs dc magnetic field at different temperatures. The level of the transmission at zero field is denoted by horizontal lines. (b) Temperature dependence of the phase shift at $H_0=0$ and 500 G. Inset: The phase shift vs dc magnetic field in normal and superconducting states.

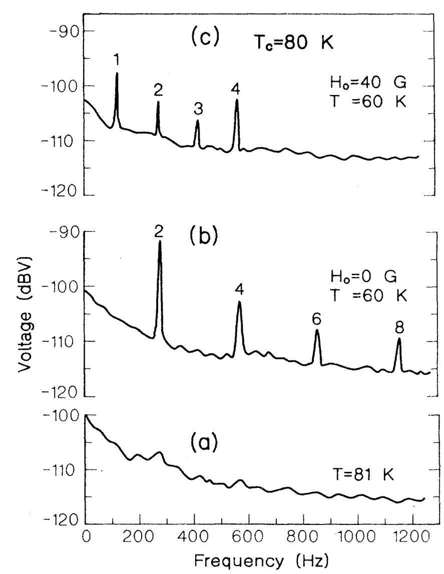

FIG. 3. The low-frequency spectrum of transmitted power through a film ($T_c=80$ K) in the presence of an ac magnetic field perpendicular to the film. (a) Normal state, $H_1=15$ G, $f=132$ Hz, $T=81$ K. (b) Superconducting state, $H_1=15$ G, $f=132$ Hz, $H_0=0$, $T=60$ K. (c) Superconducting state, $H_1=15$ G, $f=132$ Hz, $H_0=40$ G, $T=60$ K. Note the even harmonics in (b) but the generation of additional odd harmonics in (c).

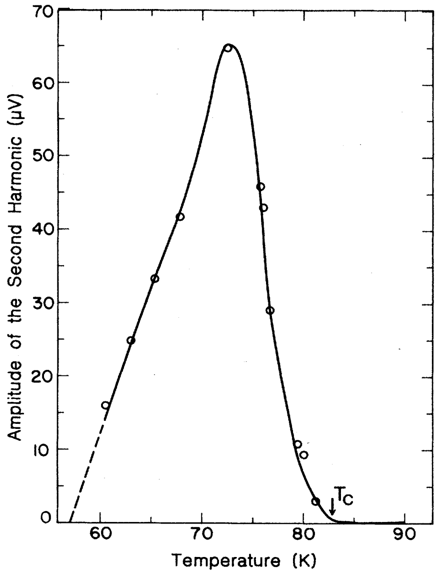

FIG. 4. The temperature dependence of the amplitude of the second harmonic.

the transmission [inset, Fig. 2(a)].

(iv) Applying an additional dc magnetic field H_0 (either parallel or perpendicular to the film) results in symmetry breaking and in the generation of odd harmonics in addition to the even harmonics (Fig. 3). Figure 5 shows the intensity of the first and second harmonic as a function of H_0. Clearly, a dc magnetic field of the order of 20 G significantly suppresses the second harmonic (Fig. 5).

Following Ref. 8, we have recalculated the transmission coefficient T and phase shift $\Delta\Theta$ for a film of thickness d on a substrate with thickness l and refraction index n placed across a *TE* waveguide with impedance Z. In our calculations we have assumed that the film thickness d is significantly smaller than the normal-state skin depth δ and the penetration depth in the superconducting state, $\lambda (d \ll \lambda, \delta)$. These assumptions are justified in our case: δ is estimated to be 80 μm from dc conductivity measurements, and λ in films is estimated to be between 1 and 3 μm.[5,6] However, λ should be significantly larger in the vicinity of T_c, where most of our experiments were carried out. One obtains for the transmission coefficient T, and the phase shift $\Delta\Theta$, the following expressions:

$$T = 4n^2\{[(2+\sigma_1 dZ)^2+(\sigma_2 dZ)^2]n^2\cos^2\kappa l + [(n^2+1+\sigma_1 dZ)^2+(\sigma_2 dZ)^2]\sin^2\kappa l - n(n^2-1)\sigma_2 dZ\sin 2\kappa l\}^{-1}, \quad (1a)$$

$$\Delta\Theta = \arctan\{[(n^2+1+\sigma_1 dZ)\sin kl - n\sigma_2 dZ\cos\kappa l] \times [(2+\sigma_1 dZ)n\cos\kappa l+\sigma_2 dZ\sin kl]^{-1}\}. \quad (1b)$$

Here κ is the propagating wave vector in the substrate. Using Eqs. (1a) and (1b) as well as the experimental data (Fig. 2) we have evaluated the real and imaginary part of the complex conductivity σ_1 and σ_2. It is clearly seen that both σ_1 and σ_2 increase upon going to the superconducting state; σ_1 goes through a maximum at $T = 65$ K, while σ_2 saturates at low temperatures. The temperature dependence of σ_1 and σ_2 is consistent with the results of Nichols *et al.*[5] and can be described either by the BCS theory with gap parameter $2\Delta(0)/k_B T_c = 3.5$ or in terms of an effective granular medium picture.[6] The leveling off of σ_2 at low temperatures can be attributed to microwave leakage through microcracks and normal regions.[5]

Certainly, the new important result obtained in these experiments is the dependence of σ_1 and σ_2 on the magnetic field (Fig. 6). We attribute the change of σ_1 and σ_2 in the magnetic field to the appearance of vortices (primarily in the weak links). The motion of vortices in the microwave field produces additional dissipation and affects σ_1. In these granular superconductors σ_2 to a large extent is determined by superconducting currents flowing through weak links. The appearance of vortices reduces

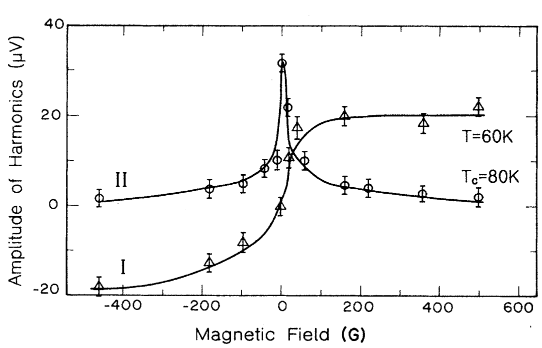

FIG. 5. The dependence of the amplitudes of the first and second harmonics on the magnitude of the dc magnetic field parallel to the film. $H_1 = 15$ G; $f = 132$ Hz.

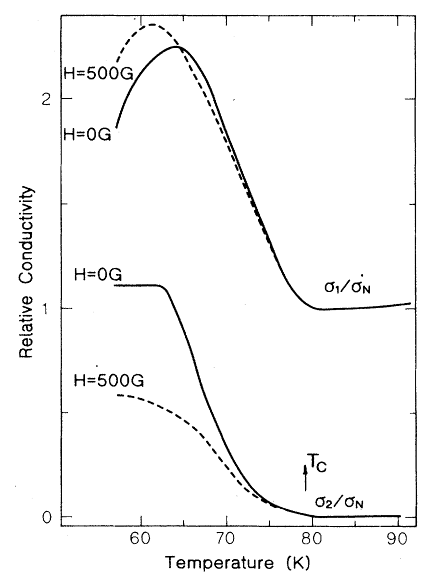

FIG. 6. The temperature dependence of σ_1 and σ_2 calculated using the data in Fig. 2 and Eqs. (1a) and (1b).

the critical current of these links and thus decreases σ_2.

The modulation of the microwave by an oscillating magnetic field and the observation of high-order harmonics can be interpreted, at least in the vicinity of T_c, in terms of the network of Josephson junctions. Following Jeffries *et al.*[2] the amplitude of the microwave current I_c flowing through a junction can be expressed as follows:

$$I_c = [\pi\Delta(T)/2eR_N]\tanh[\Delta(T)/2k_BT] \times \sin(HS/\phi_0)/(HS/\phi_0)\,, \quad (2)$$

where R_N is the resistance in the normal state, S is the junction area, ϕ_0 is the flux quantum, and H is the magnetic field which can be expressed as

$$H = H_0 + H_1 \sin(2\pi f t)\,. \quad (3)$$

Thus, I_c is time dependent and the microwave current is amplitude modulated. Using (2) and (3) together with a Fourier analysis, it is easy to demonstrate that in the absence of a dc field ($H_0=0$) only even harmonics of the ac frequency appear; while, if $H_0 \neq 0$, additional odd harmonics are generated in agreement with experimental data (Fig. 3). From (2) it is seen that with increasing H_0 the second harmonic is significantly suppressed in good agreement with the data in Fig. 5. From the field dependence of the second harmonic amplitude (Fig. 5) it is possible to estimate the junction area, S. Taking $SH_0/\phi_0=1$ for a field of 20 G at which the amplitude of the second harmonic is 0.7 of its maximum value, one obtains $S=10^{-8}$ cm^2. Certainly the assumption of a "single" Josephson junction is only an approximation and a complete analysis of the field and temperature dependence of the intensity of the various harmonics requires proper "averaging" over many junctions.[2-4]

The temperature dependence of the second harmonic amplitude (Fig. 5) can be explained, however, by Eq. (2) only at $T>70$ K. This may suggest an additional mechanism for the harmonic generation at $T<70$ K. We attribute the modulation of the microwave at lower temperatures to the motion of vortices under the action of the applied oscillating magnetic field. Note that at these temperatures the change of transmission in the magnetic field is dominated by energy dissipation in vortex motion (Fig. 6).

In summary, we have shown for the first time that the complex conductivity in the microwave range is field dependent. We have demonstrated the appearance of microwave modulation with many high-order harmonics in the presence of an oscillating magnetic field. The temperature dependence of these harmonics is consistent with dc magnetic field studies. The generation of high-order harmonics and the symmetry breaking in a dc magnetic field is analogous to those observed in the radio-frequency range.[2,3] However, we have shown that at least two mechanisms, namely (i) Josephson junctions and (ii) vortex motion, can explain this strong nonlinear behavior.

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