Non-linear response of ac conductivity in narrow YBCO film strips at the superconducting transition

J G Ossandón[1], **J L Giordano**[1], **P Esquinazi**[2], **K Kempa**[2], **U Schaufuss**[2] and **S Sergeenkov**[3]

[1] Facultad de Ingeniería, Universidad de Talca, Chile
[2] Abteilung Supraleitung und Magnetismus, Universität Leipzig, Germany
[3] Bogoliubov Laboratory, Joint Institute for Nuclear Research, Dubna, Russia

Abstract. Measurements of higher harmonics of the ac voltage response in YBCO thin film strips under low amplitude and low frequency harmonic excitation, as a function of temperature, show a non linear response of the conductivity in the superconducting transition interval. The third and fifth harmonics of the local voltage as a function of T exhibit a peak near T_c and their amplitudes seem to be closely related to the T-derivative of the first harmonic. The peaks are linearly dependent on the current amplitude and do not depend on frequency. The observed data are partially interpreted in terms of ac current induced thermal modulation of the sample temperature added to strong thermally activated fluctuations in the transition region. The fit of the model to the data gives information of some sample properties such as zero temperature critical current, zero onset resistance and thermal boundary conductance.

1. Introduction

Harmonic excitation of superconducting materials in bulk or film samples, at low or high frequencies, has been a standard technique to extract significant information of some fundamental materials (intrinsic as well as extrinsic) properties, as for instance, order parameter relaxation time, thermal conductivity, specific heat, critical temperature and others [1-7]. Moreover, measurement of higher harmonics has been shown to be a novel approach to derive the flux creep exponent in YBCO ring samples [8]. Particular interest has been generated lately due to the possibility, theoretically predicted [9-10], of determining the lifetime constant of *metastable* Cooper pairs in high-T_c superconductors (HTS). This prediction rests upon the assumption that the non linear response to a harmonic excitation near T_c has two origins: thermal oscillations from power dissipation and *depairing effects* caused by the electric field. Since the life time constant is directly involved in the depairing effects [10], its experimental determination should be possible after a careful separation of these two contributions. A particular feature of the electric field contribution, to be used for its identification, is its cubic power law dependency on the applied current amplitude. We report here detailed measurements of the ac conductivity in YBCO narrow thin film strips as a function of temperature, from far above T_c to full superconducting state, following the predicted methodology [10] with the aim of exploring such a possibility.

2. Experimental aspects.

The samples for this experiment were two rectangular pieces of $\sim 4x5\ mm^2$ size, both cut from a plate consisting of a *470 μm* thick, sapphire (Al_2O_3) substrate, having a *~240 nm* thick YBCO film deposited by laser ablation on either side. For better adhesion, a *~10 nm* thick cerium oxide layer was interposed between the YBCO films and the substrate. A *200 nm* thick gold layer covered both YBCO films on either side of the plate, which were further patterned for electric contacts. The two rectangular pieces were labeled I/3/1 and II/3/2/1 respectively.

      Using chemical etching and photolithographic techniques, the gold layer on each sample was partially removed and appropriately patterned to form a gold mask, consisting of a parallel array of

golden fringes, each fringe being *100 microns* wide and *100 microns* apart from each other. Finally, in direction perpendicular to the golden fringes, a parallel array of deep wrinkles was cut in order to create long, narrow YBCO-film strips of constant width, having on their surface a uniform sequence of golden electrodes available for electric contact. The transverse-to-the-golden-fringes wrinkles were carefully cut using two different techniques: a laser beam method in sample II/3/2/1, and an ion-beam method in sample I/3/1. The cuts were done at the Institut für Oberfläche Modifizierung (IOM) in Leipzig, Germany. In this way, narrow YBCO-film strips of, alternatively, *100*, *150*, and *200 microns* width, were prepared. The laser- as well as the ion beam- wrinkles (both ~10 microns wide) was deep enough to cut through the three layers (gold mask, YBCO film and CeO_2 buffer layer) and penetrate all the way down to the insulating substrate in order to insure electrical insulation from each other. Detailed description of the samples preparation and other experimental details can be found in reference [11]

A first series of measurements was done on a 150 microns wide strip ("Strip 3") of the laser cut sample II/3/2/1. Using four contacts geometry, a harmonic current of constant amplitude was sent through the strip and the voltage response was measured with the aid of a lock-in amplifier to detect higher harmonics. Preliminary results of these measurements are published elsewhere [11]. A new series of measurements, this time on Strip 1 of the ion beam cut sample I/3/1, has been carried out recently in order to put to a test our preliminary findings. For a wider exploration, we have expanded the range of some excitation parameters as well as the sweeping range of the sample temperature, and improved the measurements accuracy.

The harmonic excitation was in each case a sinusoidal current having alternative constant amplitudes ranging from *0.5 mA* to *20 mA* (rms-values) and frequency values ranging from *3.33 Hz* up to *1000 Hz*. A temperature controller Lakeshore 330 was programmed to set the sample temperature in the interval from *85 K* to *100 K*, stabilized within *0.05 K* at each data point, with sweeping steps of *0.1 K*. The frequency values were chosen far from any simple multiple or sub-multiple of the *50 Hz* basic line frequency in order to avoid disturbing couplings or resonance effects.
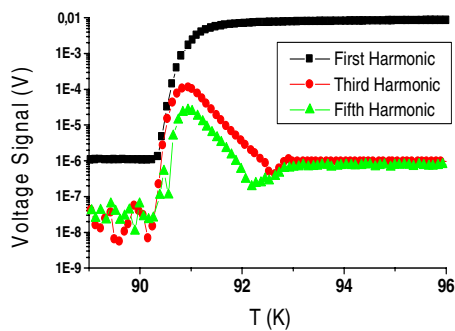

Figure 1: Higher harmonics of the voltage response as a function of T.

3. Experimental results.

Figure 1 shows a typical experimental result: it depicts on a logarithmic scale the modulus of the first, third and fifth harmonics signals as a function of temperature of the local voltage along strip 1 of sample II/3/1, for an applied ac-current of rms-amplitude $I_a = 1$ mA and frequency $f = 333$ Hz. No even harmonic signals are detected. In the temperature interval, which extends from 92.7 K (onset-T_c) down to 90.3 K, not only the *third* harmonic (as discussed in reference [12]) but also the *fifth* harmonic show clear, Λ-shaped peaks, whose amplitudes seem to be roughly proportional to the slope of the first harmonic signal. Using different applied currents, it was observed that the maxima of these so called "lambda peaks" are closely proportional to the current amplitudes and independent of the chosen frequencies. It was also observed that the position and limits of the peaks as a function of T did not shift with increasing current amplitudes or frequencies, within the experimental range. The transition from the normal to the superconducting state is very well defined by the appearance of these higher harmonics peaks as a function of T. Indeed, the Λ-peaks emerge at a critical temperature (the so called "*zero onset* T_c") where thermal fluctuations start to affect the order parameter and disappear at a T value ("*full* T_c") where the transition is complete. The presence of the lambda peaks has prevented us from detecting the theoretically predicted [9-10] contribution of the electric field to the non linear response of the conductivity at the superconducting transition ("*depairing effects*").

4. Theoretical discussion of the lambda peaks.

In our previous publication [11], some of us proposed a simple model to interpret the lambda peaks based upon two superposed effects: a current induced thermal oscillation of the sample temperature added to a stochastic, thermally activated fluctuation of the order parameter in the transition region. According to this interpretation, the normal state conductivity at each particular temperature T_o is affected by thermal oscillations around T_o whose mean amplitude Θ is directly proportional to the power dissipation and inversely proportional to the thermal conductance G between the film and the substrate. Due to these thermal oscillations, the voltage across the sample is a non linear function of T, having higher, odd harmonics (first, third, fifth…) whose amplitudes (in the limit of small enough currents) can be expressed, after a straightforward calculation [9,11], as:

$$V_{1\omega} = R_o I_a \{ 1 + \tfrac{3}{4} R'_o \Theta / R_n + \tfrac{5}{8} \Theta^2 (R'^2_o + \tfrac{1}{2} R_o R''_o) / R_n^2 \} \tag{1}$$

$$V_{3\omega} = - R_o I_a \Theta \{ \tfrac{1}{4} R'_o / R_n + 5 \Theta (R'^2_o + \tfrac{1}{2} R_o R''_o) / 16 R_n^2 \} \tag{2}$$

$$V_{5\omega} = R_o I_a \Theta^2 \{ (R'^2_o + \tfrac{1}{2} R_o R''_o) / 16 R_n^2 \tag{3}$$

where R_o is the resistance at T_o; R'_o and R''_o are its first and second T-derivatives, respectively, and R_n is the normal state resistance at zero onset T_c (just before starting the transition into the sc state) [13].

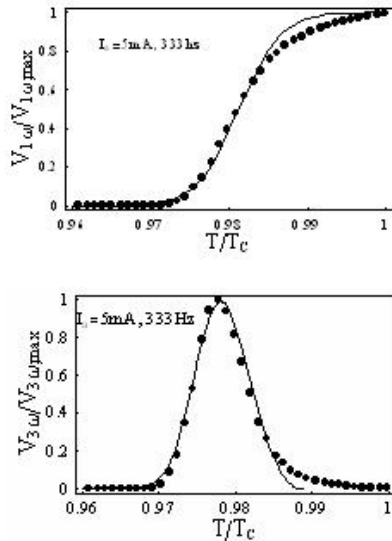On the other hand, thermally activated fluctuations, of a stochastic nature, affect the resistance due to formation of metastable Cooper pairs. According to Ambegaokar and Halperin [14], in the limit of small currents, the resistance $R(T)$ in the transition interval is reduced from its normal value just before the transition R_n by the factor $\mathcal{J}_o^{-2}(\gamma)$, where $\mathcal{J}_o$ denotes the *modified* Bessel function of zero order, and $\gamma \approx U/k_B T$ is the normalized barrier. Here U is the activation energy, which in this case we express in terms of a sample parameter L, the applied current amplitude I_a and the critical current $I_c(T)$ simply as $U = \tfrac{1}{2} L [I_a - I_c(T)]^2$. After some calculation, using the London relationship between $I_c(T)$ and $I_c(0)$, the argument of the Bessel function takes the form: $\gamma(T, I_a) = \gamma_o \{ \alpha - t^2 \}^2$, where $t \equiv T/T_c$, $\alpha \equiv 1 - I_a / I_c(0)$ and $\gamma_o \approx \tfrac{1}{2} L I_c(0)^2 / k_B T$. The parameters α and γ_o, as well as Θ, play the roll of fitting parameters in this model. Finally, we substitute the Ambegaokar and Halperin's law $R(T) = R_n \mathcal{J}_o^{-2}(\gamma)$, into the general expressions (1),(2),(3). A fit of the model to the experimental data, for the typical case of current amplitude 5 mA and frequency 333 Hz, is shown in figure 2, where we have plotted the normalized first and third voltage harmonics $V_{n\omega}/V_{n\omega max}$ ($n = 1, 3$) as a function of reduced temperature.

Figure 2: normalized first and third voltage harmonics as a function of reduced temperature T/T_c; a fit of the model to the experimental data.

5. Results

The first and third harmonics of the voltage response are fairly well described by the above simple model. The best fit through the data points in these cases yields the following approximated values for the model parameters: $\gamma_o \sim 1500$, $\alpha \sim 0.987$ and $\Theta \sim 2$ mK. From the value of α we obtain $I_c(0) \sim 0.4$ A which, in turn, together with γ_o can be used to estimate the sample inductance L. The result gives $L \sim 0.000025$ pH. Besides, using the experimental value of $R_n \approx 2.8$ Ohm for $I_a = 5$ mA, we obtain for the

estimate of the thermal boundary conductance between the strip and the substrate $G \approx 0.035\ W/K$. The above thermal modulation model does not fit well the fifth harmonic data. Indeed, the second T-derivative of $R(T_o)$ appearing in the second term of the r.h.s. in (3) introduces an oscillatory T-dependence [15]. Although the experimentally observed fifth harmonic shows an oscillatory behavior near T_c onset, its amplitude is smaller than that given by the model.

6. Conclusions

Measurements of the third as well as of the fifth higher harmonics of the ac voltage response in YBCO thin film strips under low amplitude (of a few mA) and low frequency (less than 1 kHz) harmonic excitation, as a function of temperature, exhibit a peak (so called "lambda peak") in the superconducting transition interval. The maxima of these peaks seem to be closely related to the T-derivative of the first harmonic and are linearly dependent on the current amplitude. They do not show significant changes with frequency (within the experimental range from 1 to 1000 Hz). No cubic power law dependency on the applied current has been detected. The predicted "*depairing effect*" of the electric field is apparently hidden behind the lambda peaks and has not been extracted from the data. A simple model based upon thermal oscillations added to strong thermally activated fluctuations can be fitted fairly well to the first and third harmonic data, but it does not describe well the observed fifth harmonic T-dependence. The determination of the lifetime constant of the *metastable* Cooper pairs requires further theoretical analysis and finer experimental techniques. It still remains a challenge for the experimentalist.

8. References.

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[13] We note that equations 1, 2 and 3 of this paper are slightly different from equations 5, 6, and 7 appearing in reference [11] due to small typographic errors in that publication.

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[15] This second T-derivative term was inadvertently omitted in reference [11]