

*Physics*

STUDY OF FINE PECULIARITIES OF HEAT CAPACITY IN  $\text{YBa}_2\text{Cu}_3\text{O}_y$  FILMS BY MEANS OF THE LASER-COUPLED FLAT-COIL-OSCILLATOR

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Improvements in the “hybrid” imaging method, based on traditional Laser Scanning Microscopy in combination with the single-layer flat-coil-oscillator (SFCO) sensitive technique introduced by authors earlier, are discussed. This method is capable of imaging 2D-grained structure, as well as the normal-to-superconductive phase transition in flat thin high- $T_c$  superconductive materials with 1–2  $\mu\text{m}$  spatial resolution. It enabled to detect weakly expressed peculiarities of the heat capacity in  $\text{YBa}_2\text{Cu}_3\text{O}_y$  film (preceding the well-known specific heat “jump”), which are important for true understanding of the real nature of superconductivity phenomenon. The method uses a well-focused He-Ne laser beam as a probing signal, and the SFCO-technique as a high-resolution sensing tool.

**Keywords:** Cryogenic laser scanning microscope (CLSM), imaging technique, heat capacity of  $\text{YBa}_2\text{Cu}_3\text{O}_y$  film, high- $T_c$  superconductive (HTS) & low- $T_c$  superconductive (LTS) materials, normal-to-superconductive (N/S) phase transition, single-layer flat-coil-oscillator test-method (SFCO-technique).

**Introduction.** One of the reasons, why the nature of high- $T_c$  superconductivity (HTS) is not clear yet, is the lack of test methods for non-perturbing, sensitive study of the normal-to-superconductive (N/S) phase transition in these materials at the very beginning of superconductive (SC) state formation.

The problem of electron pairing above the Meissner expel arose only after 1986, when HTS materials were discovered by Bednorz and Müller [1]. The majority of scientists presently admit [2] that in case of HTS materials one needs to distinguish between the electron pairing and the rise of phase coherence, and consider these processes separately and independently of one another. So, for superconductivity in HTS materials both the electron pairing and the Cooper-pair

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condensation are required. The latter being also known as the onset of long range phase coherence among the pairs. In other words, it is widely admitted now that in HTS the quasi-particles become paired above the Meissner expel – above  $T_0$  and starting with  $T_c$  (Fig. 1), and start the formation of SC-condensate only at  $T_0$ , whereas in low-temperature (or conventional) SC-materials (LTS) the pairing and the onset of phase coherence are assumed to occur simultaneously (at the same moment  $T_0 = T_c$ ), as due to the relatively large sizes of Cooper pairs their wave functions overlap in LTS materials. However, the “paramagnetic” precursor to superconductivity [3, 4], detected by us in LTS tin (Sn), along with the fine thermal effect indirectly related to it (Fig. 2, it was detected as a precursor to the well known “jump” of heat capacity long ago by Corak [5], but unfortunately remained unnoticed) is a weighty argument in favour of the opposite opinion (to that stated in [2]) as regards the properties of LTS materials.

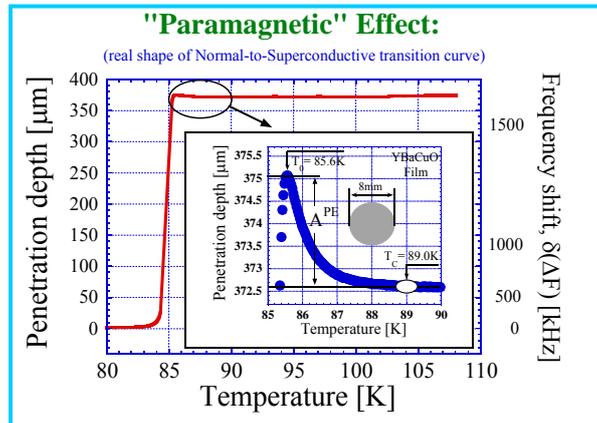


Fig. 1. N/S-phase transition of a disk-shaped YBaCuO film [4].

Inset: enlarged view of the “paramagnetic” effect (PE) at the beginnings of transition, which precedes the Meissner expel.  $A^{\text{PE}}$  is the effect’s height.

So, we assume that, there are no essential differences between the HTS and LTS materials regarding the said 2 processes: apparently, the electron pairing and onset of the phase coherence are separate and independent even in LTS. The difference is in the temperature scales of these materials, because for LTS materials these processes develop in a very narrow temperature range ( $\sim 10$  mK), while for HTS ones this range is much wider (e.g for  $\text{YBa}_2\text{Cu}_3\text{O}_y$  it is about 1 K [4], see Fig. 1). It is possibly due to this reason, why the separation of  $T_c$  from  $T_0$  in LTS materials is so problematic so far.

This problem is still outstanding due to the lack of sensitive methods for non-perturbing study of SC transition in pure (tiny) objects with very small signals, especially at the beginning of phase transition, where even SQUID-technique is unable to detect changes in the normal state’s “skin”-depth. In this connection the search for the above fine heat effect in HTS material (prior to the present study it was detected only in LTS material, see Fig. 2) becomes highly urgent. However, direct measurements of heat capacity is not an easy task (not to speak about the detection of minute changes at the very beginning of N/S-phase transition) in so small volume objects (thin-film structures, minute crystals, etc.).

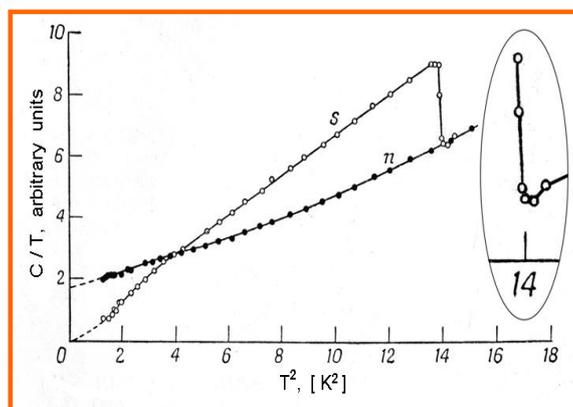


Fig. 2. Heat-capacity vs temperature curves, detected in tin (Sn) [5].

Inset: enlarged view of the effect noticed before the specific-heat's known jump. The symbol "s" corresponds to the SC-state, while "n" – to normal (superconductivity is suppressed by the magnetic field).

In the present study this problem was solved using the upgraded cryogenic laser scanning microscope introduced by our joint group earlier [6], based on a single-layer flat-coil-oscillator activated by a tunnel diode (TD) – the SFCO-technique [7, 8], integrated with focused laser scanning microscopy. It enabled us to detect fine peculiarities of the heat capacity of  $\text{YBa}_2\text{Cu}_3\text{O}_y$  film-bridge.

**Imaging Technique.** One of advantages of the SFCO-method is its ability to detect fine details of transition between the normal & SC phases in tiny, plate-like objects – with a better nanometer-scale absolute resolution at the measurement of SC-penetration depth's (by frequency shift of a testing TD-oscillator [7, 8]), and with about a nanowatt resolution at absorption measurements (by amplitude changes of the testing oscillator [9]). Besides, it can serve as a sensitive temperature probe with about 0.1 mK resolution [10]. Due to such unique capabilities of the SFCO-method, a strongly improved cryogenic imaging technique has been developed by us (so-called cryogenic laser scanning microscope – CLSM [11]), using a focused laser beam as a probing signal, capable of imaging properties of HTS thin-film structures with about 1–2  $\mu\text{m}$  spatial resolution. It operates as follows: the HTS film-bridge is illuminated point by point with a well-focused laser beam ( $\varnothing_{\text{beam}} \sim 1.5 \mu\text{m}$ ), which results in a slight local heating. As in our case the substrate of the film is transparent, and so, the amount of laser power leading to passed to the flat-coil based thermal sensor depends on peculiarities of the heat capacity (or heat conductivity) of the SC material under test in a lighted point of the film, positioned on the flat face of the detecting flat coil – leading to some changes in the amplitude and/or frequency of the SFCO-based TD-oscillator.

Moving the position of "X-Y" stage along both coordinates by stepper motors (with  $\sim 1 \mu\text{m}$  step [11]) and controlling these movements by the PC (in Lab-VIEW environment), we could make micrometer precision positioning of the laser beam and scan it over the surface of the  $\text{YBa}_2\text{Cu}_3\text{O}_y$  film. This enabled us to get 2D-images of the grained structure, as well as the N/S-phase transition of thin, plate-like HTS materials (e.g. film-structures) with about 1–2  $\mu\text{m}$  spatial resolution (see Fig. 3).

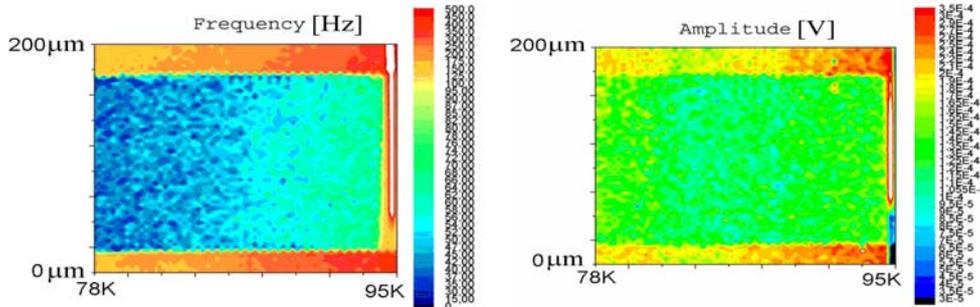


Fig. 3. Frequency shift (upper image) and amplitude change (bottom) of the SFCO-method based testing TD-oscillator, registered at the phase transition from the superconductive ( $T \sim 78\text{ K}$ ) to normal ( $T \sim 95\text{ K}$ ) of about  $160\ \mu\text{m}$  wide (vertical) HTS thin-film-bridge sample of the  $\text{YBa}_2\text{Cu}_3\text{O}_y$  composition.

**Results and Discussion.** In Fig. 3 the temperature dependence of N/S-phase transition of the test  $\text{YBa}_2\text{Cu}_3\text{O}_y$  film-bridge as detected by the *frequency shift* (the upper image) and the *amplitude dependence* (the bottom image) of the measuring TD-oscillator are shown. During the measurements the temperature of sample was varied very slowly from  $T \sim 78\text{ K}$  (superconducting state) to  $T \sim 95\text{ K}$  (normal state). In other words, the heating of the test sample progressed in a “natural” way – owing to the natural decrease of the level of liquid nitrogen in the supporting transport Dewar vessel, in consequence of which the gas flow through the cooling pipe of the Oxford Instrument’s “Microstat-HiRes Pillared” optical cryostat was slowly reduced (dropped). As a result, the sample passed via the phase transition in 3.5 hours (quasi-stationary). Besides, the scanning of bridge was made only along the  $Y$  axis, the  $X$  coordinate being fixed.

As it follows from Fig. 3, enormous changes of measured parameters (frequency and amplitude of oscillator) were detected in a narrow temperature range, in the vicinity of critical temperature. To clarify the cause of such an anomaly we present anew in Fig. 4 the modified amplitude data from Fig. 3, obtained by summarizing the *amplitude change signals* to get the 2D-raster image of laser beam power transmitted through the substrate. In other words, in Fig. 4 the 2D-raster image of the heat distribution in the HTS film under investigation is shown. Since the laser scanning microscope made by us is an instrument of bolometric nature (the flat pick-up coil detects the residual energy of the laser beam that has not been dissipated in the test film), the signals detected by the flat pick-up coil may be indirectly related to anomalies in heat capacity (as in the first approximation the heat conductivity may not have any noticeable anomalies in such a narrow range of temperatures in the vicinity of  $T_c$ ).

Actually, the curve in Fig. 4 for  $\text{YBa}_2\text{Cu}_3\text{O}_y$  film-bridge plotted by the amplitudes of the flat-coil TD-oscillator is amazingly resembling the temperature dependence of the heat capacity of LTS tin (Fig. 2), reported by Corak yet half a century ago [5]. At the same time the curve in Fig. 4 is also highly similar to the curve of SC-phase transition, shown in [3, 4], that was obtained by our group many years ago also for LTS tin and also with the help of TD-oscillator, but this time with a solenoid pick-up coil. Besides, there is an evident similarity of the weakly expressed peculiarity on our curve of heat capacity vs. temperature in Fig. 4 (shown by arrow) and the fine effect seen before the well known “jump” in specific

heat on the Corak's curve (Fig. 2) on the one hand, and the so-called “paramagnetic” effect first observed by us in LTS tin [3, 4] and then in HTS material of  $\text{YBa}_2\text{Cu}_3\text{O}$  composition both by us (Fig. 1) and Gantmakher [12] on the other hand.

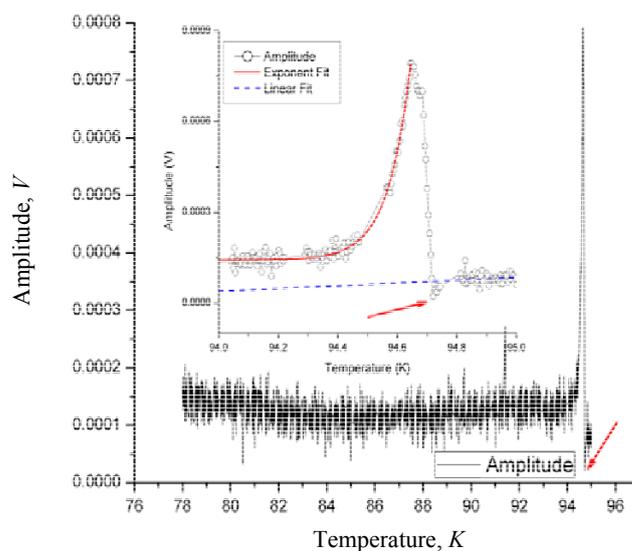


Fig. 4. SC-phase transition of the HTS thin-film bridge of the  $\text{YBa}_2\text{Cu}_3\text{O}$  composition, detected by the amplitude of the measuring TD-oscillator. The temperature of a sample was changed from 78 K (superconductive state) to 95 K (normal state) very slowly (during 3.5 hours). And besides, scanning of the bridge was done along the  $Y$  axis only, while  $X$  coordinate was fixed during the tests. Inset: enlarged part of the beginning of a phase transition. The circles are experimental data, the solid line is an exponential fit of the measured data, while the dashed line is the linear fit of a device curve (and/or the slow temperature dependence of the normal-state thermal characteristics of the sample).

Apparently, these similarities are not accidental and indicate that the reasons causing the “paramagnetic” effect also influence the thermal properties of SC materials. On the other part these similarities give evidence of a very high sensitivity of our present imaging technique (CLSM microscope) and witness the reliability of fine experimental data obtained by its means. Such a good agreement between the aforementioned independent data testifies to the importance of continuation of researches on these fine effects that would undoubtedly help to obtain substantial experimental information for better and more profound understanding of the genuine nature of superconductivity phenomenon on the whole.

**Conclusions.** As was noted by us elsewhere [13], such a comprehensive similarity of electromagnetic and thermal properties of superconductors (irrespective of whether these are HTS or LTS materials) may be explained proceeding from very recently formed concepts of the existence of 2 types of Cooper pairs – the “singlet” and “triplet” ones, in the superconducting materials (even in LTS) at the very beginning of SC-phase formation. According to our ideas [13], the amounts of “singlet” and “triplet” pairs in SC materials have unconventional temperature dependences that radically differ of one another upon cooling, as a result of which these fine effects make an appearance.

Taking into consideration the above experimental data we think that apparently the HTS and LTS materials **do not** essentially differ as regards the electron pairing (that begins from  $T_c$  temperature, see Fig. 1) and the onset of phase coherence (starting from  $T_0$ ). These are separate and independent even in LTS. The difference is only in the width of temperature scale. In case of conventional superconductors (LTS), the process develops in a very narrow temperature range ( $\sim 10$  mK), whereas in HTS materials the temperature scale is much wider (in  $\text{YBa}_2\text{Cu}_3\text{O}_y$  it exceeds 1 K), and, hence, the effects are observed incomparably easier.

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