

# HIGH-PRESSURE MICROWAVE STUDY OF $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ - $\text{Pb}(\text{Sc}_{0.5}\text{Ta}_{0.5})\text{O}_3$ COMPOSITE

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The ferroelectric relaxor  $\text{Pb}(\text{Sc}_{0.5}\text{Ta}_{0.5})\text{O}_3$ -superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  50% in weight composite exhibits the onset critical temperature  $T_c = 95$  K. High pressure studies yield the factor  $dT_c/dp$  of 1.0 K/GPa, close to the value observed for a pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  compound. The microwave absorption studies show the significant role of the intergrain weak links.

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## 1. Introduction

Since their discovery, the high-temperature superconductors have been the subject of a very intensive research effort. The high-temperature superconductivity research is unique in its interdisciplinary nature. Many aspects of solid state physics and chemistry are involved and are indispensable for the understanding of the problem.

The high-temperature superconductors are among the most complex materials studied by solid state physicists and chemists, and it is clear that excellent characterization of samples is required if the significance of experimental results is to be evaluated [1]. The correlations between the normal and superconducting state properties can be investigated by varying the basic physical parameters, such as the charge concentration or the interatomic distances. The application of hydrostatic pressure can change these parameters in a relatively clean manner, as compared to the change of charge concentration introduced by chemical substitution, where some undesirable effects may occur [2].

Information about the mechanism of superconductivity can be gained by searching for correlations between the pressure dependencies of superconducting parameters, such as the superconducting transition temperature, and the normal state properties such as the magnetic susceptibility and electrical resistivity. The pressure dependencies of  $T_c$  can be separated into two cases: (a)  $T_c(p)$  dependencies which arise from structural transitions or phase instability effects, and (b) intrinsic  $T_c(p)$  dependencies which reflect the changes in the interplanar spacings, density of states and phonon frequencies under high pressure [2]. On the other hand, the high pressure methodology allows to modify the distribution of the intergrain weak links [3] and to increase the intergrain critical currents [4, 5].

The presence of inter- and intragrain weak links causes difficulties in the separation of intrinsic and extrinsic properties of high-temperature superconductors, as well as reduces their potential applications, decreasing the critical current and critical fields. To minimise the weak-link behaviour, inclusions such as silver particles have been added to  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . The nonsuperconducting phases such as  $\text{Y}_2\text{BaCuO}_5$  or  $\text{BaTiO}_3$  have been added to increase the flux pinning and critical current density [6–8].

In this paper, we present the high pressure and microwave properties of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconductor– $\text{Pb}(\text{Sc}_{0.5}\text{Ta}_{0.5})\text{O}_3$  ferroelectric relaxor 50% in weight composite (abbreviated as YBCO–PST) with  $T_{c-\text{onset}} = 95$  K.

## 2. The sample preparation and structure

The composites were formed by mixing the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) and PST powders. Different concentrations were obtained by varying the PST weight. The mixture was then pressed at room temperature. The samples were heated in argon atmosphere up to  $900^\circ\text{C}$ , then the sintering temperature was decreased in oxygen atmosphere, with a plateau at  $450^\circ\text{C}$  during two hours. Next the samples were cooled down and air quenched.

Different characterizations were carried out systematically on the samples: (i) ac resistance measurements in a classical closed cycle helium PAR cryostat, in the range 20–300 K; (ii) scanning electron microscopy study with energy dispersion spectroscopy analysis; (iii) X-rays diffraction measurements.

X-rays diffraction patterns were obtained on a D 5000 Siemens equipment with the lambda Cu  $K_\alpha$  radiation Ni filtered. Identification of the diffractograms gives evidence that the main peaks of the two oxides are present, with relative intensities corresponding to the pure phases. The initial nominal composition of YBCO was identified as 6.8 in oxygen content. After sintering, it appears that the PST reflections mostly correspond to the disordered phase [9]. However, the traces of an additional phase corresponding to a modified pyrochlore phase appeared after the sintering. It was identified as  $\text{Pb}_{14}\text{Ta}_{10}\text{O}_{39}$  by J.C.P.D.S. computer file [10] and did not exist in the initial PST powder. No evidence of interfacial reactions between YBCO and PST is probably due to the high PST stability (melting point near  $1500^\circ\text{C}$ ).

### 3. Experimental results and discussion

The presented results concern a sample with 50% PST concentration in weight. The temperature dependence of resistivity shows that these composites are superconducting. This result is very surprising if we refer to metal-YBCO composites [11, 12], for which a metal concentration of 10% generally drastically decreases or suppresses the superconducting transition. In the normal state, the resistivity behaviour is metallic-like, as pure YBCO, with a room temperature resistivity about 0.8 m $\Omega$  cm. The onset of the superconducting state is at 95 K and the zero resistance temperature is 93 K, that is to say the same value as pure YBCO (Fig. 1).

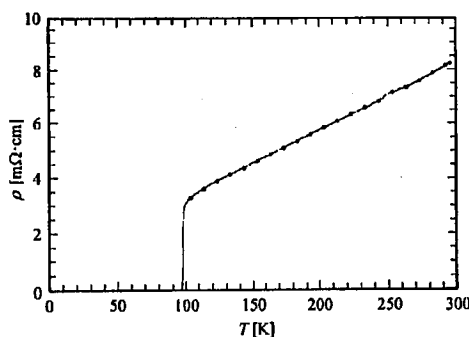


Fig. 1. The resistivity of YBCO-PST 50 weight-% composite sample versus temperature with  $T_{c-onset} = 95$  K and  $T_{c-zero} = 92.8$  K.

The superconducting transition could be monitored also by the microwave measurements. In this case, the critical temperature could be defined as the temperature at which the first appearance of the magnetically modulated microwave absorption (MMA) can be observed on cooling the sample [13].

The high-pressure measurements of the MMA were carried out by means of a high-pressure EPR probe attached to the standard EPR X-band spectrometer [3]. The pressure was applied to the steel high pressure chamber by means of transmitting liquid through a high pressure capillary [3]. In the 70–100 K temperature range, petroleum ether was used as the transmitting medium. The sample temperature was measured by a copper-constantan thermocouple. The required value of the pressure was set before cooling the sample. The temperature was then lowered at a constant pressure, using the liquid nitrogen as the cooling medium.

During the superconductivity transition, the temperature was changed very slowly (0.1 K/min) to minimise the temperature gradient between the sample and thermocouple. The sample was placed in an Al<sub>2</sub>O<sub>3</sub> microwave cavity in the maximal magnetic microwave field. The applied dc external magnetic field was modulated by a second modulation at a frequency of 80 Hz and 0.1–10 Oe amplitude. The configuration of all the magnetic fields was the same as in the standard EPR methodology, i.e. the second modulation field was parallel to the dc magnetic field and perpendicular to the microwave magnetic field. The temperature

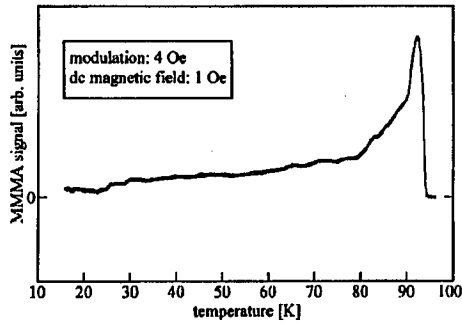


Fig. 2. Temperature dependence of the MMMA signal in YBCO-PST 50 weight-% composite.

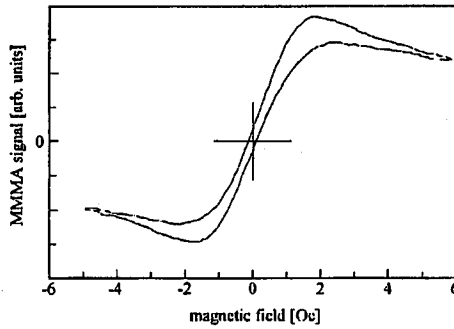


Fig. 3. Magnetic field dependence of the MMMA in YBCO-PST composite.

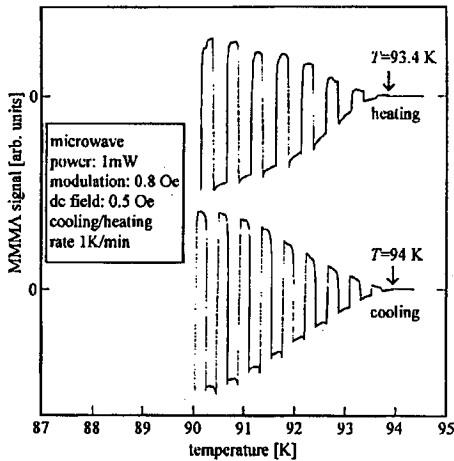


Fig. 4. The MMMA signal received using the rectangular modulation of the external magnetic field used to increase the accuracy of the determination of  $T_c$  using the microwave methodology.

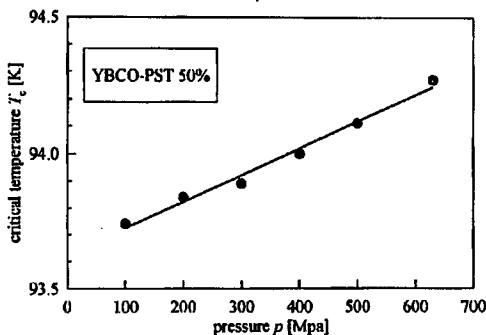


Fig. 5. Pressure dependence of  $T_c$  in YBCO-PST 50 weight-% composite.

dependence of the MMMA signal is shown in Fig. 2. One can observe the MMMA signal below the critical temperature, with a remanent non-zero value down to the lowest temperature. The magnetic field dependence of the MMMA signal is shown in Fig. 3. To increase the accuracy of the measurements, an additional rectangular modulation of the external magnetic field was applied. In this manner, instead of a monotonic change of the MMMA signal versus temperature, a step-wise, easy to identify signal can be observed (Fig. 4). Critical temperature was measured in the pressure range up to 650 MPa. The pressure dependence of  $T_c$  measured in YBCO-PST composite appears to be linear and its slope  $dT_c/dp$  equals  $1.0 \pm 0.1$  K/GPa (Fig. 5). This value is identical to the one obtained for pure YBCO sample using the same methodology [3] and is close to the  $dT_c/dp = 0.8$  K/GPa obtained by Maple et al. [14] in a wider range of pressures. The differences can be due to the different oxygen stoichiometry of the investigated samples. As shown by Medvedeva et al. [15], the pressure derivative  $dT_c/dp$  sharply increases as oxygen is removed from the fully oxygenated compound. Wühl et al. [16] presented the results of  $dT_c/dp$  investigation, which indicated a tenfold (from 0.3 to 3.0 K/GPa) change of the pressure factor for a small reduction in the oxygen content ( $O_{6.90}$  to  $O_{6.84}$ ), even though  $T_c$  only decreased by 1–2 K. It is consistent with a picture, in which increasing either pressure or the oxygen content is believed to increase the carrier density in the  $\text{CuO}_2$  planes [2]. In our case, the presence of PST in 50% in weight YBCO-PST composite, does not change the  $dT_c/dp$  factor, which indicates unchanged oxygen stoichiometry and structure of YBCO in the composite, compared to that one of pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .

The studies of MMMA allow to investigate the intergrain properties of the investigated material. The remanent low-temperature microwave absorption indicates an important role of intergrain weak links (Fig. 2). The microwave properties of the high temperature superconductors are determined by at least three different processes. Besides the dissipation due to the thermally excited carriers, described by Mattis and Bardeen [17], one can expect a dissipation introduced by the vortex nucleation and motion of the vortices in the mixed state, and dissipation due to the existence of inter- and intragrain weak links at low magnetic field. The microwave dissipation in ceramic high-temperature superconductors is significantly

higher than predicted by the Mattis–Bardeen model [18] and indicates the important role of intergrain weak links [19]. When the weak links play a dominant role in the microwave dissipation, one can investigate the low magnetic field-dependent microwave absorption by treating the sample as a network of resistively shunted junctions driven by a microwave small-signal current  $I_1 \cos \omega_1 t$  and a dc current  $I_0$  induced by an external dc magnetic field. The microwave power absorbed by the junction in linear regime is given by [20–22, 24]:

$$P = \frac{\frac{1}{2} I_1^2 R_n}{1 + [2e R_n I_c(\phi) \cos \varphi_0 / \hbar \omega]^2}, \quad (1)$$

where  $\varphi_0$  is an equilibrium phase in the absence of microwave field,  $R_n$  is a normal state resistivity of the junction and  $I_c(\phi)$  is a magnetic flux-dependent critical current of the junction.

The ceramic superconducting sample can be treated as a network of weak links and superconducting loops of different areas and orientations [23]. When the junctions do not interact with each other, the microwave power absorbed in the system of weak links can be described as a superposition originating from many weak links and loops of different areas and orientations [23].

Our previous high-pressure investigations of the microwave absorption in high temperature superconductors indicated a modification of the microwave absorption line shape under high pressure [3]. The observed previously effects indicate a change of the distribution of weak links and distribution of individual intergrain tunnelling currents under high pressure. High pressure can modify the distribution of intergrain weak links, which could be observed in the change of MMMA line-width versus temperature. On the other hand, the high-pressure influence on the intrinsic processes could be observed as the change of critical temperature.

Similar investigations of the MMMA signal in function of temperature or magnetic field for the YBCO–PST composites should allow us to determine the changes of the Josephson junctions critical current  $I_c$  and modification of their surface  $S_0$  caused by high pressure. However, our recent and unpublished results concerning the MMMA signal dependence on composition of the YBCO–PST superconductor show rather complex behaviour of the composites in a microwave field. It seems that an additional structural research is needed to explain the properties of the YBCO–PST composites. The results and the detailed discussion will soon be presented.

#### 4. Conclusion

To conclude, we have observed the remanent microwave absorption signal at low temperatures in YBCO–PST composites, which can be attributed to the presence of intergrain weak links. The microwave absorption measurements have been applied to monitor the superconducting transition under high pressure. The pressure factor  $dT_c/dp$  of YBCO–PST 50% in weight composite remains the same as in pure YBCO sample.

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