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Surface resistance of large $YBa_2Cu_3O_{7-x}$ crystals at 3 GHz with residual losses caused by flux compounds

D. Wehler, J. Dreibholz, M. Meyer, G. Müller, H. Piel and Th. Wolf *)
Bergische Universität Wuppertal, Fachbereich Physik, D-5600 Wuppertal 1
*)Kernforschungszentrum Karlsruhe, Institut für Technische Physik, D-7500 Karlsruhe

ABSTRACT

The surface resistance (R_s) of $YBa_2Cu_3O_{7-x}$ (YBCO)- crystal platelets, up to 5x10 mm^2 in size, and the microwave losses of the main flux compounds CuO and $BaCuO_{2\pm x}$ were investigated at 3GHz. R_s of all crystals drops sharply at T_c by about 3 orders of magnitude. The residual surface resistance (R_{res}) of crystals grown by the self-flux method with flux residues on the surface is dominated by the microwave losses of $BaCuO_{2\pm x}$. These losses are orders of magnitude higher than the losses of CuO and were found to increase below 40K. From the accordingly corrected measurement results on large crystals, we can estimate an upper limit of $R_{res} \approx 40\mu\Omega$. With nearly flux-free but small YBCO crystals we have achieved rf field independent R_{res} values close to the measurement limit up to 100 Oe.

1. INTRODUCTION

High frequency measurements are a very sensitive tool for the investigation of the electronic properties of high temperature superconductors (HTSC) [1]. In contrast to polycrystalline materials and epitaxial films, which suffer from grain boundaries, lattice defects and/ or phase impurities, single crystals may provide information about intrinsic high frequency properties [2,3]. These will be essential for the ultimate microwave performance and possible applications of HTSC [4]. Especially, the high residual surface resistance (R_{res}) of HTSC should be overcome by high quality single crystals. Comparing different HTSC's, R_{res} is smaller for YBCO than for $Bi_2Sr_2CaCu_2O_{8+x}$ [5], but it is orders of magnitude higher than for classical superconductors [6]. Additionally, the investigation of the sources for R_{res} provides helpful information for the optimisation of the preparation process of single crystals as well as of epitaxial thin films.

Therefore, we have studied the microwave properties of YBCO crystal platelets grown by the self flux method by exposing them to the high frequency field of a superconducting cavity resonator described in the next chapter. Since large crystals enhance the measurement sensitivity [7], we present in the main chapter the temperature and field dependence of large contaminated and small clean crystals. To explain the anomalous residual losses measured for large crystals, the microwave losses of typical flux compounds have also been studied.

2. EXPERIMENTAL TECHNIQUES

We have investigated large YBCO crystal platelets up to 50mm^2 in area and 0.2mm thick. The YBCO-crystals were grown from a nonstoichiometric melt of 4at% Y, 30at% Ba and 66at% Cu in different crucibles by slow cooling from 990°C-1010°C to 970°C with cooling rates of 0.1°C/h to 0.5°C/h. At this temperature the melt is soaked up by a porous ceramic. The oxygenation was carried out in flowing oxygen between 600°C -350°C for 3 weeks. The highest T_c-values of about 90K and the smallest transition width were achieved by crystals grown in SnO crucibles. Many of

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these crystals, especially the large ones, are contaminated with residual flux. EDX analysis of these residues showed that they consist mainly of CuO and $BaCuO_{2\pm x}$. Therefore, nearly pure samples of these compounds were separated from the melt in order to investigate their microwave losses.

To have a microwave test system with very low losses, we use a superconducting niobium resonator as a host cavity (Fig.1). The sample is placed in a high magnetic field region of the resonator by a sapphire rod with low dielectric losses, which can be heated separately from the cavity walls. The spherically shaped resonator is operated in a TM_{010}-like mode at f=2.9 GHz. The microwave power is coupled into the resonator by an antenna with variable position, allowing adjustment for optimal coupling conditions for every case. The loaded quality factor Q_L has been determined from the time constant τ of the cavity by $Q_L = 2\pi f\tau$. The overall unloaded residual Q_0 of the test system is $1.5*10^9$ at 1.9K.

The surface resistance R_s of the samples has been determined by the usual cavity pertubation formalism [1]. The partial loss contribution which depend mainly on the size of the crystal platelets has been calibrated by measurements with samples of classical superconductors and stainless steel. However, we have not corrected for dielectric loss enhancement in the sapphire caused by the electric field pertubation due to the presence of the metallic sample. Therefore, the R_s values presented here represent upper limits for the real R_s of the YBCO crystals. The resulting measurement limit for R_s is about 100μΩ for typical sample size.

In case of the dielectric flux compound samples, the product of their relative permittivity ε_r and their loss tangent tanδ has been determined under the assumption that the electric field distribution is not changed by the sample, i.e. that the geometric factor F, which takes account for the field enhancement, equals 1.

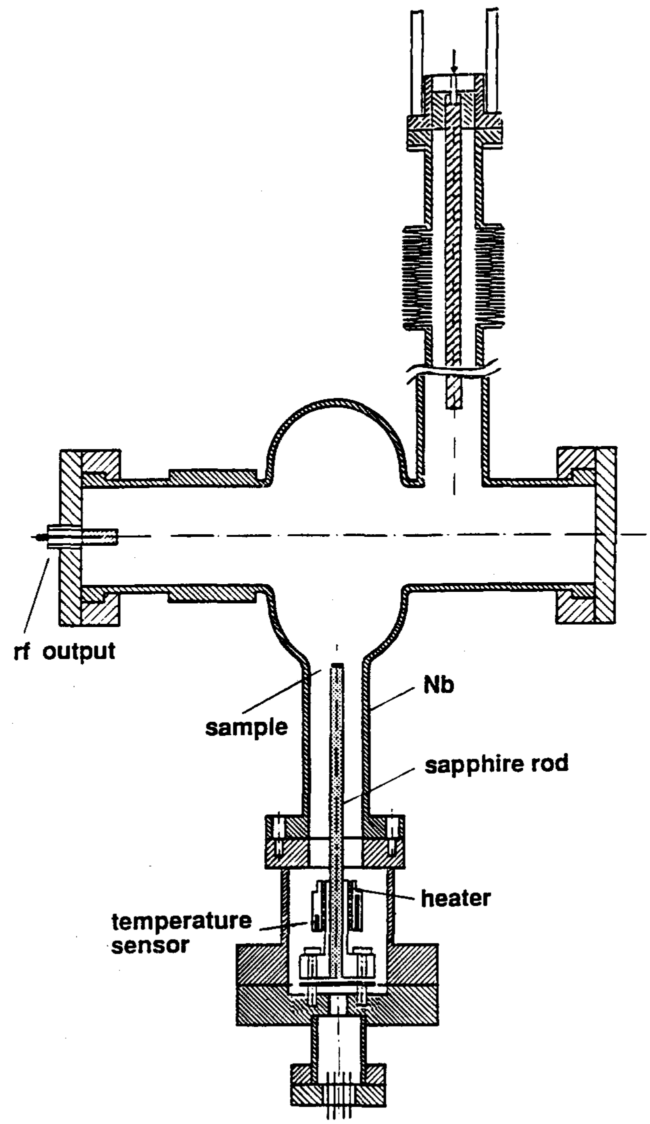

Figure 1. Schematic diagram of the 3GHz-niobium cavity. The sample is introduced by a sapphire into the high magnetic field region.

3. RESULTS AND DISCUSSION

The common feature of all YBCO-crystals is a sharp drop of R_s at T_c of about 3 decades within 3K (Fig2). The sharpness of the transition is comparable to that of the best epitaxial films measured at 87GHz [9], if the resistance in the normal conducting and superconducting regime is scaled with $\sqrt{f}$ and f^2, respectively. The temperature dependence of the real part of the conductivity agrees with calculations based on the strong coupling theory, with the assumption of a T_c-distribution of 0.5K [10].

The values of the specific resistivity ρ of the crystals derived by means of the normal skin effect formula are in the range of 50 to 250 µΩcm depending on the thickness of the crystals as with increasing thickness the contribution of ρ_c increases.

All crystals with flux contaminations show relatively high residual losses, which surprisingly increase at temperatures below 40K (Fig.2). To clarify the origin of these anomalous losses, we have investigated two nearly pure samples of the main flux compounds.

We found that $BaCuO_{2\pm x}$ which is an insulator at cryogenic temperatures [11] exhibits very high microwave losses (Fig3a). The temperature dependence of these losses agrees with that observed for the crystals. Therefore, we have substracted from the measured crystal data the fitted $BaCuO_{2\pm x}$-losses scaled by a factor corresponding to their volume fraction. The resulting $R_s(T)$-curve of the crystal provides a nearly temperature independent R_{res} as most epitaxial films [1]. However, the absolute value of 40µΩ is still more

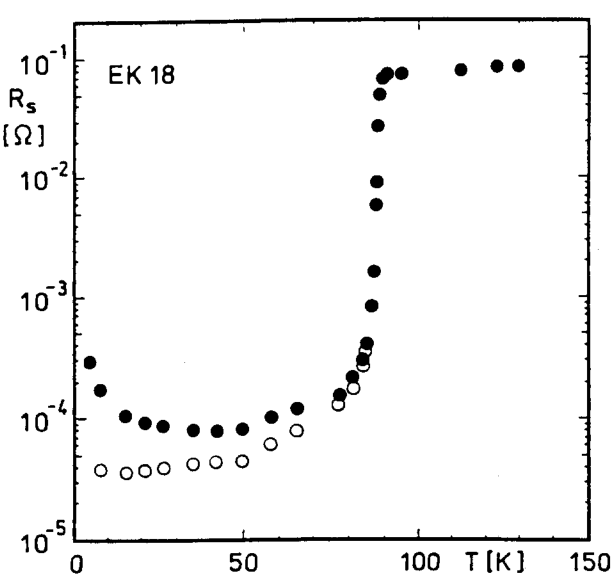

Figure 2. Temperature dependence of the surface resistance of a large (5x10mm^2) YBCO crystal. o= measured data; o= data after substraction of scaled $BaCuO_{2\pm x}$ losses.

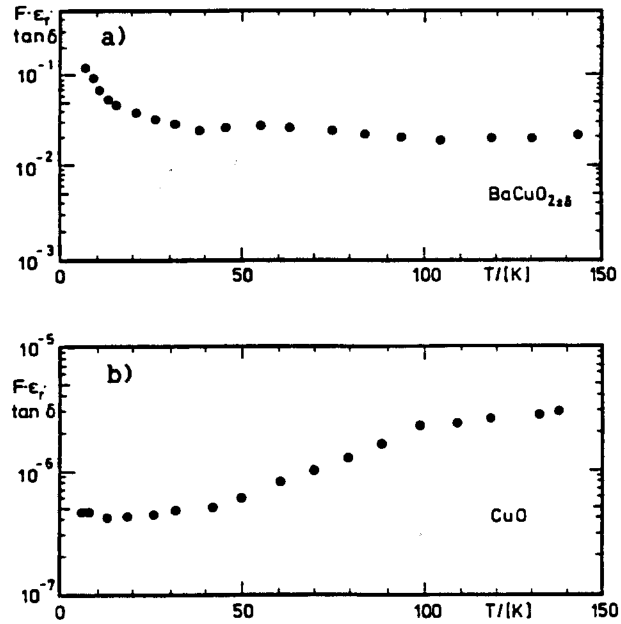

Figure 3. Microwave losses of a) $BaCuO_{2\pm x}$ and b) CuO as a function of T.

than one order of magnitude higher than the lowest measured R_{res} values of epitaxial films. At the present sensitivity limit, we cannot distinguish between the possible causes for this discrepancy, i.e. the influence of the electric field pertubation, of the side walls of the crystals and of other not corrected contaminations.

The losses of CuO show another temperature dependence (Fig.3b) and are found to be orders of magnitude smaller than the losses of $BaCuO_{2\pm x}$. This agrees with the observation that overlayers of CuO of considerable thickness do not degrade the microwave performance of epitaxial thin films [12]. For that reason, they can be neglected as major sources for R_{res} at the present level set by the $BaCuO_{2\pm x}$ losses and the measurement limit.

Crystals with clean surfaces have been of small size only, reaching surface areas not more than 2x4 mm^2. The R_s of these crystals falls just below T_c down to the measurement limit of about 100 µΩ. This is true up to surface magnetic fields of 100 Oe (Fig. 4). For the best crystals the

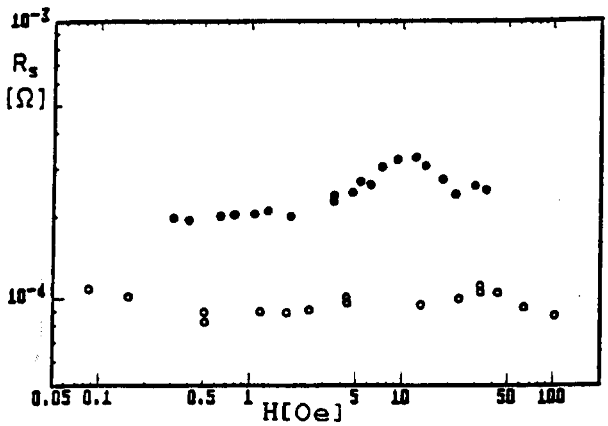

Figure 4. Dependence of the surface resistance on the surface magnetic field of (o) a contaminated YBCO crystal at 4.2 K, (o) a clean YBCO crystal at 1.8 K.

maximum achievable magnetic field (Hmax) is limited by the poor cooling conditions even in a pulsed mode operation. At H_{max} the dissipated power in the sample and the sapphire heats the sample up to temperatures above T_c during the time constant of the resonator. The highest magnetic field which we could achieve still in the superconducting state of one YBCO crystal was 225 Oe. Typical for crystals with flux residues is a local maximum in the $R_s(H)$ curve, which might be correlated to the partially inverse temperature dependence of $BaCuO_{2\pm x}$.

4. CONCLUSIONS

To obtain YBCO crystals with low microwave losses, it is essential to avoid contaminations of $BaCuO_{2\pm x}$. This material yields high microwave losses. It has been shown that the anomalous temperature dependence of the surface resistance of large ($5x10mm^2$) YBCO crystals are explainable by the losses of $BaCuO_{2\pm x}$ residues. CuO shows microwave losses orders of magnitude lower than $BaCuO_{2\pm x}$. The R_s of YBCO crystals drop sharply at T_c comparable to the best epitaxial films. Though we are using a superconducting niobium resonator as host cavity, lowest achieved R_{res} values of the YBCO crystals are close to the measurement limit. Therefore, it is necessary to enhance the ratio of resonator to sample surface area. For this purpose, a similar system operating at 21 GHz is under construction, which should reduce the measurement limit by a factor of 30.

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