

## PERFORMANCE STATUS OF HIGH- $T_C$ SUPERCONDUCTORS FOR CAVITY APPLICATIONS

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### Abstract

Systematic measurements of the surface resistance  $R_S$  of  $YBa_2Cu_3O_{7-\delta}$  samples between 3 and 90 GHz have given much higher residual losses than for classical superconductors but smaller ones than for copper. The lowest  $R_S$  values of about 10 m $\Omega$  at 77 K and 87 GHz have been obtained with epitaxially grown films on dielectric substrates, while single crystals have tolerated the highest field levels of more than 100 Oe at 4.2 K and 3 GHz. Polycrystalline samples provide sufficiently low  $R_S$  only at very low field levels. Their granularity can be partially suppressed by c-axis texturing of the grains in high magnetic fields. We have developed an electrophoretic deposition technique which allows to coat arbitrary shaped silver cavities with textured  $YBa_2Cu_3O_{7-\delta}$  layers. As a first application, a compact hydrogen maser cavity was coated with untextured  $YBa_2Cu_3O_{7-\delta}$  yielding  $R_S$  values of 1 m $\Omega$  at 77 K and 1.42 GHz which should be low enough for a successful maser operation.

### Introduction

Superconducting cavities built from high purity niobium have become one of the major technical improvements for many new accelerator projects [1] due to the enlarged field gradients they provide to cw particle beams. Moreover, their routinely achievable unloaded quality factor  $Q_0$  of more than  $10^9$  is about five orders of magnitude higher than for copper cavities and leads to reduced energy consumption as well as to new applications for fundamental physics experiments [2]. The substitution of niobium by high- $T_C$  superconductors promises much higher accelerating fields or at least higher operating temperatures for such cavity applications [3, 4]. From an economic point of view,  $Q_0$  values of about  $10^8$  at 77 K corresponding to a surface resistance  $R_S$  of about 3  $\mu\Omega$  would be sufficient due to the improved Carnot and technical efficiency of the refrigerators. The scope of new applications especially for space technology basically depends on reduced microwave losses of high- $T_C$  superconductors in comparison to copper at temperatures well above 20 K.

Since the encouraging first results on the microwave properties of the high- $T_C$  superconductors reported at the EPAC two years ago [5], many measurements around the world have revealed mainly two difficulties which hinder the rapid use of these materials for cavity applications [6]. First, anomalously high residual losses are observed especially at high field levels. Secondly, these losses are even more pronounced for polycrystalline material which will be necessary for large and curved surfaces. Therefore, we have splitted our efforts into two opposite directions. One deals with the investigation of the best available single crystals and epitaxially grown thin films to uncover the intrinsic properties of the high- $T_C$  superconductors. The other tries to optimize the performance of bulk ceramic and electrophoretically deposited layers on silver substrates. The latter technique is well suited for the construction of cavities of complex shape and provides improved mechanical as well as thermal stability for these rather brittle and poor conducting oxides. In this report, the progress achieved for  $YBa_2Cu_3O_{7-\delta}$  as the present material of choice will be summarized with respect to both issues.

### Experimental Techniques

Four different kinds of  $YBa_2Cu_3O_{7-\delta}$  samples were systematically investigated concerning their microwave properties. Polycrystalline bulk pellets and thick films on Ag substrates were produced at Wuppertal, while epitaxially grown thin films on dielectric  $SrTiO_3$  and  $LaAlO_3$  substrates and single crystals were supplied by Siemens Research Laboratories in Erlangen and Kernforschungszentrum Karlsruhe, respectively. All polycrystalline samples were synthesized from high purity oxides and carbonates by solid state reaction as described in more detail elsewhere [5, 6]. The superconductor powder was either pressed to pellets of 13 or 25 mm diameter and 1 to 2 mm thickness or suspended preferably in Acetone for the electrophoretic deposition process. The coating was usually performed in four steps with short intermediate bakings resulting in homogenous layers of about 20  $\mu m$  thickness. Applying a magnetic field  $H$  of 8 T during the deposition, a high degree of c-axis texturing parallel to  $H$  can be achieved in such layers [7]. Long sintering periods of more than 100 h have been applied to all of these polycrystalline samples to ensure sufficient grain growth. The thin films ( $d \approx 0.8 \mu m$ ) were in situ grown on heated substrates of 1 cm<sup>2</sup> size by laser ablation of stoichiometric targets [8]. Single crystals typically a few mm in size were flux grown from Ba and Cu rich melts in  $ZrO_2$  crucibles and postannealed below 600°C in an oxygen atmosphere for at least one week [9].

The fabrication of all of these samples were optimized using X-ray diffractometry and scanning electron microscopy with energy dispersive X-ray analysis for the control of phase purity, crystallinity and homogeneity. For the here reported best samples, the inductively measured transition widths (10% to 90%) never exceeded 1 K. Zero dc resistance was always achieved at about 92 K except for the thin films which resulted in  $T_C$  values around 90 K depending on the substrate. The granularity of the samples, i.e. the quality of the grain contacts, was tested in dc magnetic fields up to 8 T resulting in reduced  $T_C$  values and enlarged transition widths especially for the untextured polycrystalline samples.

The microwave performance of the samples was tested in host cavities at four different frequency bands. Plane samples, i.e. pellets and thin films as well as thick layers on Ag discs, with a net diameter of at least 22 mm or 8 mm were mostly measured as endplates of cylindrical copper cavities in  $TE_{0np}$  modes at about 21 GHz or 87 GHz, respectively. For this well defined test arrangement with no currents across the contact surface, the sensitivity limit is given by the residual losses of the copper cavity [6]. Moreover, the maximum achievable magnetic surface field levels for the given microwave power of about 1 W at 21 GHz and 10 mW at 87 GHz are accordingly below 700 A/m and 20 A/m. Both the sensitivity and the field limits have been slightly increased for measurements at 4.2 K by use of niobium host cavities. An alternative measurement set-up which enables much higher field levels in arbitrary shaped samples at all temperatures by means of a niobium accelerator cavity at 3 GHz is shown in Fig. 1. This technique is based on an independent temperature control of the sample on top of a thermally insulated sapphire support. Because of the positioning of the sample in the maximum magnetic field region, fields up to typically 40000 A/m ( $\approx 500$  Oe) can

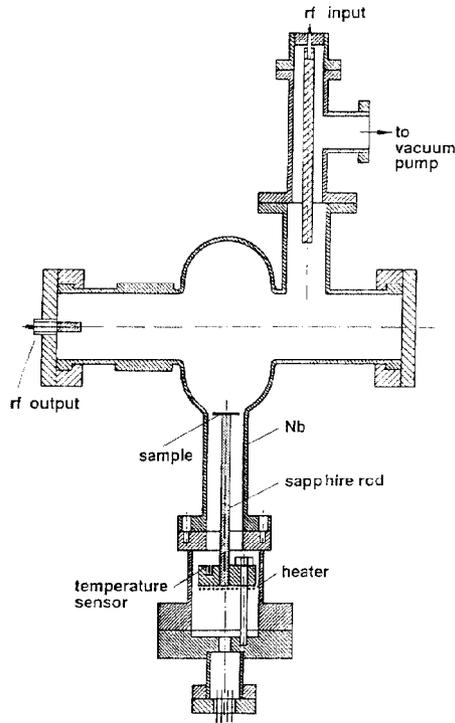


Fig. 1: Spherical Nb cavity for the test of small samples on an independently heated sapphire rod in the maximum magnetic field region of the  $TM_{010}$  mode at 3 GHz.

be applied. This limitation is set by the breakdown of the superconducting host cavity. However, sufficient cooling of the sample has to be provided either by He gas pressure of 100 mbar (at 4.2 K) or by pulsed power with a short duty cycle. At low fields, the sensitivity limit for direct surface impedance measurements mainly depends on the size of the samples and on the residual losses given by the sapphire rod. Moreover,  $R_S$  can be estimated from calorimetric measurements with the temperature sensor inside the heating system. Finally, the inner cylindrically shaped electrodes of a compact hydrogen maser cavity [10] have been electrophoretically coated and tested at 1.4 GHz. At all of these frequencies,  $R_S$  was measured as a function of temperature between 4.2 and 300 K as described in more detail elsewhere [4].

### Results and Discussion

The sharpest microwave transition curves for  $YBa_2Cu_3O_{7-\delta}$  have been obtained with single crystals at 3 GHz (Fig. 2) and 6 GHz [11] and with epitaxially grown thin films on  $LaAlO_3$  at 6 and 10 GHz [12] and 87 GHz [13]. On a reduced temperature scale, they are comparably sharp as those for classical superconductors like  $Nb_3Sn$  (see Fig. 2), i.e. as expected from BCS theory. In contrast, polycrystalline bulk samples yield a more gradual decrease of  $R_S(T)$  down to 4.2 K as also shown in Fig. 2. The same is true for untextured layers on the electrodes of the maser cavity which resulted at 1.4 GHz in  $R_S$  values of 1 m $\Omega$  at 77 K and 50  $\mu\Omega$  at 4.2 K [10]. Systematic test series on polycrystalline bulk and layer samples at 21.5 GHz have confirmed the important role of a homogenous microstructure [5] and of an improved crystallinity due to c-axis texturing [7] for steeper fall-offs near  $T_C$ . It is most remarkable that all kinds of oxide superconductors provide several orders of magnitude higher residual losses than Nb or  $Nb_3Sn$ . For both the best polycrystalline as well as single crystalline  $YBa_2Cu_3O_{7-\delta}$  samples, they increase about quadratically with frequency but are about a factor of 30 lower for the latter

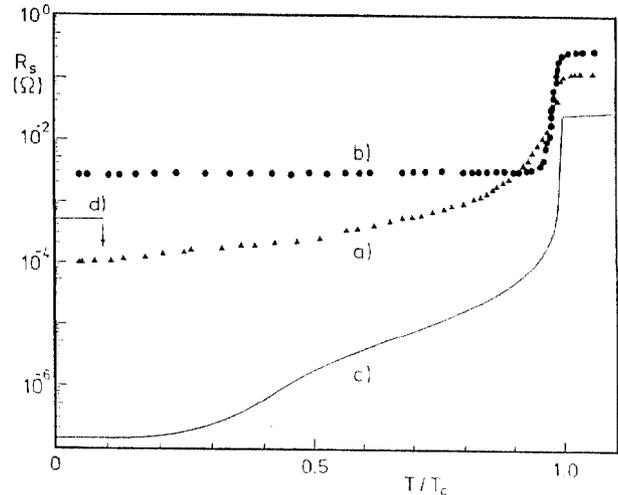


Fig. 2: Typical temperature dependencies of  $R_S$  measured at 3 GHz for a polycrystalline pellet (a) and a single crystal of  $YBa_2Cu_3O_{7-\delta}$  (b) both with  $T_C = 92$  K. Also shown are data for polycrystalline  $Nb_3Sn$  layers (c) and for larger single crystals of  $YBa_2Cu_3O_{7-\delta}$  (d).

ones [6]. The difference is somewhat smaller at 77 K, where the lowest values approach already the  $R_S(\omega)$  data of Nb and  $Nb_3Sn$  at the same reduced temperature. From a more practical point of view, the comparison of the present data for  $YBa_2Cu_3O_{7-\delta}$  with  $R_S(\omega)$  of OFHC copper at 77 K yield crossover frequencies of about 20 GHz for polycrystalline and more than 300 GHz for single crystalline material.

For the application of oxide superconductors in accelerator cavities, however, such low  $R_S$  values have to be achieved also at high field levels. This requirement seems to be a much bigger obstacle especially for polycrystalline  $YBa_2Cu_3O_{7-\delta}$  as demonstrated in Fig. 3. Even the best bulk samples show a drastic increase of losses already at magnetic surface fields  $H_S$  below 100 A/m which in a typical accelerating structure correspond to field gradients far below 0.1 MV/m. However, the situation is not hopeless since single crystalline material behaves much different in that respect. For the single crystal data in Fig. 3,  $R_S$  stays nearly stable at 4.2 K as well as at 77 K up to  $H_S$  values of at least 10000 A/m ( $\approx 120$  Oe, or about 3 MV/m for electron accelerators). Similar results have been previously obtained at 6 GHz for single crystals [11] as well as for two thin films on  $LaAlO_3$  substrates [14]. It is

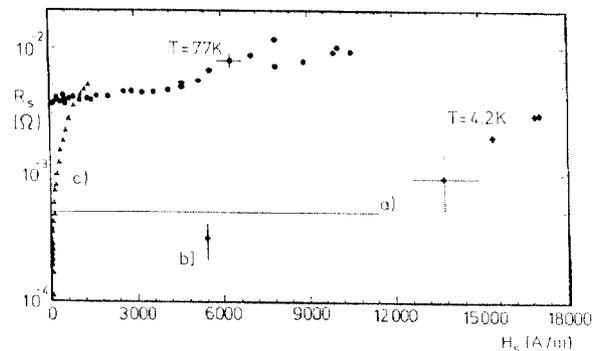


Fig. 3: Dependence of  $R_S$  on the magnetic surface field  $H_S$  at 3 GHz for the best polycrystalline pellet at 4.2 K (triangles) and a single crystal of  $YBa_2Cu_3O_{7-\delta}$  at 4.2 K (rhombes) and 77 K (circles). At 4.2 K and low  $H_S$ , the single crystal losses fall below the sensitivity limit of the microwave measurements (line a) but can be estimated calorimetrically (point b).

worthwhile to mention that for the test arrangement shown in Fig.1 the surface currents have to flow in all directions of the crystals, i.e. also across the planes of these strongly anisotropic superconductors. Preliminary measurements at 3 GHz for a thin film on LaAlO<sub>3</sub> provide a R<sub>S</sub> degradation from 0.6 mΩ at 1 A/m to a saturation value of 7 mΩ between 100 A/m and 2000 A/m. Similar field dependencies have been observed for directly cooled polycrystalline wire samples at frequencies below 1 GHz, large portions of which remained superconducting up to the highest fields of 50000 A/m [15]. All of these results suggest that the intrinsic microwave field limitation of the oxide superconductors is not reached yet, and that the increase of losses is caused by the granularity especially for polycrystalline samples.

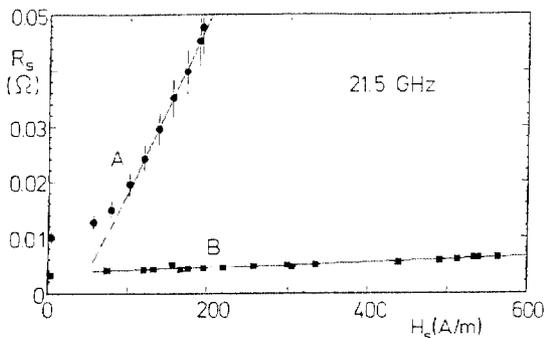


Fig. 4: Typical H<sub>S</sub> dependence of R<sub>S</sub>(4.2 K) measured at 21.5 GHz for electrophoretically deposited untextured (circles) and c-axis textured (squares) YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> layers.

At this point the question arises if and how far the high field performance of polycrystalline YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> can be improved by c-axis texturing of electrophoretically deposited layers. The results shown in Fig. 4 are rather encouraging in that respect, since R<sub>S</sub> stays nearly constant for the textured layer up to the maximum available microwave power. Further systematic test series on such layers have revealed a strong correlation between their sensitivity against high magnetic surface fields expressed by the linear slopes dR<sub>S</sub>/dH<sub>S</sub> at 4.2 K and the absolute value of R<sub>S</sub> at 77 K (Fig. 5a). Moreover, R<sub>S</sub>(77 K) decreases not only for the c-axis textured layers but also for bulk samples with enhanced grain growth due to improved sintering conditions [10] as shown in Fig. 5b. Therefore, reducing the number of grain boundaries and enhancing the grain contacts seems to be the key for further improvements of polycrystalline YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>.

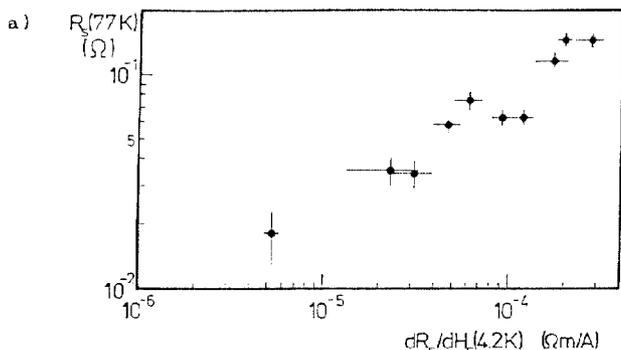
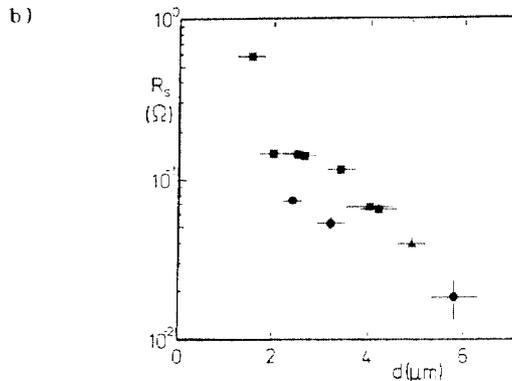


Fig. 5: Correlation between R<sub>S</sub>(77 K) measured at 21.5 GHz and a) the linear slopes dR<sub>S</sub>/dH<sub>S</sub> at high fields and 4.2 K (see Fig. 4); b) the average grain size d of several untextured (squares) and c-axis textured layers (circles) and a bulk sample (triangle).



Conclusions

Oxide superconductors provide not only high T<sub>C</sub> values but also many difficulties to reach as low R<sub>S</sub> values and as high levels as classical superconductors. Nevertheless, for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> there are already some promising results for cavity applications at least at low field levels. Suppression of the granularity of these materials by further refinements of the fabrication techniques will be necessary to make their excellent intrinsic properties accessible for more applications.

Acknowledgements

We gratefully acknowledge the collaboration with T. Wolf from the Kernforschungszentrum Karlsruhe and with B. Roas and L. Schultz from the Siemens Research Laboratories for providing us with high quality single crystals and thin films, respectively. The contributions of H. Chaloupka, E. Mahner, M. Meyer and R.W. Röth are appreciated. This work was funded by the German Federal Minister for Research and Technology (BMFT) under the contract number 13 N 5502.

References

- [1] For recent status reports see Proc. of the 4th Workshop on RF Superconductivity, Tsukuba, ed. by Y. Kojima, KEK Report 89-21 (1990).
- [2] N. Klein, G. Müller, H. Piel and J. Schurr, IEEE Trans. Magn. **MAG-25**, 1362 (1989).
- [3] H. Padamsee, J. Superconduct. **1**, 377 (1988).
- [4] H. Piel, Nucl. Instr. Meth. in Phys. Res. **A287**, 294 (1990).
- [5] G. Müller, M. Hein, N. Klein, H. Piel, L. Ponto, U. Klein and M. Peiniger, Proc. of the EPAC, Rom, p. 40 (1988).
- [6] For a recent review see G. Müller, *ibid.* ref. 1, p. 267 (1990).
- [7] M. Hein, G. Müller, H. Piel, L. Ponto, M. Becks, U. Klein and M. Peiniger, J. Appl. Phys. **66**, 5940 (1989).
- [8] B. Roas, L. Schultz and G. Endres, Appl. Phys. Lett. **53**, 1557 (1988).
- [9] T. Wolf et al., J. Crys. Growth **96**, 1010 (1989).
- [10] M. Hein, S. Kraut, E. Mahner, G. Müller, D. Opie, H. Piel, L. Ponto, D. Wehler, M. Becks, U. Klein and M. Peiniger, *subm. to J. Superconduct.* (1990).
- [11] D.L. Rubin, K. Green, J. Gruschus, J. Kirchgessner, D. Moffat, H. Padamsee, J. Sears, Q.S. Shu, L.F. Schneemeyer and J.V. Waszczak, Phys. Rev. **B38**, 6538 (1988).
- [12] A. Inam, X.D. Wu, L. Nazar, M.S. Hedge, C.T. Rogers, T. Venkatesan, R.W. Simon, K. Daly, H. Padamsee, J. Kirchgessner, D. Moffat, D. Rubin, Q.S. Shu, D. Kalokitis, A. Fathy, V. Pendrick, R. Brown, B. Brycki, E. Belohoubek, L. Drabeck, G. Grüner, R. Hammond, F. Gamble, B.M. Lairson and J.C. Bravman, Appl Phys. Lett. **56**, 1178 (1990).
- [13] N. Klein, H. Chaloupka, G. Müller, S. Orbach, H. Piel, B. Roas, L. Schultz, U. Klein and M. Peiniger, J. Appl. Phys., June (1990).
- [14] H. Padamsee, J. Kirchgessner, D. Moffat, D.L. Rubin, Q.S. Shu, A. Inam, X.D. Wu, L. Nazar, M.S. Hedge, T. Venkatesan, Cornell University, CLNS 89/929 (1989).
- [15] J.R. Delaven and C.L. Bohn, Phys. Rev. **B40**, 5151 (1989).