G. Müller, M. Hein, N. Klein, H. Piel and L. Ponto Fachbereich Physik, Bergische Universität-Gesamthochschule Wuppertal, D-5600 Wuppertal 1, FRG

U. Klein and M. Peiniger

Interatom GmbH, D-5060 Bergisch-Gladbach 1, FRG

Abstract

The potential application of the new high T_c superconductors for low loss accelerator cavities at liquid nitrogen temperatures depends on their residual surface resistance and high field performance as well as on the development of appropriate coating techniques. We have exposed polycrystalline bulk samples and thick layers of Y1Ba2Cu3O7 to the microwave field of normal and superconducting host cavities at S- and K-band frequencies. Increasing the sintering time of the samples to more than 200 hours, residual surface resistances of less than 200 $\mu\Omega$ at 3 GHz and 20 m Ω at 21.5 GHz have been obtained which are already significantly lower than that of pure copper. Further improvements of the microscopic stoichiometry of the bulk samples resulted in rf transitions as narrow as those of single crystals, providing a surface resistance of about 30 m Ω at 77 K and 21.5 GHz. First results on electrophoretically deposited layers on silver substrates are promising for the fabrication of completely coated cavities of arbitrary shape.

Introduction

Since the discovery of the oxide superconductors [1,2], their impact on present applications of conventional superconductors in accelerators, i.e. magnets and rf cavities, is under discussion. For applications the most promising of the new superconductors is the oxygen deficient perovskite $Y_1Ba_2Cu_3O_7$. Its easy fabrication as well as its high critical temperature $T_c=93$ K makes this ceramic like material most interesting for rf applications because of three reasons.

At first, layers of only some μm thickness are necessary due to the small penetration depth λ of the rf magnetic surface field H into the superconductor. The temperature dependence of λ follows the empirical law

$$\lambda(T) = \lambda(0) / \sqrt{1 - (T/T_c)^4}$$
⁽¹⁾

with $\lambda(0) \approx 0.1 \,\mu\text{m}$ for typical superconductors. Therefore, only thin superconducting coatings are required which can be deposited on mechanically rigid substrate cavities

Secondly, due to the presence of unpaired conduction electrons rf losses don't vanish just below T_c . The rf power P absorbed by these electrons produces Joule heating which is proportional to the surface resistance R_s of the superconductor. Its frequency and temperature dependence can be described according to the BCS theory for conventional superconductors like Nb and Nb₃Sn in good approximation by

$$R_{s}(\omega,T) = 2P/H^{2} = A\omega^{2}/T e^{-\Delta(T)/(kT_{c}) \cdot T_{c}/T}$$
 (2)

where the reduced energy gap of the superconductor $\Delta/kT_{\rm C}$ is nearly constant for $T \leq T_{\rm C}/2$. Depending on the ratio $\hbar\omega/2\Delta$, $R_{\rm S}(T)$ falls just below $T_{\rm C}$ several orders of magnitude due to the drastic reduction of $\lambda(T)$ (1) and the increase of $\Delta(T)$. Below $T_{\rm C}/2$ the exponential temperature dependence governs until an additive residual surface resistance dominates the rf losses at very low temperatures. Therefore, the higher the $T_{\rm C}$ and $\Delta/kT_{\rm C}$ of a superconductor the higher the operating temperature of an cryogenic accelerator can be chosen.

The third reason arises from the anisotropic nature of $Y_1Ba_2Cu_3O_7$ and from the granularity of polycrystalline material which causes low critical current densities between single grains [3]. While this feature sets a very hard constraint for the construction of dc and ac wires and magnets [4], capacitive rf coupling between the grains could be possible without additional losses. There is no doubt that the critical current density in single crystals exceeds 10^{11} A/m² corresponding to peak magnetic surface fields of at least 10^4 A/m (=12 mT) for an assumed λ of 0.1 μ m. From a more fundamental point of view the critical rf magnetic field should be given by the superheating field which is about $0.75 \cdot H_c$ for superconductors with large λ compared to the coherence length [5]. Literature data of H_c provide at least 1T for Y₁Ba₂Cu₃O₇ which would correspond to accelerating gradients of about 200 MV/m in velocity of light structures.

Because of these reasons, we have started to investigate the rf properties of polycristalline $Y_1Ba_2Cu_3O_7$ samples [6] using normal and superconducting host cavities at S- and K-band frequencies [7]. Moreover we have tried to improve the rf performance by a systematic variation of the fabrication techniques [8]. Last but not least we have developed an electrophoretic deposition technique adequate for the coating of arbitrary shaped cavities [9]. In this paper the main results on the surface resistance and high field performance of bulk samples and thick layers will be presented and compared to data obtained recently on large single crystals [10].

Sample Fabrication and Characterization

Stoichiometric powder mixtures of high purity ($\geq 99.99\%$) Y₂O₃, BaCO₃ and CuO were extensively ball milled in agate devices resulting in a typical particle size between 1 and 10 µm. Calcination was performed at 930°C in air for at least 24 hours partially with repeated regrindings. From the resulting pulverized Y₁Ba₂Cu₃O_{7- δ} pellets of 13 and 25 mm diameter and 1 to 3 mm thickness were pressed with 7 kbar. Alternatively colloidal suspensions with organic solvents were used for the electrophoretic deposition of Y₁Ba₂Cu₃O_{7- δ} onto silver plates [9] of 25 and 125 mm diameter. After five deposition steps with short intermediate baking smooth black layers of about 50 µm thickness were obtained. Finally these samples were sintered at 930°C in a pure oxygen atmosphere for extented time periods up to 10 days and slowly cooled down to room temperature.

Scanning electron microscopy with energy dispersive X-ray analysis was used to judge about the density and homogeneity of typical samples. The electrophoretically deposited layers appeared to be much denser than the bulk samples. Furthermore sufficient layer adhesion but no silver diffusion was recognized. Despite of the correct overall stoichiometry, local deviations from the nominal composition were found especially at the grain boundaries. The dc resistivity of some bulk samples was controlled by the standard four wire technique providing always transition temperatures and widths around 92 K and 1 K, respectively. Magnetic shielding measurements resulted in ac transition widths of less than 1 K for the best samples. X-ray diffractometry provided within its limited resolution (1%) for all samples single-phase $Y_1Ba_2Cu_3O_7$.

RF Measurement Techniques

The rf properties of the samples were tested in different normal and superconducting host cavities. The change of the unloaded quality factor of these cavities $Q_0^{\ C}$ due to the $Y_1Ba_2Cu_3O_7$ samples to $Q_0^{\ C,S}$ allows the determination of their surface resistance according to the formula [7]

$$\mathbf{R}_{s} = \mathbf{G}_{s} \left(\frac{1}{Q_{0}^{c,s}} - \frac{1}{Q_{0}^{c}} \right) + \mathbf{R}_{w}$$
(3)

where R_w is the measured surface resistance of the empty cavity. G_s can be either calculated from the geometry of the sample and the cavity field distribution or calibrated by an identical sample of known surface resistance. Copper cavities are well suited to resolve $R_s(T)$ of $Y_1Ba_2Cu_3O_7$ as long as it is significantly higher or not too much lower from that of copper itself. However, for the measurement of extremely low residual surface resistances and high rf field levels superconducting host cavities must be used. As a first step we have constructed two cylindrical copper cavities for different frequency bands as shown in Figs. 1 and 2 which are optimized for the TE_{011} mode and our sample dimensions. For the samples of 13 mm diameter a superconducting niobium cavity at 3 GHz was used as described previously [8].



Fig. 1: Cylindrical copper cavity for testing large $Y_1Ba_2Cu_3O_7$ samples and layers in the TE_{011} mode at 3.5 GHz.



Fig. 2: Same as Fig. 1 but for 21.5 GHz using waveguides.

Both cavities are supplied with mode traps to separate the TE_{011} from the degenerate TM_{111} mode sufficiently. Due to the very slow warm up of the cavities in helium cryostats within one day from 4.2 K to about 150 K, the temperature of the samples can be measured with sufficient accuracy by platinum resistors on the cavity wall. These $R_s(T)$ measurements were performed with a computer controlled system based on a synthesized sweep oscillator with 1 Hz step width.

Results and Discussion

In Fig.3 the residual surface resistance $R_s(4.2 \text{ K})$ measured on the bulk samples at 3 GHz (13 mm) and 21.5 GHz (25 mm) is shown as a function of the total sintering time, i.e. of the powder and the pellets. At both frequencies we have found a correlation $R_s(4.2 \text{ K}) = R(f) \cdot t(h)^{-\alpha(f)}$ with $R(3 \text{ GHz}) = 8.93 \Omega$ $\pm 0.15 \Omega$, $\alpha(3 \text{ GHz}) = 0.702 \pm .007$, $R(21.5 \text{ GHz}) = (3.5 \pm 1.2) \cdot 10^3 \Omega$ and $\alpha(21.5 \text{ GHz}) = 2.13 \pm .06$ which seem to be also valid for electrophoretically deposited layers from the same powder. Obviously a limit is not obtained yet, but longer annealing times are disadvantageous for applications.



Fig. 3: Dependence of the residual surface resistance at 4.2 K on the total annealing time of the $Y_1Ba_2Cu_3O_7$ bulk samples for 3 and 21.5 GHz.

The measured temperature dependence of R_s is influenced strongly by the preparation conditions of the samples too, as shown in Fig. 4 and 5. All curves exhibit the same transition temperature of about 92 K. For sample M21 the fall off has become gradually steeper with decreasing residual surface resistance which is still temperature dependent even at 4.2 K. Nevertheless the initial drop of $R_s(T)$ for this sample was much weaker than expected from (2) even after the longest sintering time. Therefore we have modified our fabrication technique by regrinding steps during the calcination. For the first time we achieved with this polycrystalline sample (M26) already after a relative short total sintering time of 120 h a much steeper $R_s(T)$ just below T_c which is nearly saturated at 77 K (Fig. 4). Its relative high residual R_s at 4.2 K was systematically improved by longer sintering as for the other samples. This clearly demonstrates that the fall off and the residual value of R_s are not correlated. This is also true for conventional superconductors where non superconducting surface contaminations dominate R_{res}.



Fig. 4: Temperature dependence of the surface resistance R_s at 21.5 GHz for different $Y_1Ba_2Cu_3O_7$ bulk samples. In the brackets the sintering time of the powder and the pellets are given.



Fig. 5: $R_s(T)$ for an electrophoretically deposited $Y_1Ba_2Cu_3O_7$ twice sintered layer at 21.5 GHz. In addition the first result on a large (130 cm²) plate at 3.5 GHz is given.

These results support our assumption that the residual losses of our polycrystalline samples originate at least partially from remaining surface corrosion or intergrain resistivity while the fall off seems to be influenced mostly by the microscopic stoichiometry. At this point a comparison with the best available single crystal data obtained at 6 GHz [10] is most interesting. In Fig.6 the actualized data of $R_{c}(77 \text{ K})$ for our polycrystalline samples as well as for the single crystals are plotted as a function of the frequency. The fall off of $R_{s}(T)$ is similar steep now for both, but single crystals provide a somewhat reduced T_c due to oxygen deficiency. It is worthwhile to mention that their performance scatters for different batches too. The arrows in Fig. 6 on the best values remind to the fact that the real surface resistance of superconducting Y₁Ba₂Cu₃O₇ should be even lower because of the high residual losses in both cases.



Fig. 6: Frequency dependence of R_s at 77 K for the different bulk samples and layers. For comparison the former status at various laboratories [7] (straight line) and recent data on single crystals [10] are given. The dashed line corresponds to copper at cryogenic temperatures.

Finally, the high field performance of superconducting $Y_1Ba_2Cu_3O_7$ will decide over its application in particle accelerators. We have found no degradation of $R_s(4.2 \text{ K})$ up to magnetic surface fields of about 100 A/m in polycrystalline samples. For higher field levels which are at present available only for the S-band cavities, a significant increase of losses has been observed in the 13 mm diameter samples [7] which have not been fabricated so far with the modified technique. It is promising that at least single crystals have shown no strong field dependence of rf losses at 20 K up to 9 mT, i.e. close to 10^4 A/m [11].

Conclusions

First measurements on polycristalline bulk samples and electrophoretically deposited layers of $Y_1Ba_2Cu_3O_7$ have been performed at S- and K-band frequencies. Surface resistance values significantly below those of copper have been obtained at low temperatures. The temperature dependence of R_s has been found to be for the best bulk samples as steep as for single crystals which is most important for operation at 77 K. Compared to the conventional superconductors this high T_c superconductor provides moderate Q and field values until now. Nevertheless the significant improvements achieved systematically during the first year after its discovery are promising for the future work and possible application of oxide superconductors for accelerating cavities.

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