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# Microwave surface resistance of bulk $YBa_2Cu_3O_{6+x}$ material

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Superconducting Y-Ba-Cu-O samples were prepared by conventional solid-state reaction. The microwave surface resistance of 1:2:3 compound superconductor material was measured in a special disk resonator structure at 10 GHz. At liquid-nitrogen temperatures, the microwave surface resistance is comparable to that of Au. At lower temperatures ($\sim 10$ K), the surface resistance is an order of magnitude lower than that of Au at the same temperature.

## INTRODUCTION

Since the observation by Müller and Bednorz that an oxide of barium, lanthanum, and copper might be superconducting at temperatures up to 35 K, many groups have pushed superconducting transition temperatures in this class of copper oxide materials, especially the Y-Ba-Cu-O group with the 1:2:3 layered perovskite superstructure, to 90 K and slightly above.

Previous superconductor applications were made from alloys of lead or niobium. Since their $T_c$ is low, they will operate at temperatures only a few degrees above absolute zero. These temperatures could only be achieved by employing expensive refrigeration units using liquid He as the coolant. However, the recently discovered high-$T_c$ ceramic superconducting materials with operating temperatures around 90 K and slightly above promise to reduce the cooling requirements tremendously and permit the use of cheaper refrigerants such as liquid nitrogen.

Low-$T_c$ superconductors found early applications in the field of microwave and millimeter wave devices. For some materials, ultralow loss factors at frequencies well beyond microwave frequencies have been demonstrated. This superior low-loss performance permits orders of magnitude improvement in conventional devices such as filters, high $Q$-stabilized cavities, and delay lines. Superconductors also have demonstrated parametric and Josephson phenomena which led to a number of new devices and applications for the generation, amplification, and detection of microwave radiation.

As the interest focuses now on high-$T_c$ applications in the microwave area, material properties such as maximum current density, sheet resistance as a function of frequency and temperature, effects of surface finish, porosity, possible anisotropies, moisture absorption, passivation, annealing, and thermal expansion have to be characterized and understood. In this paper, we present some early results on the microwave surface resistance of 1:2:3 compound. Our initial effort was concentrated on bulk material in disk form as it is easy to fabricate. While from a long-range point of view we are convinced that superconducting thin-film configurations have the broadest application possibilities, a wealth of important information can be derived from the current studies of the relatively porous, solid disk samples.

There are different methods to measure the microwave surface resistance of this new material. Aside from the indirect qualitative measurement techniques like microwave absorption measurements,[1] there are perturbation and direct measurement schemes. The superconducting material in the perturbation technique is only a small portion of the microwave resonant structure.[2,3] Meanwhile, in direct measurements, the whole resonator or the high-current carrying parts of the resonator are made from superconducting material.[4] In general, it is difficult and elaborate to judge quantitatively the true conductivity improvement of the ceramic material using perturbation techniques. Hence, it is preferred to use direct measurement techniques.

The microwave measurements described in this paper were taken on a resonator structure consisting of two superconductor disks. In this arrangement, the conductor losses dominate the resonator quality factor $Q$ and, thus, a higher accuracy can be achieved from this novel direct measurement technique.

## MICROWAVE TEST ARRANGEMENT

The best test structure for evaluating the surface resistance of superconductor bulk material would be a closed resonator such as a waveguide cavity.[4] Since most research groups currently produce superconductor material in simple disk form, a closed resonator is difficult to realize; large homogeneous blocks are not readily available, and machining leaves substantial surface damage with an associated increase in surface resistance.

Here, the disks are used in their original form for microwave measurements. Our test configuration is shown in Fig. 1. A small disk is separated from a larger disk (acting as a ground plane) by a very thin low-loss dielectric. Such a disk resonator configuration has a quality factor $Q$, which is determined by conductor losses, radiation, and dielectric losses. Since the latter is generally very low for suitable dielectric materials, and the radiation losses can be reduced by keeping the spacing between

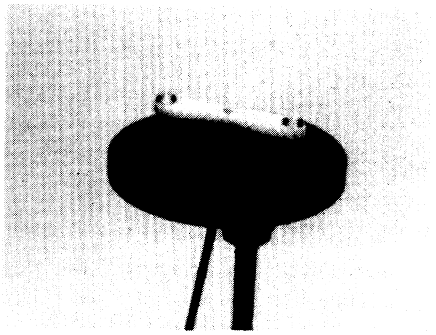

FIG. 1. Disk resonator with probe coupling.

the two disks at a minimum, the overall $Q$ will be dominated by the conductor losses of the disks. A small probe penetrates through the ground disk to excite the resonator at its dominant $TM_{11}$ mode at 10 GHz.

The disk resonator is mounted in a 3-in. inner diameter cryostat that permits cooling to liquid-helium temperatures in a single evacuated envelope. An HP8510B network analyzer is used for the characterization of the single-port resonator. The resonator response is deembedded for this purpose from the external connection and the rf feed by a computer controlled calibration procedure at different temperatures. Both phase and amplitude are used for the determination of the $Q$ factor.

## SAMPLE PREPARATION AND CHARACTERIZATION

Samples of nominal composition $YBa_2Cu_3O_{6+x}$ were prepared by mixing appropriate amounts of 99.9% pure $Y_2O_3$ from Rhone Poulenc and $BaCO_3$ and CuO from Fisher Scientific. The starting mixtures were attrition milled. This process produces the desired particle size in a short time, before agglomeration takes place. The detailed experimental procedure is depicted in Fig. 2. The samples prepared have been identified from their powder x-ray diffractograms reported earlier and match well with those of Wong-Ng *et al.*,[5] showing that they all have similar crystal structure. Densities of the sintered pellets as determined by Archimedes method were 5.64 g/cm$^3$. Average grain size obtained by scanning electron micrographs was ~20 $\mu$m. Superconductivity was measured by four-probe conductivity and by magnetic susceptibility measurements. Resistivity data for these materials sintered at different temperatures have been reported earlier[6,7] showing a $T_c$ at 90 K with a room-temperature resistivity of just 4 m$\Omega$ cm. The magnetic susceptibility was measured by filling the secondary coil of a 15-Hz mutual inductance bridge. The bridge has a resolution of 3 ppm of the full Meissner signal when the sample is filled completely in the secondary. The magnetic susceptibility of the attrition milled sample is shown in Fig. 3. The estimated volume fraction of the superconducting phase in these samples was more than 90% of the sample volume.

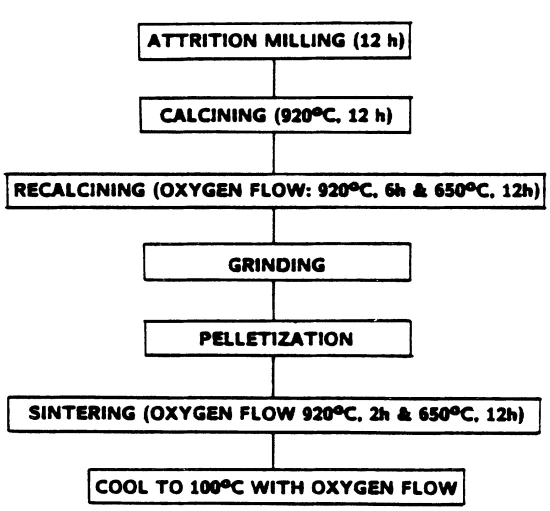

FIG. 2. Flow chart of sample preparation procedure.

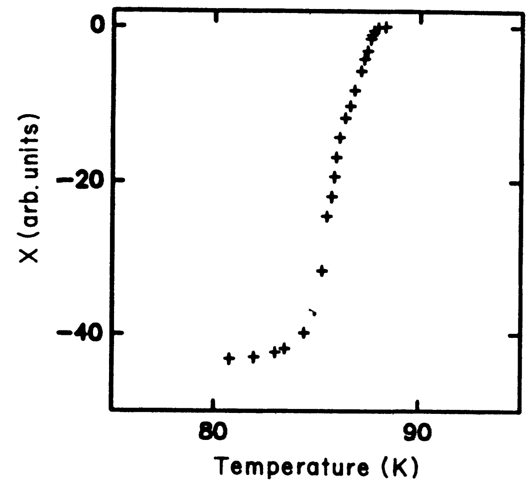

FIG. 3. Magnetic susceptibility as a function of temperature.

## MICROWAVE TEST RESULTS

The entire test set was calibrated first by measuring the $Q$ of a resonator structure consisting of Au-plated brass

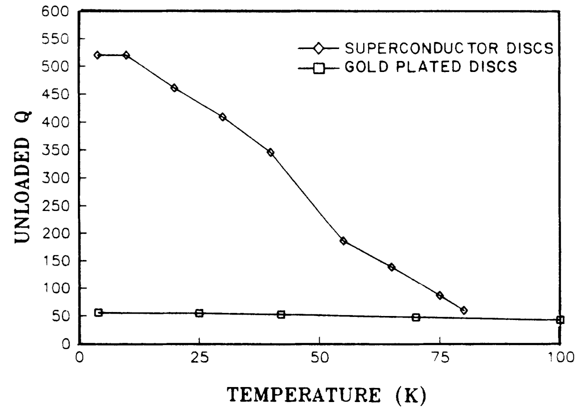

FIG. 4. $Q$ values vs temperature for superconductor and gold-plated disks.

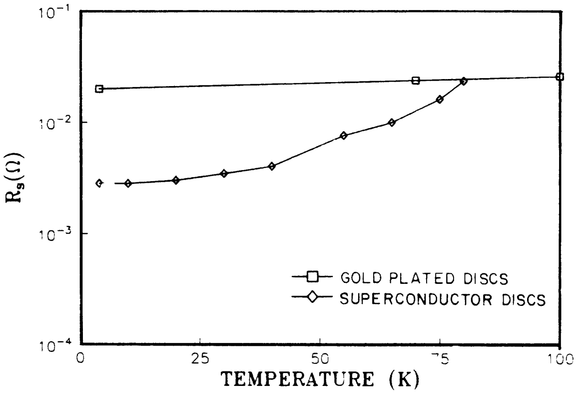

FIG. 5. Surface resistance vs temperature for gold-plated and superconductor disks.

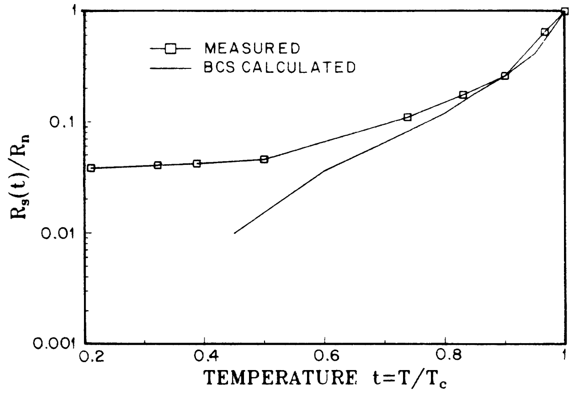

FIG. 6. Comparison of measured and BCS calculated resistance vs temperature.

disks having the same dimensions as the superconducting disks. A thin dielectric spacer of 1.2 mil polyethylene was used to keep radiation losses low. The same arrangement was used for testing various solid superconducting materials. So far, the best results have been obtained on material prepared by attrition milling. Figure 4 shows a comparison of $Q$ values for the gold-plated disks with superconducting disks as a function of temperature. Close to the transition temperature of the superconducting material as defined by dc measurements ($\sim$85 K) the $Q$'s of plated gold and the superconductor bulk material are approximately comparable. The $Q$ of the superconducting material continues to improve towards lower temperatures reaching a value an order of magnitude higher than that of plated gold at 10 K. To reduce radiation losses of the structure, thinner dielectrics could be used. This will become important especially when superconductor materials with better conductivities become available.

Since the $Q$ values in the particular resonator structure are dominated by conductor losses, they can be converted to surface resistance as shown in Fig. 5. The resulting surface resistance of Au at room temperature is 38 m$\Omega$ compared to the theoretical value of 30 m$\Omega$ and follows a gradual reduction towards lower temperatures close to theoretical expectations. The surface resistance of the bulk 1:2:3 compound reaches a value 10 times lower than that of gold at approximately 10 K.

The results obtained so far show these materials are not yet really useful for microwave applications. They do show, however, that even the current bulk material (rather coarse and porous by microwave standards) can reach surface resistance values an order of magnitude better than that of a metal, albeit at temperatures well below the transition point. A comparison of test results with numerical computations of the surface resistance, based upon the Bardeen-Cooper-Schrieffer (BCS) theory,[8] indicates that the overall temperature dependence of the surface resistance is reasonably similar to that of BCS close to $T_c$ as shown in Fig. 6. However, saturation at temperatures well below $T_c$ (i.e., $t < 0.3$) was noticed and measured. The saturation is a real physical effect, and the measured values do not agree with the simple BCS theory at low temperatures ($t < 0.5$) as seen in Fig. 6.

During the course of testing several materials, a number of significant observations were made. Machining and polishing of as-pressed and annealed surfaces reduced the $Q$ by about 10%–20%, apparently due to surface damage. The original $Q$ can be restored, however, by reannealing. Material prepared as described before appears to be quite stable against normal atmospheric conditions. Measurements on the same samples performed over a 5-month period showed no marked deterioration, even though no special precautions, such as storage in a controlled atmosphere were taken.[9] Changes in rf drive level from −20 to 0 dBm did not affect the $Q$ values measured, indicating that the surface resistance at these power levels was not current limited. A number of superconductor samples from other sources that showed well-pronounced Meissner effects and sharp dc transitions did not show good microwave performance. The exact conditions responsible for low microwave surface resistance are still under investigation.

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[1] M. Poirier *et al.*, J. Appl. Phys. **63**, 1646 (1988).

[2] J. S. Martens, J. B. Beyer, and D. S. Ginley, Appl. Phys. Lett. **52**, 1822 (1988).

[3] D. L. Moffat *et al.*, in *Extended Abstracts: High Temperature Superconductors II*, edited by D. W. Capone, W. H. Butler, B. Batlogg, and C. W. Chu (Materials Research Society, Pittsburgh, 1988), Vol. EA-14, p. 347.

[4] C. Zahoponlos, W. L. Kennedy, and S. Sridhar, Appl. Phys. Lett. **52**, 2168 (1988).

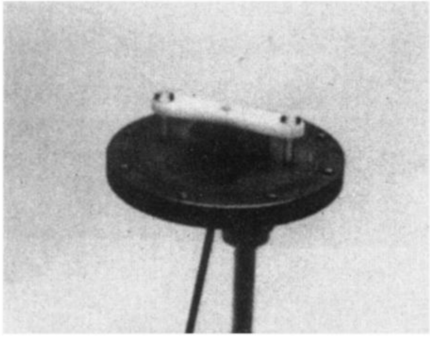
[5] W. Wong-Ng *et al.*, Adv. Ceram. Mater. **2**, 565 (1987).

[6] A. Safari *et al.*, in *High Temperature Superconductors*, edited by M. B. Brodsky, R. C. Dynes, K. Kitazawa, and H. L. Tuller (Materials Research Society, Pittsburgh, 1988), p. 269.

[7] A. Safari *et al.*, in *Proceedings of the 38th IEEE Electronic Components Conference* (IEEE, New York, 1988), p. 181.

[8] S. Sridhar, C. A. Shiffiman, and H. Hamdeh, Phys. Rev. B **36**, 2301 (1987).

[9] It does change markedly, however, when soaked in water at elevated temperature as noticed by susceptibility and x-ray photoelectron spectroscopy studies (Ref. 7).



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