RF PERFORMANCE OF POLYCRYSTALLINE HIGH-T_c SUPERCONDUCTORS

C. L. Bohn, J. R. Delayen, and C. T. Roche
Argonne National Laboratory
Argonne, Illinois 60439

ABSTRACT

We have measured the rf properties of bulk and thick-film high-T_c superconductors as functions of temperature, frequency, and rf field amplitude. The rf surface resistance typically depended strongly on field amplitude, but the samples remained superconducting up to the highest fields we could apply (~640 gauss). For example, at 220 MHz, the surface resistance of a bulk polycrystalline $YBa_2Cu_3O_{7-x}$ sample ranged from $\lesssim 2\mu\Omega$ at $\lesssim 0.05$ gauss and 4.2 K to 6.3 mΩ at 640 gauss and 77 K. The stability of the rf properties of this sample after storage both in air and in vacuum were also investigated.

1. INTRODUCTION

The low rf surface resistance of superconductors make these materials desirable for the construction of high-Q cavities. With critical temperatures (T_c) higher than the temperature of liquid nitrogen, and with the potential for sustaining surface fields as high as 27,000 gauss while remaining superconducting,[1] the ceramic-oxide superconductors are candidates for use in this application.

Unlike the dc resistance, the rf surface resistance of a material in the superconducting state is not zero, nor is it necessarily low. In the framework of a two-fluid model, most, but not all, of the electrons are bound in Cooper pairs; the remainder are normal-conducting.[2] The inertia of the Cooper pairs keeps them from responding quickly enough to high-frequency radiation to prevent penetration of the radiation into the superconductor. The rf field which leaks into the superconductor drives the normal-conducting electrons, which in turn collide with the lattice and deposit thermal energy. According to Bardeen-Cooper-Schrieffer (BCS) s-wave theory, for temperatures $T \lesssim 0.5\ T_c$ the rf surface resistance is approximately:[3]

$$R_s \approx \frac{A}{T} f^2 \exp\left(-\frac{\Delta}{T}\right)$$

where A is a constant, f is the rf frequency, and Δ is the binding energy of a Cooper pair. In general, a constant term R_0 called the residual surface resistance needs to be added. It is caused by irregularities in the material and is therefore unpredictable theoretically. The rf surface resistance of most low-T_c superconductors, such as lead and niobium, follow this relation. On the other hand, that of the high-T_c ceramics appears to deviate from the theory, particularly with regard to temperature dependence.[4,5]

The high-T_c ceramics are characteristically poor thermal conductors;[6] therefore, the use of high-conductivity substrates will probably be required to keep the ceramics cold while they are exposed to high rf fields. From a materials-processing standpoint, silver has been found to be compatible.[7] We have fabricated a variety of bulk samples, and thick films on silver substrates, of $YBa_2Cu_3O_{7-x}$ (YBCO) and Bi-Sr-Ca-Cu-O (BSCCO), and have measured their rf surface resistances versus temperature and rf field amplitude at frequencies from 150 MHz to 40 GHz. In this paper, we describe our measurement techniques and notable results, and we comment on future initiatives for research and development of high-T_c superconductors for high-power rf applications in light of the results.

2. MEASUREMENT APPARATUSES

A standard technique for determining the rf surface resistance (R_s) of a sample involves the use of resonant cavities. The sample is inserted in the cavity and modifies the quality factor of the cavity. Measurement of the quality factor before and after sample insertion enables the calculation of R_s.[8,9]

We employ two types of cavities in our measurements. In the transverse electromagnetic (TEM) cavity (Figure 1), a rod-shaped sample is supported on-axis inside a long, cylindrical outer conductor using a quartz tube. The sample behaves as a resonant coaxial line such that its length corresponds to one half-wavelength of the rf field. In the transverse electric (TE) cavity (Figure 2), the sample forms the bottom plate of a cylindrical cavity which we made to resonate in the TE_{012} and/or TE_{011} modes. We have constructed 8 cavities, 6 TE and 2 TEM, for experiments on samples ranging from small size up to 15-cm-diameter plates (Table 1).

In a typical experiment to measure R_s versus temperature, the temperature of the cavity was monitored with sensors mounted at the top and bottom outer surfaces of the cavity. The cavity and sample were cooled to 4.2 K with liquid helium and a measurement was taken. The helium was subsequently boiled away using a small heater, and the cavity was allowed to warm slowly enough that the two sensors recorded the same temperature to within 0.5 K. This assured a condition of approximate thermal equilibrium in the cavity.

For isothermal measurements of R_s as a function of rf field amplitude, a TEM cavity was used. The cavity and quartz tube were flooded with either liquid helium or liquid nitrogen so that, at all times, the temperature of the sample was known unambiguously. The peak rf magnetic field at the surface of the sample was determined from the voltage reading of a calibrated pick-up probe located halfway between the end-plates of the cavity, as shown in Figure 1.

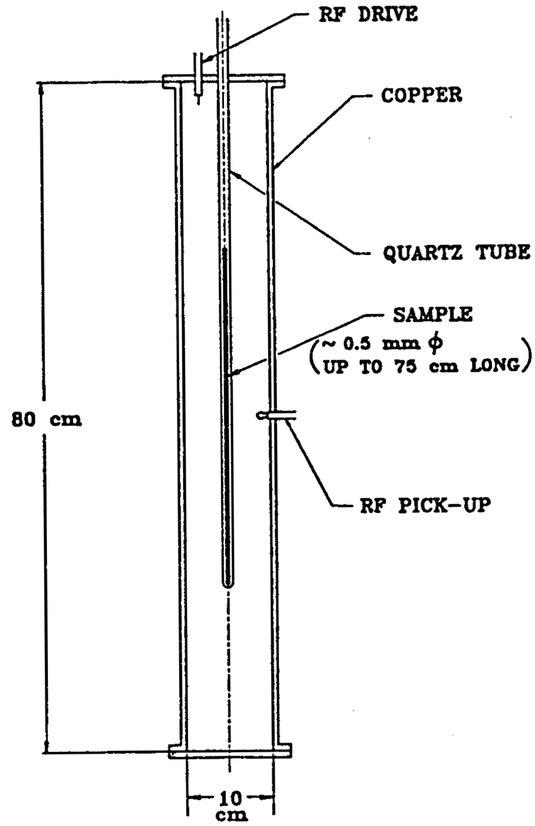

Figure 1. Generic TEM cavity. The sample is a rod which acts as a resonant coaxial line.

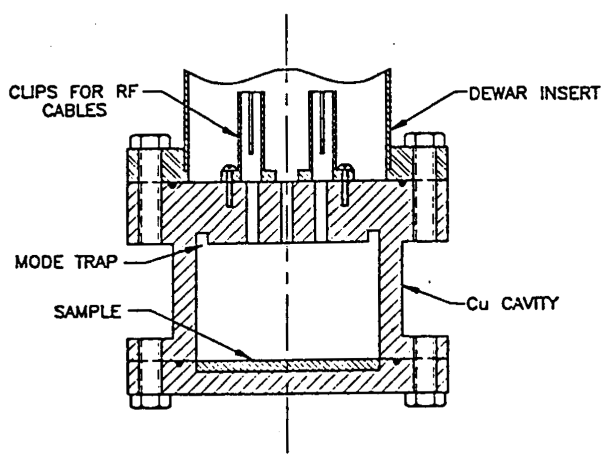

Figure 2. Generic TE cavity. The sample is the bottom surface of the cavity.

Table 1. Apparatuses for RF Measurements

Cavity	Frequencies	Capabilities
TEM Cu	150-600 MHz	Large Samples (rods) RF Breakdown
TEM Cu	600-1500 MHz	Large Samples (rods) RF Breakdown
$TE_{011,012}$, TEM Cu, Nb	1.5-4 GHz	Large Samples (rods and plates to 200 cm^2) Small Samples (disks, single crystals) RF Breakdown
$TE_{011,012}$ Cu	8.0-12.4 GHz	Large Samples (disks and plates to 20 cm^2)
$TE_{011,012}$ Cu	12.4-18 GHz	Medium Samples (disks and plates to 7 cm^2)
$TE_{011,012}$ Cu, Nb	26.5-40 GHz	Small Samples (disks and plates to 1.7 cm^2, single crystals)

Additional details on these cavities and the measurement techniques can be found elsewhere.[10-12]

3. RESULTS

3.1 BULK POLYCRYSTALLINE RODS

We fabricated several bulk ceramic YBCO rods from phase-pure YBCO powder which was combined with several organics to form a plastic mass and then extruded.[13] After firing, one such rod had a diameter of 0.44 mm and a length of 80 cm. To do measurements at different frequencies, we broke the rod to obtain the desired length. Initial measurements at low rf field amplitude (B_{rf}) were taken at 4.2 K and 77 K after fabrication, and the rod was stored in air for ten months. Both low-field and high-field measurements were then made. The low-field measurements from the two time periods agreed, indicating that the rf properties of the sample were stable over the ten-month interval. Additional low-field measurements were made after storing the sample in vacuum for 38 days. No significant change in the low-field data was observed. The lowest R_s measured was $\lesssim$1.1 μΩ at 4.2 K and 175 MHz.

The low-field behavior of R_s is plotted in Figure 3. It is characterized by a sharp transition between the superconducting state and the normal state. The frequency dependence of R_s was approximately quadratic at all temperatures below $T_c \approx 91$ K and was approximately square-root just above

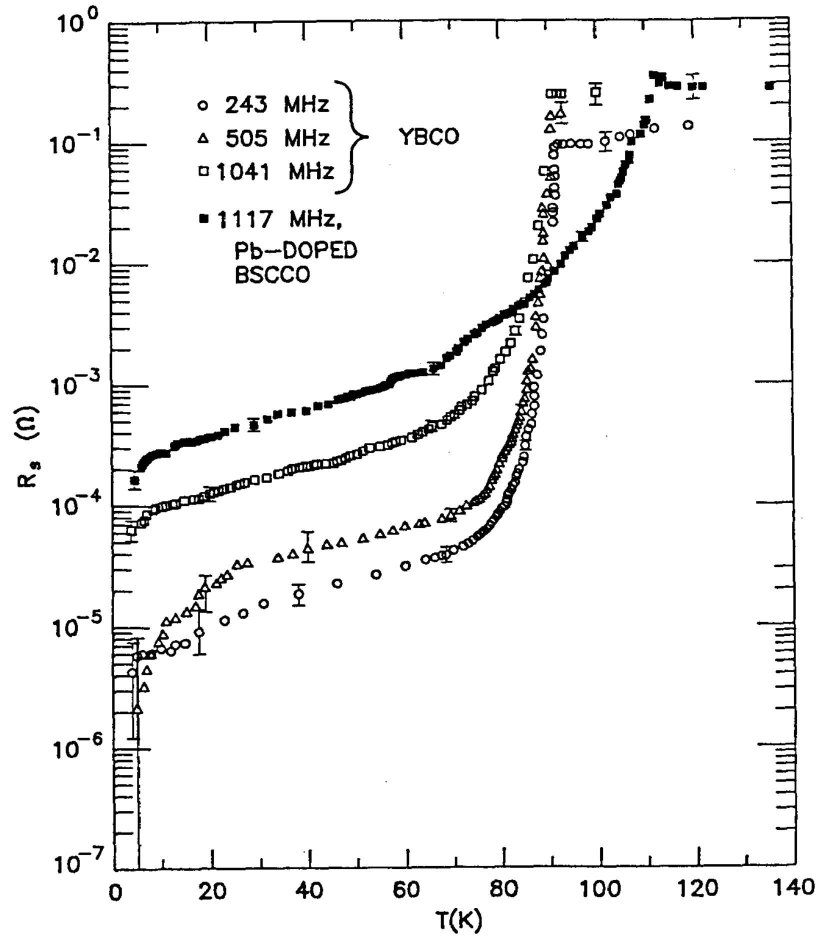

Figure 3. Surface resistance of bulk YBCO versus temperature at three frequencies and at low field ($B_{rf} \lesssim 0.05$ G). Low-field data for a bulk Pb-doped BSCCO sample at 1.1 GHz is also shown.

T_c. The temperature dependences of Figure 3 did not follow either BCS theory or a single power law over a wide temperature range. For comparison, the low-field R_s at ~1 GHz of a typical, similarly fabricated bulk rod of $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ is also shown in Figure 3. This sample has a transition temperature of approximately 110 K.

The dependence of R_s on the rf field amplitude at the center of the YBCO sample for T = 4.2 K and 77 K is illustrated in Figure 4. The surface resistance increased monotonically as B_{rf} was raised, passing through a transition region characterized by a strong B_{rf}-dependence, and saturating at a value roughly 5 percent of the normal-state surface resistance just above T_c. The sample remained superconducting out to the highest field achieved,

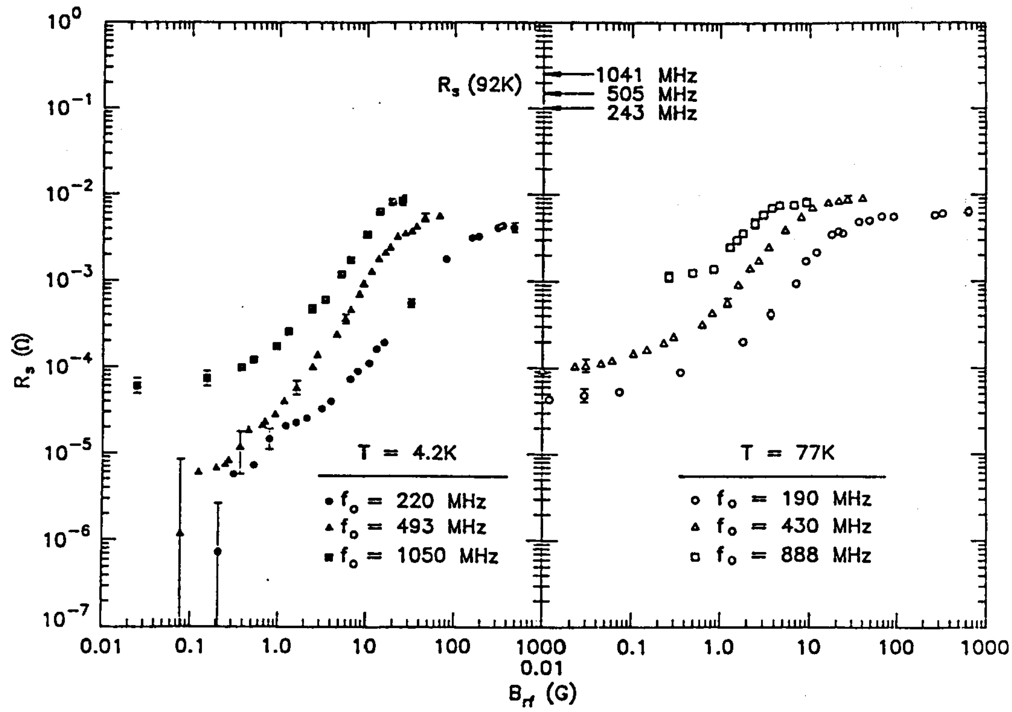

Figure 4. Surface resistance of bulk YBCO versus rf field amplitude at T = 4.2 K and T = 77 K.

$B_{rf} \cong 640$ G (at 77 K and 190 MHz). In the transition regions, R_s was strongly dependent on temperature and exhibited a frequency dependence which was approximately quadratic. In the high-field region, on the other hand, R_s showed a weak dependence on both temperature and frequency.

3.2 THICK POLYCRYSTALLINE FILMS ON SILVER SUBSTRATES

In earlier efforts, thick films of YBCO were fabricated by applying slurries to silver substrates, and several of these films had rf surface resistances comparable to bulk specimens.[14,15] We have recently fabricated thick films of BSCCO on silver, again by applying high-viscosity slurries.[16] Two different processing techniques were used; "4336" samples were produced using powders derived from the compound $Bi_4Sr_3Ca_3Cu_6O_x$, and a "2212" sample was produced using powders derived from $Bi_2Sr_2CaCu_2O_x$. All of the films were approximately 80-μm thick; the 4336 samples had diameters of 1.3 cm and 15 cm, and the 2212 sample had a 5.1 cm diameter. None of the samples was entirely phase-pure as judged by x-ray diffraction.

The rf surface resistance of these samples was measured as a function of temperature at low rf field amplitude ($\lesssim$0.1 gauss) using three of the copper TE cavities described in Table 1. The data is shown in Figure 5. The 4336 samples had $T_c \approx 81$ K, and the 2212 sample had $T_c \approx 83$ K. The surface resistances measured at low temperatures were approximately consistent with a quadratic frequency dependence, indicating that the rf performance of the 4336 and 2212 films were comparable despite their very different processing procedures. At low temperatures, the 2212 sample behaved like room-temperature copper at the X-band frequencies of the rf fields to which it was

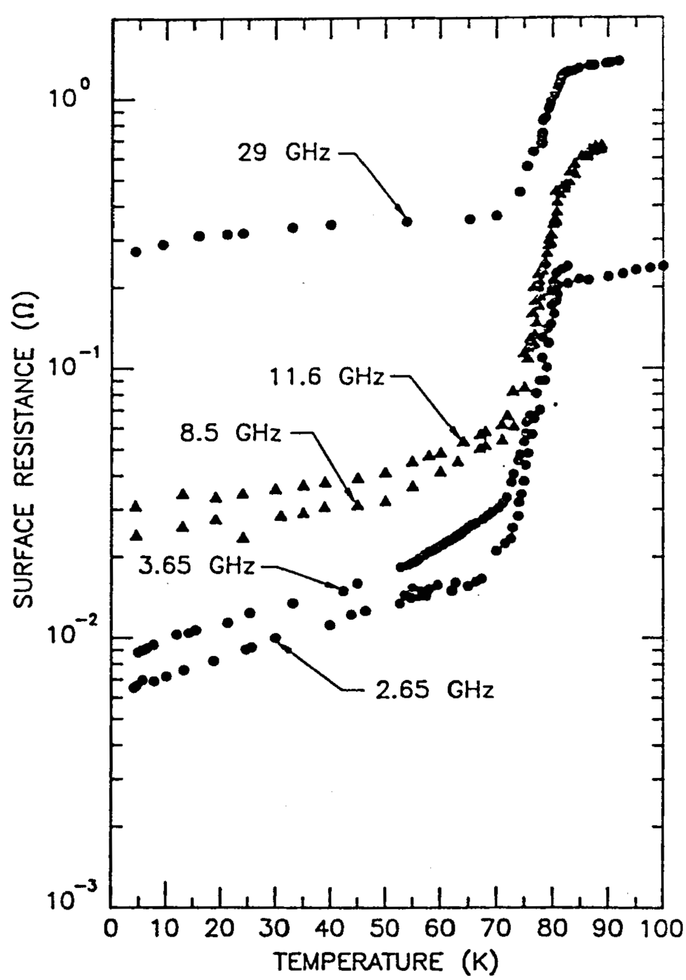

Figure 5. RF surface resistance versus temperature of BSCCO thick films on silver at low field ($B_{rf} \lesssim 0.1$ G). Circles correspond to the 4336 samples; triangles correspond to the 2212 sample.

exposed. The performance of these films was also similar to that achieved recently in bulk BSCCO. For a 2212 pellet of surface area 0.013 cm^2, a surface resistance of 3.0 mΩ at 3 GHz, 4.2 K, and low field has been reported,[17] which is comparable in magnitude to R_s of the 15-cm-diameter 2212 film with larger surface area (182 cm^2).

As a first step toward identifying ways to improve the rf properties of polycrystalline BSCCO, we acquired a highly textured bulk sample of $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ of dimensions 2 cm in diameter by 0.5 mm in thickness. The introduction of Pb into the BSCCO lattice results in zero dc resistance at temperatures near 110 K. We measured the surface resistance of the sample as a function of temperature at 29.2 GHz in a TE cavity at low field. As is seen in Figure 6, the transition from the normal state to the superconducting state started at $T_c \approx 110$ K; it was broad by comparison with the transition of the thick film on silver. At temperatures $T \lesssim 77$ K, the surface resistance of the Pb-doped sample was lower than that of the undoped sample by a factor ~5.

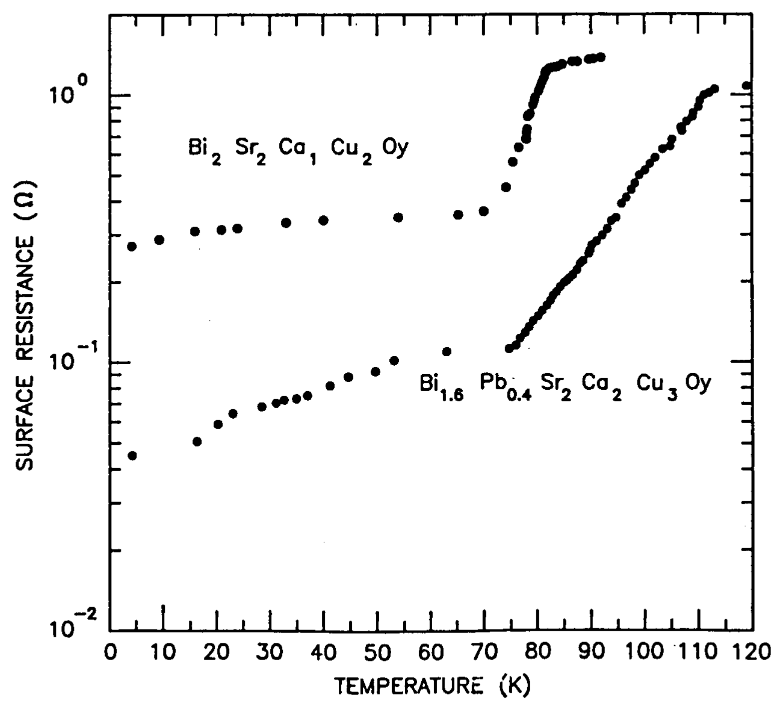

Figure 6. Comparison of rf surface resistance versus temperature at low field of bulk Pb-doped BSCCO versus BSCCO thick film on silver at 29.2 GHz.

4. DISCUSSION

Resonant superconducting cavities for high-power rf applications need to have low surface resistances while sustaining high peak fields on the cavity walls. For example, a resonant structure for accelerating charged particles will require surface resistances no higher than a few tens of micro-ohms at surface fields as high as a few hundred gauss and at frequencies in the range 200-1800 MHz. Our data on bulk YBCO (Figure 4) indicate that at low fields, sufficiently low surface resistances are achieved, but at high fields (e.g., $\gtrsim$100 gauss) the surface resistance is too large by a factor of ~100. Our observation that the sample remains superconducting at high rf field is favorable, however.

The BSCCO thick films may also have surface resistances of interest at low fields. If they have a quadratic frequency dependence over a sufficiently wide range of frequencies, then, for example, their low-field, low-temperature surface resistance at 200 MHz would be ~40 $\mu\Omega$. If the highly textured bulk sample of Pb-doped BSCCO also exhibits a quadratic frequency dependence, its projected surface resistance at low field and low temperature is ~2 $\mu\Omega$ at 200 MHz, a value comparable to that observed in the bulk YBCO sample.

Other investigators have also seen a pronounced rf-field dependence of R_S in polycrystalline high-T_c superconductors,[18] though it is uncertain whether single crystals exhibit this property. An attempt to find field-dependence in YBCO single crystals gave results which were nearly independent of field.[19] However, it seems that this data refers to upper bounds. Thus, although it is clear that the surface resistance of single crystals can be lower than polycrystals, the field dependence of the surface resistance of single crystals is still an open question.

The poor thermal conductivity of the ceramic oxides motivates the development of thin films on high-conductivity substrates. The minimum film thickness must be of the order of 1 μm, corresponding to a few rf penetration depths, in order to shield the substrate from the rf field. To coat complicated geometries, for example structures used in ion accelerators,[20] a method that does not involve line-of-sight deposition may be needed, such as chemical vapor deposition. Accordingly, the next step toward practical high-power rf applications of these materials is the production of suitable thin films, first on large surface areas, and second on complicated shapes.

In view of the high-field properties of polycrystalline ceramics described here, it is necessary to measure the surface resistance versus field of thin films. Information from these measurements will aid those who fabricate the films in an iterative process to reduce the high-field surface resistance to useful levels. To initiate this process, we are now building a niobium cavity for high-field measurements at 850 MHz of flat samples with surface areas nominally 6.5 cm^2. In the meantime, until a method for producing films with adequate rf properties is found, niobium will continue to be the material of choice for the construction of high-power resonators.

ACKNOWLEDGMENT

We are grateful to Dr. Huey-Lin Luo of the University of California at San Diego for providing the Pb-doped BSCCO sample. This work was supported by the U.S. Department of Energy under contract W-31-109-ENG-38 and by the U.S. Army Strategic Defense Command.

REFERENCES

1. T.K. Worthington, W.J. Gallagher, and T.R. Dinger, Phys. Rev. Lett., **59** (1987) 1160.

2. I.M. Khalatnikov and A.A. Abrikosov, Adv. in Physics, **29** (1959) 45.

3. A. Abrikosov, L. Gorkov, and I. Khalatnikov, Sov. Phys. JETP, **8** (1959) 1982.

4. W.L. Kennedy and S. Sridhar, Sol. State Comm., **68** (1988) 71.

5. T.L. Hylton, A. Kapitulnik, M.R. Beasley, J.P. Carini, L. Drabeck, and G. Gruner, Appl. Phys. Lett., **53** (1988) 1343.

6. J. Heremans, D.T. Morelli, G.W. Smith, and S.C. Strite III, Phys. Rev. B., **37** (1988) 1604.

7. M.T. Lanagan, K.C. Goretta, J.P. Singh, G.T. Goudey, J.T. Dusek, D.I. Dos Santos, and R.B. Poeppel, in Superconductivity and Its Applications, H.S. Kwok and D.T. Shaw, eds., Elsevier: New York (1988) 118-123.

8. S. Sridhar and W.L. Kennedy, Rev. Sci. Instru., **59** (1988) 531.

9. J.R. Delayen, K.C. Goretta, R.B. Poeppel, and K.W. Shepard, Appl. Phys. Lett., **52** (1988) 930.

10. C.L. Bohn, J.R. Delayen, D.I. Dos Santos, M.T. Lanagan, and K.W. Shepard, IEEE Trans. Magnetics, **25** (1989) 2406.

11. J.R. Delayen and C.L. Bohn, Phys. Rev. B (to appear in 1 Sep 89 issue).

12. C.L. Bohn, J.R. Delayen, U. Balachandran, and M.T. Lanagan, Appl. Phys. Lett., **55** (1989) 304-306.

13. M.T. Lanagan, R.B. Poeppel, J.P. Singh, D.I. Dos Santos, J.K. Lumpp, U. Balachandran, J.T. Dusek, and K.C. Goretta, J. Less-Com. Met. (in press).

14. J.R. Delayen and M.T. Lanagan, "High-Power RF Applications of High-T_c Superconductors," in Superconductivity and Its Applications, H.S. Kwok and D.T. Shaw, eds., Elsevier: New York (1988) 286-290.

15. C.L. Bohn, J.R. Delayen, D.I. Dos Santos, M.T. Lanagan, and K.W. Shepard, op. cit.

16. C.L. Bohn, J.R. Delayen, U. Balachandran, and M.T. Lanagan, op. cit.

17. D.W. Cooke, E.R. Gray, R.J. Houlton, B. Rusnak, E. Meyer, G.P. Lawrence, M.A. Maez, B. Bennett, J. D. Doss, A. Mayer, W.L. Hults, and J.L. Smith, "RF Surface Resistance of Bulk $Tl_2Ca_2Ba_2Cu_3O_{10}$, $Bi_2CaSr_2Cu_2O_8$, and $YBa_2Cu_3O_x$" (unpublished).

18. H. Padamsee, K. Green, J. Gruschus, J. Kirchgessner, D. Moffat, D.L. Rubin, J. Sears, Q.S. Shu, R. Buhrman, D. Lathrop, T.W. Noh, S. Russek, and A. Sievers, in Superconductivity and Its Applications, H.S. Kwok and D.T. Shaw, eds., Elsevier: New York (1988) 249-254.

19. 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. B, **38** (1988) 6538.

20. J.R. Delayen, "Heavy-Ion Superconducting Linacs," in Proc. of the 1989 Particle Accelerator Conf., Chicago, Illinois, 20-24 March 1989.