Absorption at High Microwave Power by Large-Area Tl-Based Superconducting Films on Metallic Substrates

D. W. Cooke, P. N. Arendt, E. R. Gray, and A. M. Portis

Abstract—**Microwave surface resistance measurements have been made on large-area Tl-Ba-Ca-Cu-O thick films that are magnetron-sputtered onto oriented Ag alloy substrates by replacing the end wall of an 18 GHz TE_{011} mode Cu cavity with the superconducting film. The best surface resistance values obtained are 4 and 14 mΩ at 10 K and 77 K, respectively; corresponding Cu values are 8 mΩ and 21 mΩ. The dependence of the surface resistance on microwave power was measured in a similar way except that a Nb cavity was used instead of a Cu cavity. Typically, the surface resistance of the film begins to rise in 1–10 Oe of microwave field and saturates in 20–60 Oe. A model is presented relating the observed saturation to critical penetration of Josephson junctions. Films exhibiting the highest degree of *c*-axis texturing show the weakest dependence of surface resistance on power and also exhibit the sharpest transition to the superconducting state as measured at high frequency. These results are important for the development of high-power microwave cavities.**

I. Introduction

FOLLOWING the discovery of copper-oxide-based superconductors with transition temperatures higher than 77 K, it was recognized that these materials were potentially important for radio-frequency (RF) and microwave applications. This pioneering work has been widely reviewed [1]–[5]. Passive microwave components requiring small-area films deposited onto planar surfaces were expected to be the first application of high-temperature superconductors (HTS's), and in fact such devices have already been constructed [6], [7]. A more stringent application of HTS will be microwave cavities, where large-area films deposited onto nonplanar, metallic substrates will be required. And, unlike the low-power applications in passive microwave devices, cavity applications will require films with good power-handling characteristics in order to obtain the accelerating gradients necessary for HTS devices to be competitive with Cu and Nb [8]–[11]. Accordingly, we have investigated the surface resistance (R_s) and power dependence of Tl-based films deposited onto large-area metallic substrates.

II. Tl Film Preparation

A description of the technique for fabricating Tl-based films has been given recently [12], and we mention here only the salient features. Precursor films were prepared by magnetron sputter deposition onto 37-mm-diameter substrates of the Ag-based alloy Consil 995. To ensure compositional uniformity of the films, the substates were rotated and offset from the central axis of the sputter gun during deposition. Two targets were used for the deposition—metallic Tl-Ba-Ca-Cu and oxides of Ba-Ca-Cu. The advantage of using the oxide target is that the deposition system does not have to be contained within a filtered hood.

Following deposition, the films were placed in an alumina crucible, which was contained in a box oven. The films were annealed at high temperatures in an environment of O_2 and TlO_x. Annealing protocols varied from 2 to 20 min and from 860° to 905° C maximum temperature. Three of the four films described here were fabricated with intermediate buffer layers of BaF_2 and are characterized as unoriented, partially oriented, or oriented. One film was fabricated without the buffer layer and is characterized as well oriented. X-ray diffraction data for the unoriented and well-oriented films are shown in Fig. 1.

III. Surface Resistance

A. Temperature Dependence

The temperature dependence of R_s for the four Tl-based films was measured with an 18 GHz Cu cavity resonant in its fundamental TE_{011} mode. The end wall of the cavity was replaced by either the superconducting film or, for determination of the sample geometry factor, G, by a stainless-steel standard [10], [11]. R_s for the film is

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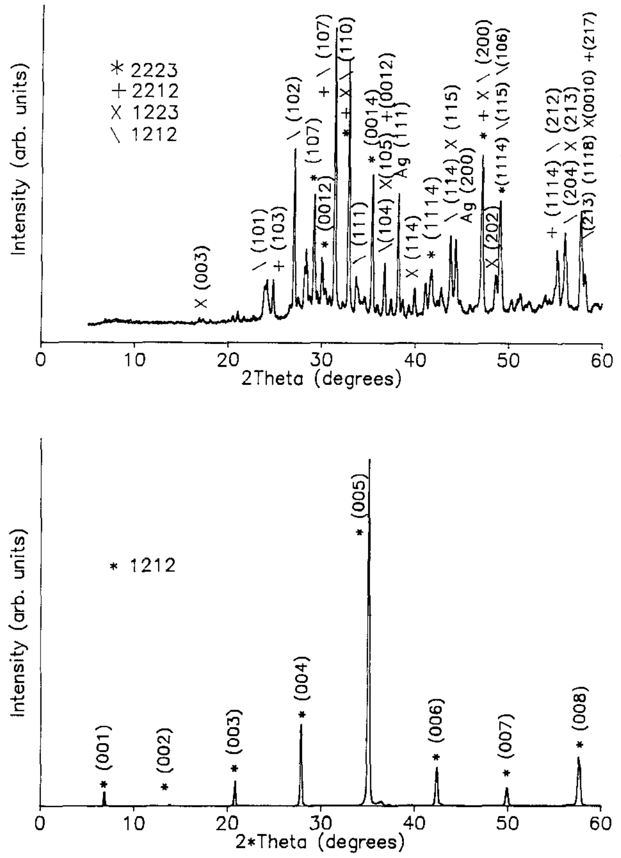

Fig. 1. X-ray diffraction pattern for the unoriented (upper panel) and well-oriented (lower panel) Tl films.

obtained from the relation

$$R_S = G\Delta\left(\frac{1}{Q}\right) \tag{1}$$

where the quality factor, Q, is determined from the frequency dependence of the transmission through the cavity.

Results of the $R_s(T)$ measurements are shown in Fig. 2. The data have been normalized to the superconducting transition temperature, T_c, for each film so that the high-frequency transition widths can be compared. The well-oriented film exhibits both the sharpest transition and lowest value of R_s at 4 K (4 mΩ). Interestingly, the sharpness of the transition increases with c-axis texturing, but the low-temperature values of R_s are not correlated with texturing. Perhaps this is not surprising since the "residual resistance" of all four of the films is more than two orders of magnitude higher than that of Nb. This suggests that R_s is representative of intrinsic film properties only near T_c whereas at lower temperatures R_s is dominated by extrinsic properties. However, recent data obtained on epitaxial Tl-based thin films show that R_s is a factor of 8 and 14 lower at 4 and 77 K, respectively, than present values obtained on polycrystalline samples. This indicates that either $a-b$ plane orientation is important in reducing R_s or, alternatively, that laser-ablated films contain fewer impurities and defects that adversely

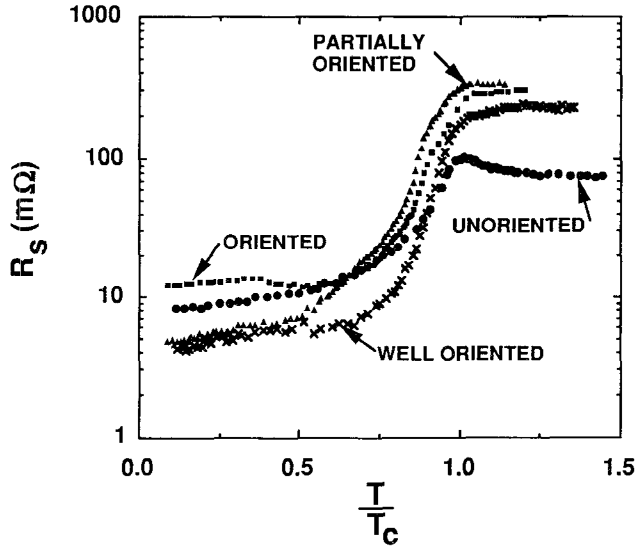

Fig. 2. Surface resistance versus reduced temperature for four Tl-based films. The sharpest transition occurs for the well-oriented film.

affect R_s [13]. In either case the salient point is that with improved processing techniques one can expect lower values of R_s for Tl films deposited on large-area, metallic substrates, which are necessary for RF cavity applications.

B. DC Field Dependence

A number of investigators have observed increases in R_s in applied static magnetic fields. Simultaneous measurement of R_s and X_s, the surface reactance, permits a determination of the variation of both λ (the magnetic penetration depth) and δ (the classical skin depth) with H. The surface resistance and the cavity frequency ν near 18 GHz have been measured in a Cu cavity [10] with H perpendicular to the plane of the film. The observed increase of R_s with H is a sigmoidal curve that saturates in fields of about 100 Oe. The fractional frequency shift is related to the change in surface reactance by

$$\frac{\Delta\nu}{\nu} = -\frac{1}{2}\frac{\Delta X_s}{G} \tag{2}$$

where G is again the sample geometry factor determined with a stainless-steel standard.

The square of the wavevector in the superconductor is

$$k^2 = \omega^2\mu\epsilon = -1/\lambda^2 + 2i/\delta^2 \tag{3}$$

with the real part of k^2 written in terms of a penetration depth, λ, and the imaginary part in terms of a skin depth, δ. The advantage of defining λ and δ in this way is that they are related directly to the real and imaginary parts of the film specific admittance $1/z = -i\omega\epsilon$. The surface impedance is

$$\begin{aligned} Z_s = R_s - iX_s &= (\mu/\epsilon)^{1/2} = (-i\omega\mu z)^{1/2} \\ &= -i\omega\mu\lambda\left[1 - 2i(\lambda/\delta)^2\right]^{-1/2}. \end{aligned} \tag{4}$$

The resistive part of the surface impedance climbs toward $R_c = \frac{1}{2}\omega\mu\delta$ as λ increases with respect to δ, the result of junction decoupling. The surface reactance similarly increases toward $R_c = \frac{1}{2}\omega\mu\delta$ as defects are decoupled. The films are represented by a lumped-element circuit. A *specific* kinetic inductance (or inductivity) $l_1 = \mu\lambda_1^2$ represents superconducting transport in a defect-free film with the London penetration depth $\lambda_1 \approx 0.2$ μm. An element $l_2 = \mu\lambda_2^2$ represents the Josephson kinetic inductance of defects, with λ_2 being an averaged Josephson penetration length. A resistivity $\rho_2 = \frac{1}{2}\omega\mu\delta_2^2$ in shunt with l_2 represents junction losses. This much of the circuit is equivalent to the model applied to granular superconductors by Hylton and Beasley [14]. A shunt screening inductance l_3 must be added for high-quality films to represent currents that flow around defects. This element is not required for granular superconductors, where RF current must flow across grain boundaries. Screening may also be negligible in a patterned stripline where the bulk of the current is forced across defects.

The assumption of the analysis is that dc magnetic fields increase the Josephson inductivity l_2 but do not affect the shunt resistivity ρ_2. To further simplify the analysis we have assumed that the film impedance is dominated by defects and that l_1 can be neglected.

C. RF Field Dependence

Cavity applications of HTS require low values of R_s that must be sustained in moderate surface magnetic fields H_{RF}. Measurement of the field dependence of R_s for HTS material has been difficult because of the poor thermal conductivity of the superconductor and the consequent inability to remove the RF-generated heat. Although pulsed RF measurements mitigate this problem, it is difficult to ensure that microscopic heating does not occur with an increase in R_s caused by a temperature rise rather than by intrinsic effects. Nevertheless, various groups have reported $R_s(H_{RF})$ for HTS single crystals, films, and bulk specimens [10], [13], [16]–[20].

We have constructed an 18 GHz Nb cavity resonant in the TE_{011} fundamental mode which places the metallic substrate of the Tl-based film in direct contact with liquid helium, thus providing a good thermal sink [10]. In a typical experiment we sweep through the cavity resonance in a time period ranging from 50 to 150 ms. Values of surface resistance vary by approximately 20% depending on the sweep time as a result of thermal heating of the sample under test. Shown in Fig. 3 are the results for the four samples investigated in this work. It is evident that c-axis texturing reduces the field dependence of R_s. For the unoriented film the increase is $\Delta R_s \sim 10$ for $H_{RF} \sim$ 20 Oe; a similar ΔR_s for the well-oriented film requires $H_{RF} \sim 55$ Oe. Thus, c-axis texturing provides two improvements in the high-frequency characteristics of Tl-based films:

i) a sharper transition into the superconducting state, as discussed above;
ii) a weaker dependence of R_s on H_{RF}.

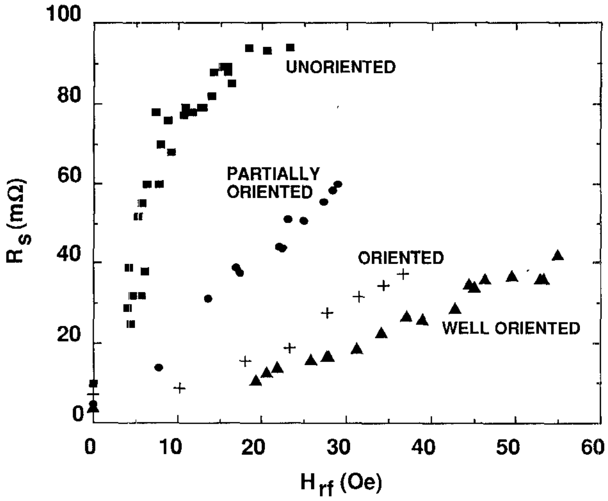

Fig. 3. Field dependence of the surface resistance for Tl-based films exhibiting varying degrees of c-axis texturing.

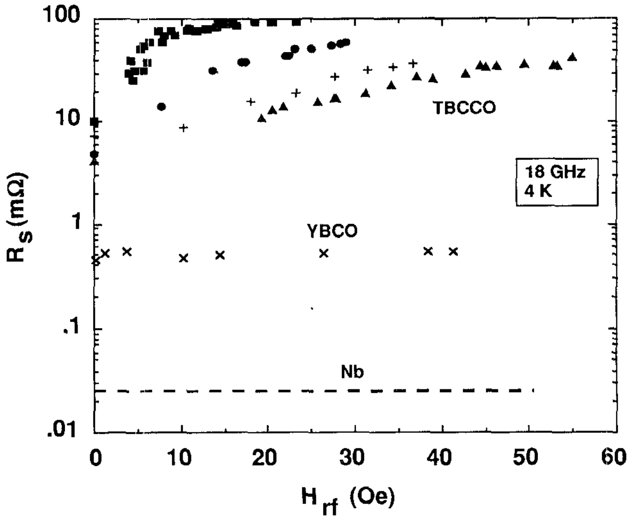

Fig. 4. Field dependence of the surface resistance for the four films of Fig. 3 compared with an epitaxial YBCO film and with Nb.

The first improvement is important for device operation at 77 K, and the second improvement is necessary for cavity applications. It is important, however, to put these results in proper perspective. Fig. 4 shows the data of Fig. 3 together with recent data obtained from an epitaxial film of $YBa_2Cu_3O_7$ (YBCO) that has been coevaporated onto MgO. This latter film shows that R_s is independent of H_{RF} for fields up to about 40 Oe. Assuming that Tl films behave in a similar manner, one can expect comparable improvements in field dependence with improved processing techniques. Also, it is noteworthy that the magnitude of R_s for these polycrystalline Tl films in low field is an order of magnitude higher than for YBCO and more than two orders of magnitude higher than for Nb. A comparison of R_s data for Tl-based films deposited onto both metallic and dielectric substrates along with bulk data is discussed below.

Returning to the data of Fig. 3, we briefly discuss present efforts to understand the dependence of R_s on H_{RF}. Although not shown in Fig. 3, we have also measured the dependence of R_s on H_{dc} and have found that static fields are similar but less effective than RF fields in increasing R_s. The static case is easier to understand and is thought to be due to the decoupling of superconducting grains by intergranular magnetic flux [15] as discussed above. The increase in R_s with elevated microwave power is significantly different and is associated with the development of an intergranular critical state of microwave flux, as evidenced by the measured linear increase in R_s with H_{RF} [21] and with frequency, as discussed below.

D. Frequency Dependence

Measurements of the RF surface resistance at 820 MHz have been made at 4.2 K in a pulsed quarter-wave coaxial Nb cavity, resonant in its fundamental transverse electromagnetic (TEM) mode [18]. The sample, a silver disk coated on both sides with the Tl-based superconducting film, was placed in a recession in the base of the cavity at a position of maximum RF magnetic field. For a determination of the sample geometry factor G, the specimen was replaced in turn by superconducting niobium and stainless-steel disks. The RF surface resistance of the superconducting film was computed from (1) with Q properly determined from the initial cavity decay [19].

In summary, dc magnetic fields increase surface resistance by granular decoupling. Radio-frequency fields are considerably more effective than dc fields in increasing the surface resistance and by a distinctly different mechanism. The obtained linear frequency dependence is strongly suggestive of ac loss in which magnetic work is performed over a period of the RF field.

IV. Geometrical Limiting and ac Loss

A. The Model

Power-dependent increases in surface resistance have been observed at RF and microwave frequencies in samples of ceramic YBCO [24]–[27] as well as in Tl-based films [10], [11], [15] and are suggestive of a geometrical origin. As described above, the characteristics of the loss are:

i) The power-induced surface resistance increases linearly with the frequency.
ii) The surface resistance is a sigmoidal function of RF field, increasing quadratically at the lowest fields, passing through a linear region, and finally saturating at higher fields. The initial quadratic dependence appears to be absent for resonant open structures such as striplines [22] and loop gap resonators [24], where intense RF fields may build up at edges.
iii) The form of the functional dependence on RF magnetic field is sensitive to sample preparation.
iv) The form of the dependence on RF field is insensitive to frequency.

All these characteristics may be explained by a model in which the RF penetration of intergranular regions near the surface is limited by critical currents. Much like bulk critical-state penetration [28], work is performed around a cycle of the ac magnetic field as a result of irreversible processes, leading to a dissipation rate that is proportional to the ac frequency. Because intergranular critical currents are small, the ac loss in these structures is expected to saturate at RF fields well below those expected in bulk. Further, where intergranular flux penetrates into the sample only partially, ac loss saturates at a correspondingly reduced field.

B. Critical Absorption and Dispersion

In a medium of depth r_c and macroscopic critical current density J_c, the surface reactance is expected to increase linearly with RF field up to the penetration field [28] $H^* = J_c r_c$ and to asymptotically approach a constant at higher fields. For J_c independent of RF field, the computed surface reactance for $H_{RF} < H^*$ is

$$X_s(H_{RF}) = \omega\mu r_c H_{RF}/2H^*. \tag{5}$$

For $H_{RF} \gg H^*$, the expected asymptotic value of $X_s(H_{RF})$ is

$$X_s = \omega\mu r_c. \tag{6}$$

For $H_{RF} < H^*$ the surface resistance is also expected to increase linearly with H_{RF} as

$$R_s(H_{RF}) = (4/3\pi)\omega\mu r_c H_{RF}/2H^*. \tag{7}$$

For $H_{RF} > H^*$ we expect

$$R_s(H_{RF}) = (2/\pi)\omega\mu r_c(H^*/H_{RF}) \cdot [1-(2/3)(H^*/H_{RF}] \tag{8}$$

which goes through a maximum at $H_{RF} = \frac{4}{3}H^*$ with the value $R_s = (3/4\pi)\omega\mu r_c$ and falls off slowly at higher RF fields. For $H_{RF} < H^*$ the critical surface resistance should be a fraction $4/3\pi = 0.4244$ of the critical surface reactance. The maximum value of R_s is expected to be $3/4\pi = 0.2387$ of the asymptotic value of the surface reactance. The logarithmic integral of R_s is

$$LI = \int_0^\infty R_s(H_{RF})\, d\ln H_{RF} = \frac{2}{\pi}\omega\mu r_c. \tag{9}$$

C. Low-Frequency AC Loss

Lam *et al.* [29] have measured the ac permeability of ceramic YBCO rods as a function of ac amplitude and find two maximum in the imaginary part of the permeability μ''. The lower-field maximum, which disappears when the rod is powdered, is believed to be associated with intergranular penetration. The higher-field maximum is taken to be an intragranular penetration field. Associated with these maxima in μ'' are steps in the real part of the permeability μ', as expected for critical-state behavior. The obtained ratios of the logarithmic integrals to the steps in the dispersion are 0.53 for intergranular flux and

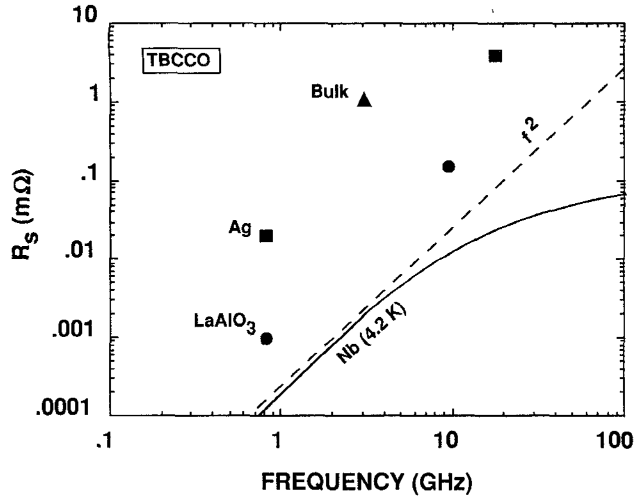

Fig. 5. Frequency dependence of the surface resistance for Tl-based HTS material. The triangle represents a bulk specimen, squares correspond to films deposited onto metallic substrates, and the circles correspond to films deposited onto dielectric substrates. Measurements were made at 4 K.

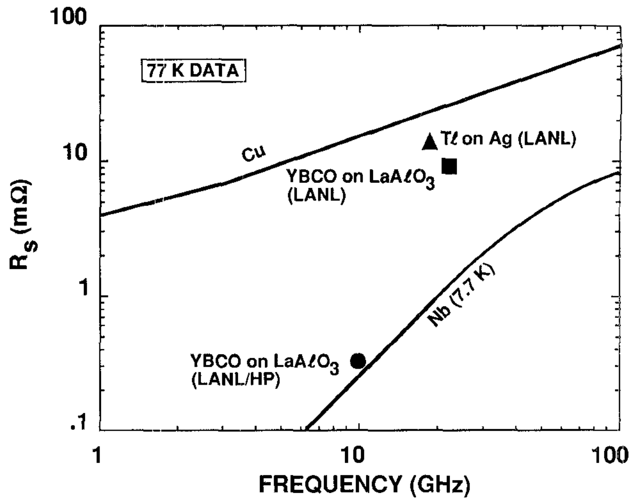

Fig. 6. Frequency dependence of the surface resistance for a large-area Tl-based film on a metallic substrate (triangle), large-area YBCO film on a dielectric substrate (square), and small-area YBCO on a dielectric substrate (circle). Cu and Nb are shown for comparison.

a lower limit of 0.28 for intragranular flux. These quantities are to be compared with a calculated value of $2/\pi = 0.64$.

In the case of intergranular critical flux penetration, J_c is an effective-medium critical current density that is related to the Josephson critical current density, J_0, by $J_c \approx (a/2\lambda_L)J_0$, where a is a grain diameter and λ_L is the London depth for the penetration of flux into grains. The depth r_c must similarly be interpreted as an effective-medium depth and is to be related to the junction depth r_0 by $r_c = (2\lambda_L/a)r_0$.

Zannella *et al.* [30] have measured ac loss at 50 Hz on fine silver tubes that contain sintered $Bi_2Sr_2CaCu_2O_{8+x}$ powder and find that the power dissipated, P, is linear in frequency, as expected for ac loss. Whereas P is expected [28] to increase as H_{RF}^3, it is found to increase only as H_{RF}^2. On the basis of the microwave experiments that have been performed [15], [17]–[20], [27], it appears that Zannella *et al.* [30] may have been in the saturation range of H_{RF} even at their lowest current of 5 A. In fact, a current of 5 A through a fine wire of radius 0.029 cm produces a surface RF field, $H_{RF} = 34$ Oe. This is approximately the field at which saturation is observed in Tl-based films at both 820 MHz and 18 GHz [15]. Measurements in progress of the ac loss of Tl-based films whose microwave loss characteristics have been reported should provide a good test of this model.

V. Comparisons and Conclusions

A comparison of bulk Tl-Ba-Ca-Cu-O, of Tl-based thick films on large-area, metallic substrates, of Tl-based thin films on small-area dielectric substrates, and of Nb is illustrated in Fig. 5. All R_s values were measured at 4 K. The trend of the data is clear—bulk polycrystalline material exhibits higher values of R_s than do the polycrystalline, textured thick films, which exhibit higher R_s values than the epitaxial thin films. The squares in Fig. 5 correspond to Tl-based films magnetron-sputtered onto Ag alloy (Consil 995) substrates, and the circles represent Tl-based films laser-ablated or coevaporated onto $LaAlO_3$ substrates. Although the measurements were made at different laboratories, the approximately quadratic dependence of R_s with microwave frequency is observed. The trend of the data, i.e., a reduction in R_s going from unoriented, bulk material to highly oriented, epitaxial films, suggests that elimination of grain boundaries is crucial for attainment of lower R_s values. Therefore, improved methods of producing epitaxial Tl-based films on Ag must be developed if further reductions in residual R_s are to be expected.

Finally, in Fig. 6 we compare R_s values for HTS films prepared by different techniques, using both dielectric and metallic substrates of varying size measured at 77 K. For large-area films deposited onto metallic substrates the lowest value obtained is a factor of 2 higher than that of a 5.1 cm^2 coevaporated film deposited onto a dielectric substrate, which is a factor of 10 higher than a small-area, laser-ablated film deposited onto a dielectric substrate. The salient feature of the data in Fig. 6 is that the lowest R_s values have been obtained for small-area epitaxial films. For large-area applications, such as RF cavities, it will be necessary to scale those deposition processes that yield epitaxial films. Whether this scaling can be accomplished for large-area films on metallic substrates is an open question.

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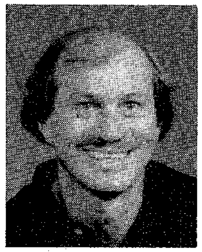

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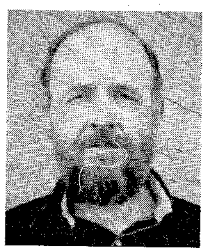

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