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HIGH TEMPERATURE SUPERCONDUCTING FILMS AND MULTILAYERS FOR ELECTRONICS

John R. Gavaler and John Talvacchio Cryogenic Electronics

Annual Report for the Period February 20, 1991 to February 20, 1992

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1. ANNUAL REPORT: HIGH TEMPERATURE SUPERCONDUCTING FILMS AND MULTILAYERS FOR ELECTRONICS

February 20, 1991 to February 20, 1992 AFOSR Contract No. F49620-91-C-0034 J. R. Gavaler and J. Talvacchio

2. ABSTRACT

The four tasks of this three-year program address properties fundamental to (1) enhancing the superconducting properties of HTS films, (2) the application of HTS films in passive microwave circuits, (3) the realization of HTS digital electronics, and (4) the development of new superconducting devices. Progress during the first year included significant enhancements in the properties of superconducting YBCO films. One of these, the elimination of surface roughness due to Cu-O particles, was essential for the subsequent growth of high quality multilayer structures. For the first time, uniform large-area YBCO films were sputtered on both sides of a substrate with properties equal or superior to those of single-sided smaller-area films. Two new device configurations were developed: the integration of a ferroelectric film on a YBCO electrode compatible with ferroelectric memory cells and a step-edge S-N-S Josephson junction using in-situ deposited gold as the normal metal. SQUIDs fabricated with such junctions had the highest yield of any HTS junction configuration. Epitaxial superconducting Ba-K-Bi-O films were deposited and the gap energy was measured as a function of temperature in low-leakage S-I-N tunnel junctions. Epitaxial YBCO/insulator/YBCO trilayers using Sr-Ti-O and La-Al-O as the thick insulating layers were prepared and evaluated.

3. OBJECTIVES

The objectives of the Westinghouse-AFOSR program are:

- 1. Search for thin film superconductors with enhanced T_c 's and other superconducting properties.
- 2. Determine the fundamental lower limit of HTS rf surface resistance.
- 3. Investigate epitaxial multilayers, including Josephson junctions, incorporating deposited insulators and normal conductors with HTS films.
- 4. Develop materials and processing for alternative HTS devices.

4. ACCOMPLISHMENTS

4.1 PREAMBLE

The research reported here was performed under a Westinghouse-AFOSR Program which began February 20, 1991. The specific objectives of this program are listed in Section 3 of this report. These objectives are identical to the Tasks of the Work Statement. The overall objective is to investigate the fundamental physics and materials science of superconductors to enhance their properties for application in passive devices based on transmission line structures, active devices based on Josephson junctions, and novel active device structures based on the unique properties of superconductors.

4.2 ENHANCED SUPERCONDUCTING FILM PROPERTIES

The work on this task during the course of this program will be focussed on two distinct areas. The research in the first of these areas can be considered more speculative in that it is motivated by the hoped-for but unproven possibility that there exist even higher- T_c superconductors than those presently known. Using thin film growth methods the effort will be made to find some of these unknown materials. The research in the second area has a more modest but also a more immediately practical goal. In this case the effort will be directed toward improving properties of already existing superconducting materials thereby making them more useful in practical device applications.

During this reporting period, progress of a practical nature was made in the deposition of YBCO films. One of the most significant was identifying the source and finding means for the elimination of second phase Cu-O growth. It has been observed by many workers that YBCO films deposited either by off-axis sputtering or by laser ablation frequently contain Cu-O particles protruding from an otherwise smooth surface. Viewed with a scanning electron

microscope, these ~0.5-1 micron particles resemble boulders covering an otherwise level landscape. The presence of such "boulders" can have an extremely deleterious effect on the properties of Josephson junctions and on other structures requiring a thick epitaxial-film insulator. If present in large quantities, they can also degrade the performance of microwave devices.

Based on data collected from three different sputtering systems, it has been determined that these boulders form when the epitaxial growth of the YBCO is in some way inhibited. When this occurs, the Cu-O phase grows in preference to YBCO. There are a number of causes which can and do inhibit good epitaxy in YBCO films. These include structural or chemical defects in the single crystal substrates, foreign matter on the surfaces, and non-uniform growth temperatures. The latter cause was found to be particularly important when growing large-area films which are heated primarily by radiation rather than by conduction. One of the most significant sources of foreign matter on the substrate surface was found to be the backsputtering of material from the hardware immediately surrounding the substrates. The diminution of boulder formation from this source was accomplished by instituting two modifications in the sputtering process: (1) shielding of the hottest parts of the substrate holder/heater assembly from the substrates and (2) the application of a negative bias voltage to a conductive ring around the substrates which excluded the plasma from that vicinity. These two procedures were found to affectively minimized the backsputtering of material toward the substrates.

Before these solutions were found, only the second sides of the largearea films grown on both sides of two-inch diameter substrates were consistently free of Cu-O boulders. The reason for this result is now understood. Because of the radiation absorbed by the film deposited on the back side of the substrate and since its large area completely covered the substrate holder, the substrate surface had both a uniform temperature and the hottest temperature in the entire deposition region. Apparently the high temperature was sufficient to cause any backsputtered material to deposit preferentially on the somewhat cooler surfaces adjacent to the substrate rather than on the substrate itself.

All of these and other experimental results which led to the understanding and solution of the problem of boulders are discussed in more detail in an extensive review of the deposition of YBCO by sputtering which was published in the October 15, 1991 issue of the <u>Journal of Applied Physics</u>.

Some representative films which were completely free of boulders as determined by scanning electron microscopy were sent to NIST in Boulder for study in a scanning tunneling microscope (STM) for comparison with films made at other laboratories by a variety of techniques. The Westinghouse films were found to be the smoothest of all the films studied. The films were observed to have grown with a spiral patterns centered on screw dislocations approximately 0.1 μ m apart.

As alluded to above, another improvement in YBCO film properties stemmed from the use of a new substrate holder which was designed to heat the substrates predominantly by radiation. Previous designs employed a metal block to which the wafer was bonded with silver paint. The new design permitted the routine deposition of large-area films with more uniform properties than had been obtained with bonded substrates. This new technique eliminated the breakage associated with the removal of wafers silver-pasted to a block and it also permitted deposition of YBCO on both sides of polished wafers. The first report of low rf surface resistance films on both sides of large-area substrates made with this new holder was presented at the ICMC and has been accepted for publication in the proceedings.

The search for new materials with superconducting properties superior to presently available compounds was begun during this period. Experiments were performed to synthesize Ca-doped YBCO films (10% of the Y replaced by Ca) with a 1:2:4 structure and composition. This material was discovered in 1990 in bulk form and found to have a T_c of 90K. In contrast to 1:2:3, the 1:2:4 structure is not susceptible to oxygen depletion. Thus far, however, a negligible effort has been made to grow films – the preferred form in which to measure applications-related properties, such as rf surface resistance and critical current density, for comparison with the 1:2:3 structure.

Films were deposited by off-axis sputtering from a $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_7$ target. Only mixed-phase films with the 1:2:3 and 1:2:4 compositions were

prepared. These were obtained by first growing a new metastable precursor phase (non-superconducting) characterized by a 7.0 angstrom lattice constant at 450°C and then post-annealing at 780°C. Another approach for obtaining single=phase films is to follow the bulk synthesis route which would include annealing in a high oxygen pressure amorphous films of the correct composition. To do this annealing, however, requires a collaborator who has a suitable high-pressure furnace. Such a collaborator is being sought.

The search for higher T_c superconductors, begun in this period, focussed on the study of the the effect of incorporating small quantities of a fourth element into the YBCO structure with the hope of stabilizing a higher- T_c phase. A series of experiments were performed in which up to 5 at. % of either calcium or praseodymium was added into the deposited films. These types of experiments have previously been performed by workers using bulk samples. The rationale here is that because of the unique capability of thin film growth methods to form metastable phases it might be possible to prepare a material as a thin film that would not be c'able in bulk form. There is no evidence in the work completed to date, however, that this in fact occurred.

Another area studied was the critical oxidation step during which the high- T_c YBCO orthorhombic structure is formed. As has been reported previously, it was found that molecular oxygen is not an adequate oxidizing agent for forming this high- T_c structure and that a stronger agent such as water vapor is required. Some of YBCO films were annealed at much higher water pressures than used during film deposition to determine the effect of this treatment on film properties. The most interesting result was obtained for films held for one hour in 2 Torr of water vapor at 700°C. These films exhibited a large decrease in room-temperature resistivity from 300 to 200 micro-ohm-cm and an increase in resistivity ratio (300K/100K) from 3 to 4. Despite their significantly increased metallic character, however, T_c did not increase. Possible changes in other superconducting properties have not yet been determined.

4.3 RF SURFACE RESISTANCE

The rf surface resistance, R_s , of superconducting films was measured using a low-Q parallel-plate resonator with a resonance at approximately 8 GHz. This technique is now routinely used to evaluate films developed for this program. This low-Q parallel-plate resonator method is the only one presently available in this frequency range which is capable of measuring R_s of highquality YBCO films over a large temperature range without losses from the measurement apparatus dominating the measurement.

Films made by sputtering, laser ablation, and MOCVD in other laboratories were measured for comparison with Westinghouse films. None of the films to date have R_s as low as those grown by off-axis sputtering here. The attempt is now being made to correlate the various R_s measurements with surface morphology measured by STM at NIST. A preliminary conclusion is that the surface resistance scales with the density of imperfections in the film due to voids, non-superconducting particles, or misoriented grains of YBCO.

A series of R_s measurements was performed on films which were exposed to water vapor to evaluate long-term environmental effects on R_s . Surprisingly, there was no degradation of R_s as a function of exposure, even after the films were immersed in water. These experiments left open the possibility that atmospheric degradation may have occurred but was complete before the first R_s measurement was performed – perhaps within minutes after the films were exposed to the atmosphere. To investigate this possibility, R_s was measured on films which were encapsulated in situ with either gold or epitaxial LaAlO₃ layers. So far, the encapsulating layers have been found to only increase the net surface resistance. These results were presented at the IEEE Microwave Theory and Techniques Symposium.

4.4 EPITAXIAL FILMS AND LAYERED STRUCTURES

Thick epitaxial insulating layers are required in the fabrication of a number of devices to permit integration of ground planes and crossovers in active circuits, fabrication of lumped-element capacitors, and implementation of flux transformers. Insulating film requirements were estimated for each of these applications. Epitaxial bilayers and trilayers of YBCO/insulator and

YBCO/insulator/YBCO were fabricated with $SrTiO_3$, $LaAIO_3$, and MgO as the insulator. Vertical transport and capacitance measurements were made to obtain values for the resistivity and dielectric constant of the insulator. The highest resistivity was found for $SrTiO_3$ insulators – higher than any value reported in the literature for an epitaxial insulator on an HTS film and sufficiently high for any application. The problem with $SrTiO_3$ is its high real and imaginary dielectric constants which slow and attenuate signals.

The resistivity of epitaxial LaAlO₃ films was four orders of magnitude lower than that of SrTiO₃ and just marginally acceptable for lumped-element capacitors and digital circuits. In contrast to the smoothness of SrTiO₃ films, LaAlO₃ films were rough and, in the case of trilayers, the roughness was transferred to the top YBCO layer. Epitaxial MgO overlayers had the worst morphology and the lowest resistance in vertical transport measurements. Smooth YBCO film surfaces free of precipitates were critical for this work. It was found that insulating overlayers grown on YBCO with CuO precipitates on the surface had four orders of magnitude lower resistivity than for those grown on smooth YBCO. These results were discussed at the DARPA Workshop on High-T_c Superconductors. Several new insulating materials have been identified as promising candidates. They have acceptably low dielectric constants, a better lattice match to YBCO than MgO, and greater structural stability than LaAlO₃. Sputtering targets of two of these, NdGaO₃ and YAlO₃, have been ordered to permit comparison of these materials with the three already tested.

Superconducting $Ba_{1-x}K_xBiO_3$ (BKBO) is the highest T_c compound to have isotropic properties and for which an unambiguous gap voltage has been observed in tunneling measurements. Study of this material thus could produce important insights into the many problems that have been encountered with tunneling into anisotropic oxides such as YBCO. The development of a BKBO thin-film Josephson tunnel junction technology, however, has itself been hampered by the difficulty of making high quality films due to the high volatility of potassium.

The problem of K loss was addressed in this program by three different techniques: co-sputtering BKBO films from a stoichiometric $Ba_{0.6}K_{0.4}BiO_3$ target and a KO_2 target, sputtering from a stoichiometric target

in a flux of evaporated KOH, and sputtering from a K-rich target. The best results were obtained with the K-rich target. Epitaxial and superconducting films were obtained on SrTiO₃(110), LaAlO₃(100), MgO(100), and α -Al₂O₃(1T02) substrates. All films had smooth, featureless surfaces up to a magnification of 10,000X. Tunneling measurements were performed using normal-metal Ag as the counterelectrode in which the temperature dependence of the superconducting gap was measured. The BCS coupling parameter, $2\Delta/k_{\rm B}T_{\rm c}$ was 4.1. These results were presented at the Fall Meeting of the Materials Research Society.

Since one of the goals of synthesizing and studying BKBO is to fabricate all-BKBO tunnel junctions, a study was begun of the film surface which forms upon exposure to air. This surface may form a natural barrier. RHEED patterns were obtained from the surfaces of BKBO films indicating that the perovskite structure was present in the surface layer. Measurements of composition by XPS indicated that, despite its volatility, K was present. The surface layer consisted of carbon compounds, presumably K and Ba carbonates. This layer could be removed by annealing in pure O_2 prior to deposition of an artificial tunnel barrier. Candidate materials for the artificial barrier would have to have a structural and lattice match to BKBO to support epitaxial growth of the top BKBO electrode. Results of the surface preparation and analysis work are scheduled for presentation at the March Meeting of the American Physical Society.

EDS and x-ray data were obtained which show that a Y-Ba-Cu-O compound can be grown which has a similar c-axis structure and the same cation composition as YBCO but is not superconducting. This material was grown by using a YBCO target whose oxygen content had been depleted by long sputtering in pure argon. Efforts are now underway to grow this material in a controlled and reproducible way for potential use as a barrier layer.

Non-superconducting Y-Ba-Cu-O films grown under standard conditions except at reduced temperatures are also being studied as a possible barrier layer for junction use. YBCO deposited on a 150 angstrom thick Y-Ba-Cu-O layer grown at 500°C on LaAlO₃ showed a near-optimum critical temperature of

85K. X-ray diffraction data indicated that this multilayer film had grown with c-axis epitaxy and had no random components.

4.5 MATERIALS AND PROCESSING FOR HTS DEVICES

The integration of ferroelectrics with superconductors offers new opportunities for the development of non-volatile memory. Epitaxial ferroelectric films are desirable both to optimize the anisotropic polarization and to extend the lifetime of the memory cell. In the first work of this kind, ferroelectric bismuth titanate (BTO) films were successfully grown by laser ablation on YBCO films with and without SrTiO₃ buffer layers. The SrTiO₃ buffer layer permitted higher BTO deposition temperatures (up to 640°C) to be used without degrading the properties of the YBCO. The high deposition temperatures was desirable since the BTO grew epitaxially only at temperatures greater than 600°C. Both YBCO and BTO have perovskite-related structures with a lattice mismatch of approximately 1%. Capacitance hysteresis loops were measured at 77K which determined the quality of the ferroelectric. Results were presented at the 3rd International Conference on Integrated Ferroelectrics and will be published in the proceedings in the journal, <u>Ferroelectrics.</u>

Several different configurations for YBCO Josephson junctions are being developed at Westinghouse under various programs. One of the configurations which is being investigated under this program is a step-edge S-N-S junction in which a thin YBCO film is made discontinuous by covering a sharp step in the substrate which is high compared to the film thickness. The YBCO banks are then connected by a much thicker normal metal. The yield of these junctions has been remarkably high when gold, deposited in situ, was used as the normal metal layer. Modulation of the Josephson current was observed up to 65K.

Surprisingly, the I_cR_n products of these junctions degraded when the junctions were annealed at 400 and 600°C, in contrast to the improvements in contact resistance typically observed between gold and YBCO upon annealing. Scanning electron microscopy of cross-sections of the steps provided the explanation. Steps milled in the LaAlO₃ substrates initially used for this work

were well-defined and reproducible until the substrate was cycled to temperatures near or above the LaAlO₃ cubic-to-rhombohedral structural transformation. After cycling, the steps were found to have changed height, angle, and even position on the wafer. Subsequently, NdGaO₃ substrates were used for this work. These results were presented at an ONR/DARPA workshop on Substrate Materials for High-T_c Superconductors.

Junctions made by this technique were used to fabricate flip-flop circuits under another Westinghouse program. This program will continue to use this technique as a way of evaluating both Josephson junctions and interfaces between YBCO and normal metals. The emphasis will now be placed on higher resistivity normal metals such as epitaxial layers of over-doped $La_{2-x}Sr_xCuO_4$. Films of LSCO with x = 0.3 have been grown which are not superconducting but which have a lower normal-state resistivity than superconducting LSCO (x = 0.17). This result is in agreement with that for bulk samples which must be fabricated under high pressures (100 bar O_2) to get the excess Sr to remain in the LSCO structure. The results of junctions fabricated with LSCO normal metals are scheduled for presentation at the March Meeting of the American Physical Society.

4.6 OTHER ACCOMPLISHMENTS

In support of the work described above, a file of reprints and preprints on high-T_c superconductivity started in 1987 was maintained. The updated list of papers was entered into a computer and keywords were assigned to them to aid in retrieval. The entire database has been made available to the research community through the computer facilities at High-T_c Update. The 1991 additions to the database will be published in the <u>Journal of Superconductivity</u>.

The first half of 1992 marks the completion of a three-year. \$4M upgrade of facilities at Westinghouse dedicated to superconducting electronics. This program will now have the benefit of new equipment for the deposition, patterning, and characterization of superconducting films and circuits. The number of vacuum deposition systems dedicated to HTS film structures will have increased from two at the start of the program to seven by the middle of 1992. The most important new equipment is a dedicated ion miller with

capability for up to a 6"-diameter wafer. This equipment is located in a superconducting electronics clean room is now double its previous area. The most significant new characterization tool is a state-of-the-art four-circle x-ray diffractometer.

To obtain optimum use of the unique material and measurement capabilities developed under the present and previous AFOSR-Westinghouse programs, collaborations with other research institutions have been continued and expanded. These collaborations are made with researchers whose work falls within the overall objectives of this program. A table of technical collaborations which were active in 1991 is shown in Section 7 of this report.

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5. PUBLICATIONS

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- 5. T. T. Braggins, J. R. Gavaler, and J. Talvacchio, "In-Situ Deposition of YBaCuO Films on Both Sides of Two-Inch-Diameter Wafers by Off-Axis Sputtering," accepted for Proc. ICMC, <u>Advances in Cryogenic Engineering</u> (<u>Materials</u>), (Plenum, New York, 1991).
- 6. J. Talvacchio, S. H. Talisa, and G. R. Wagner, "High-T_c Film Development for Electronic Applications," Microwave Journal, 105 (June, 1991).
- 7. D. H. Kim, D. J. Miller, J. C. Smith, R. A. Holoboff, J. H. Kang, and J. Talvacchio, "The Effects of Microstructure on Flux Pinning in Epitaxial YBa₂Cu₃O₇ Films," Phys. Rev. B <u>44</u>, 7607 (1991).
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- M. M. Driscoll, J. T. Haynes, S. Horwitz, R. A. Jelen, R. W. Weinert, J. R. Gavaler, J. Talvacchio, G. R. Wagner, K. A. Zaki, and X.-P. Lang, "Cooled, Ultra-high Q, Sapphire Dielectric Resonators for Low Noise Microwave Signal Generation," Proc. 45th IEEE Symposium on Frequency Control, 700 (1991); and IEEE Trans. Ultrasonics, Ferroelectrics, and Frequency Control (1992).

6. PERSONNEL

- J. R. Gavaler
- J. Talvacchio

Principal Co-Investigators

- M. G. Forrester
- J. H. Kang
- G. R. Wagner
- B. A. Baumert (Carnegie Mellon University)
- J. D. McCambridge (Yale University)

7. COUPLING ACTIVITIES^{*}

- 1. "The Effect of Uniaxial Strain on J_c and T_c of thin YBaCuO Films," <u>G. L. Belenky</u>, S. M. Green, S. J. Hagen, R. L. Greene, M. G. Forrester, and J. Talvacchio, Contributed presentation to the American Physical Society March Mtg., March 1991.
- 2. "Heterostructures Formed with High Temperature Superconductors," J. Talvacchio, Invited presentation to the Materials Research Society Spring Meeting, Anaheim, April 1991.
- 3. "Superconducting Materials and Electronics Research at Westinghouse STC," <u>M. G. Forrester</u>, University of Maryland Physics Dept. Colloquium, April 1991.
- Pulsed Laser Deposition of Oriented Bismuth Titanate Films for Integrated Electronic Applications," <u>H. Buhay</u>, S. Sinharoy,
 M. H. Francombe, W. H. Kaser, J. Talvacchio, B. K. Park, N. J. Doyle,
 D. R. Lampe, and M. Polinsky, Contributed presentation to the 3rd International Conf. on Integrated Ferroelectrics, Colorado Springs, April 1991.
- 5. "Electronic Applications of Superconductors," J. Talvacchio, Invited presentation to the Pittsburgh Chapter of the IEEE Magnetics Soc., Pittsburgh, May 1991.
- 6. "In-Situ Deposition of YBaCuO Films on Both Sides of Two-Inch-Diameter Wafers by Off-Axis Sputtering," <u>T. T. Braggins</u>, J. R. Gavaler, and J. Talvacchio, Contributed presentation to the International Cryogenics Materials Conf., Huntsville, June 1991.
- 7. "Present and Projected Performance of High-Temperature Superconducting Filters," <u>S. H. Talisa</u>, M. A. Janocko, J. Talvacchio, and C. Moskowitz, Contributed presentation to the IEEE Microwave Symposium (MTT-S), Boston, June 1991.
- 8. "Sputtered YBCO Film Structures for Microstrip and S-N-S Devices," J. Talvacchio, Invited presentation to the Third Annual DARPA HTSC Workshop, Seattle, October 1991.

^{*}Speaker's name is underlined.

- "Cooled, Ultra-high Q, Sapphire Dielectric Resonators for Low Noise Microwave Signal Generation," <u>M. M. Driscoll</u>, J. T. Haynes, S. Horwitz, R. A. Jelen, R. W. Weinert, J. R. Gavaler, J. Talvacchio, G. R. Wagner, K. A. Zaki, and X.-P. Lang, Invited presentation to the 3rd DARPA HTSC Workshop, October 1991.
- 10. "Current Status of Single-Target Sputtering of YBCO," J. R. Gavaler, Invited presentation to the Fall Meeting of the Materials Research Society, Boston, December 1992.
- 11. "Surface Properties and Microstructure of Sputtered Ba-K-Bi-O Films," <u>B. A. Baumert</u> and J. Talvacchio, Contributed presentation to the Fall Meeting of the Materials Research Society, Boston, December 1992.
- "Pulsed Laser Deposition and Characterization of Epitaxial Bi-Ti-O/YBCO 'Ferroic-Superconductor' Structures," <u>H. Buhay</u>, S. Sinharoy, J. Talvacchio, W. H. Kasner, N. J. Doyle, and M. H. Francombe, Contributed presentation to the Fall Meeting of the Materials Research Society, Boston, December 1992.
- 13. "Requirements for Substrates and Deposited Insulators Used in Analog and Digital HTS Circuits," J. Talvacchio, Invited presentation to the Workshop on Substrate Materials for High Temperature Superconductors, Williamsburg, February 1992.

OUTSIDE COLLABORATIONS

Institution/Collaborator	Effort / Special Requirements
Argonne National Laboratory	Vortex dynamics in superconductors
Dr. D. H. Kim	- epitaxial YBCO and NbN patterned films
University of Maryland	Strain effects in YBCO
Prof. C. J. Lobb	- patterned YBCO bridges
NIST (Boulder)	Scanning tunneling microscopy
Dr. A. Roshko	- YBCO films with measured R _s
EMCORE Corp.	rf surface resistance measurements
Dr. P. Norris	- YBCO films grown by MOCVD
Carnegie Mellon University	Flux pinning and creep in BKBO
Prof. M. E. McHenry	- Epitaxial BKBO films
University of Florida Prof. D. Tanner	Infrared reflection and absorption - single crystal LSCO films
Yale University Prof. D. E. Prober	S-N-S Josephson junction development - in-situ S-N interface formation
CVC Products, Inc.	rf surface resistance measurements
Dr. P. Ballentine	- 2" dia. YBCO films
Carnegie Mellon University	Low-field microwave absorption
Prof. N. S. VanderVen	- Low-R _s YBCO films
Northeastern University	Calibration of new R _s meas. technique
Prof. C. Vittoria	- YBCO with measured R _s

8. PATENTS

1. N. B. Singh and J. Talvacchio, "Thin-Film Growth of Thallium-Based Superconductor," Disclosure No. RDS 91-027, disclosed April 1991.