②

WL-TR-93-5031

EXPLORATORY RESEARCH FOR A HIGH TEMPERATURE SUPERCONDUCTING INTEGRATED CIRCUIT

E. K. Track & O. Mukhanov
Hypres Inc., 175 Clearbrook Rd., Elmsford, N. Y. 10523

J. N. Eckstein, I. Bozovic, & G. F. Virshup
Varian Associates, Palo Alto CA 94304-1025

Z. -X. Shen, & W. E. Spicer
Stanford University, Stanford CA 90305-4055

September 1993

Final Report for Period July 1992 - Aug. 1993

Approved for public release; distribution is unlimited.

Solid State Electronics Directorate
Wright Laboratory
Air Force Materiel Command
Wright-Patterson Air Force Base, OH 45433-7331

42-pg

NOTICE

When Government drawings, specifications, or other data are used for any purpose other than in connection with a definitely Government-related procurement, the United States Government incurs no responsibility or any obligation whatsoever. The fact that the government may have formulated or in any way supplied the said drawings, specifications, or other data, is not to be regarded by implication, or otherwise in any manner construed, as licensing the holder, or any other person or corporation; or as conveying any rights or permission to manufacture, use, or sell any patented invention that may in any way be related thereto.

This report is releasable to the National technical Information Service (NITS). at NITS, it will be available to the general public, including foreign nations.

This technical report has been reviewed and is approved for publication.

Joseph E. Brandelik
JOSEPH E. BRANDELIK, Sr. EE
E-O Techniques & Appl. Branch
Electro-Optics Division

John O. Crist
JOHN O. CRIST, Actg. Chief
E-O Techniques & Appl. Branch
Electro-Optics Division

C. Richard Lane
C. RICHARD LANE, Actg. Chief
Electro-optics Division
S. S. Electronics Directorate

If your address has changed, if you wish to be removed from our mailing list, or if the addressee is no longer employed by your organization please notify WL/ELOT, WPAFB, OH 45433-7331 to help us maintain a current mailing list.

Copies of this report should not be returned unless return is required by security considerations, contractual obligations, or notice on a specific document.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
93 Sept

3. REPORT TYPE AND DATES COVERED
Final 07/01/92--08/01/93

4. TITLE AND SUBTITLE
EXPLORATORY RESEARCH FOR A HIGH
TEMPERATURE SUPERCONDUCTING
INTEGRATED CIRCUIT

5. FUNDING NUMBERS
C F33615-90-C-1456
PE 65502
PR 3005
TA 65
WU 02

6. AUTHOR(S)
E.K. TRACK & O. MUKHANOV, J.N. ECKSTEIN,
I. BOZOVIC, & G.F. VIRSHUP, Z.-X. SHEN
& E. SPICER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
HYPRES, INC.
VARIAN ASSOCIATES, INC.
STANFORD UNIVERSITY

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
SOLID STATE ELECTRONICS DIRECTORATE
WRIGHT LABORATORY
AIR FORCE MATERIEL COMMAND
WRIGHT-PATTERSON AFB OH 45433-7331

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
WL-TR-93-5031

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
APPROVED FOR PUBLIC RELEASE
DISTRIBUTION IS UNLIMITED

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

The objective of this effort was the exploration investigation of the molecular-beam-epitaxy-trilayer Josephson Junction process under development by VARIAN Corporation. Under this effort, Stanford University provided fundamental materials characterization to understand and improve the surfaces and interfaces of the thin film structures. HYPRES Inc. provided an independent assessment of the junctions produced by Varian and addressed the possibilities of rapid single fluxquantum (RSFQ) circuit designs. The material system chosen for this investigation was Bismuth-Strontium Calcium-Copper-Oxide (BSSCO). The Josephson Junction character of the devices was confirmed by the observation of microwave induced (Shapior) steps in the I-V curves. Contract resistance by three orders of magnitude by modulation doping the top few molecular layers of the upper superconductive electrode. The desired properties to warrant RSFQ circuit fabrication were not obtained. A material system with a higher Josephson Junction critical temperature and higher critical current is necessary for circuits.

14. SUBJECT TERMS
Microwaves Superconductors Josephsons junctions

15. NUMBER OF PAGES
42

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT
UNCLASSIFIED

18. SECURITY CLASSIFICATION OF THIS PAGE
UNCLASSIFIED

19. SECURITY CLASSIFICATION OF ABSTRACT
UNCLASSIFIED

20. LIMITATION OF ABSTRACT
UNLIMITED

TABLE OF CONTENTS

This report contains an executive summary containing the main elements of the program and its conclusions. This is followed by three appendices detailing specific aspects of the program. The results presented in this report were fully funded by the advanced research projects agency, ARPA/DSO.

Appendix A covers the film and junction fabrication efforts at Varian.

Appendix B contains a discussion of the requirements for the implementation of rapid single flux quantum circuits with high temperature superconductor trilayer structures, such as the Varian process.

Appendix C involves the surface characterization efforts at Stanford University.

Executive Summary

Motivation and Goals

This program is an extension to a phase II SBIR which examined optically modulated superconducting delay lines. The phase II program was a theoretical and experimental investigation which defined the issues and the necessary parameters for the successful implementation of long optically modulated superconducting delay lines and their application as variable phase shifters for use, for example, in phased array radar. In these applications, it is important to provide switching and multiplexing elements in integrating a useful subsystem. The best approach is to implement these active elements using superconducting technology. This opens the possibility of monolithic integration of the subsystems in a compact package which can be efficiently cryocooled. In addressing this issue, the goal of this program is to explore the potential of molecular-beam-epitaxy trilayer Josephson junction process, being developed by the VARIAN Corporation, to be used for the implementation of high temperature superconductor (HTS) circuits designed by

HYPRES. Stanford University provides fundamental materials characterization to understand and improve the surfaces/interfaces of the thin film structures. The elements of the program are as follows:

1) The development of the Varian MBE process so that junction characteristics suitable for fabrication of rapid-single-flux-quantum (RSFQ) circuits can be reproducibly fabricated. The focus is on exploring the uniformity and repeatability of the junction process and on engineering the barrier material so that the junction properties (critical current, normal resistance) can be tuned to the desired parameters. [Varian]

3) To provide an independent test of the Varian junctions produced and to address the issues and possibilities of RSFQ circuit designs for practical circuits based on the Varian process. [HYPRES]

3) The placement of a fundamental footing under the Varian process to understand the surface and interface properties of the thin film devices, and to address the problems of contact resistance and proximity effect. [Stanford University]

Program Activities and Results

The material chosen in this program is the superconductor BSSCO (Bismuth-Strontium-Calcium-Copper-Oxide) in the atomic stoichiometry ratio 2212 for Bi, Sr, Ca, Cu, and O. The transition temperature of this material is around 80 K, and Varian has under other programs set-up an MBE system for the deposition of HTS films of this material. In addition, Varian has developed the capability of forming trilayers of this material incorporating a base- and a counter-electrode "sandwiching" a barrier layer produced by changing the composition and/or stoichiometry of the 2212 compound to produce an insulating or a non-superconducting phase. The focus of the program was on engineering this barrier material in order to tune the junction parameters: critical current, I_c and normal state resistance R_n.

Based on calculations and projections done by HYPRES, the target I_c for reliable operation of HTS RSFQ circuits require an I_c of over 0.5 mA at a temperature of 40 ^{0}K. Another parameter relevant to the circuits is the I_cR_n product, which should be ≥ 0.5 mV. In addition to engineering the barrier material, another problem was addressed: the problem of high contact resistance of the material.

The following progress was achieved by Varian on these issues:

A total of 54 depositions of superconducting Bi-Sr-Ca-Cu-O films were performed. The films that showed surfaces predominantly free of second-phase defects were processed into four terminal devices. These were tested by current-voltage (I-V) characteristics and resistance-versus-temperature (R(T)) measurements, with and without external microwave radiation or magnetic field. The results are as follows:

1. Deposition procedures, including the specific shuttering sequence have been developed which provide films with clean, defect-free surfaces over most of the wafer. A good yield of Josephson junctions has been achieved in some of the films. Josephson junction character of the devices was confirmed by clear microwave-induced (Shapiro) steps observed in the I-V characteristics.

2. The problem of high contact resistance has been solved by appropriate modulation doping in the top few molecular layers of the upper Bi-Sr-Ca-Cu-O electrode. With this technique, the contact resistances were reduced from as much as thousands of Ω's to less than 1 Ω, over the area of about 10^{-6} cm^2.

3. A new barrier design has been developed with which we have obtained I_cR_n of 5-7 mV at 4.2 K. This is an order-of-magnitude improvement over what is achieved with most other types of HTSC Josephson junctions.

4. Within the limited statistics currently available with the new barrier designs, we have also observed an order of magnitude decrease in variability of junction properties such as I_c and R_n.

HYPRES has set-up a measurement probe equipped with magnets and rf ports for the testing of the Varian junctions. The measurements that can be implemented include current-voltage characteristics, rf response, and magnetic field modulation for both axial and transverse magnetic fields. We used the set-up to retest Varian samples. Two sample batches were tested. The first batch involved earlier samples containing junctions with critical currents below 50 µA at 4.2 °K. The results of this initial test indicated a decrease of more than 50% in the critical current density of the junctions since their initial evaluation at Varian. No significant rf response or dc magnetic field modulation was observed. The second batch included two chips (No. 905 and No. 908) each containing several devices. For this batch, all devices tested showed critical currents and normal state resistances close to those determined previously at Varian, so these junctions successfully underwent temperature cycling, storage and transport. Magnetic fields sufficient to produce in excess of 100 flux quanta (ϕ_0) in the junctions were applied. No significant magnetic modulation was observed on any of the devices. Device 908.10 had the largest modulation of the critical current -- approximately 5%. No response to 12 GHz microwaves was observed.

The effort in surface/interface characterization at Stanford University centered on establishing baseline X-ray photoelectron spectroscopy spectra for the BSSCO material by measuring single crystals, and then comparing the spectra of the Varian BSSCO thin films to this baseline. In addition, an investigation of the surface stability of BSSCO and its interaction and proximity effect with Au, Ag, Bi, and A_xC_{60} was carried out. The thin film surface characterization yielded a technical breakthrough by developing a technique to cleave the thin films insitu (i.e., without breaking vacuum). The spectra of these thin films compared well with the spectra from the bulk material and demonstrated the first direct observation of the gap of BSSCO through this technique. The method also demonstrated the good single crystalline quality of the Varian BSSCO thin films. The interface studies indicated that no reaction and no proximity effect is observed with Au and Ag overlayers on BSSCO. This is attributed to the high electron density of these materials. Bi and A_xC_{60} on the other hand are low electron density materials, but were found to be too reactive with the BSSCO surface to produce a good interface for proximity effects.

Conclusions

This program has demonstrated the potential for the Varian MBE process to control the properties of HTS Josephson junctions through atomic layer-by-layer engineering. Although the independent measurement of the devices at HYPRES progressed to the point of demonstrating the stability and repeatability of the Varian junctions, the desired parameters for successful implementation of HTS RSFQ circuits were not obtained in this initial investigation. The process controls are nevertheless well understood, and continued research in this field is believed to lead to successful devices and circuits. The circuit process will need to incorporate two additional superconducting layers for ground plane and wiring. The design analysis of HTS circuits indicates the feasibility of implementing practical devices once this process is developed. The surface and interface characterization implemented under this program have demonstrated that X-ray photoelectron spectroscopy has the sensitivity required to analyze the superconducting properties of thin film surfaces and interfaces. Through this technique, and a uniquely developed thin film cleavage method, the energy gap of BSSCO thin films were unambiguously identified. The technique can be used in future programs to correlate the surface and interface properties of such films and junctions with their transport properties and the related characteristics of devices made with these films and junctions.

APPENDIX A

MOLECULAR BEAM EPITAXY GROWTH OF HTS FILMS AND JUNCTIONS

THE FINAL REPORT ON HYPRES/VARIAN SUBCONTRACT UNDER AFSC/WL CONTRACT F33615-90-C-1456 BY HYPRES PO 703768, ENTITLED "DEVELOPMENT OF A HIGH TEMPERATURE SUPERCONDUCTING INTEGRATED CIRCUIT."

J. N. ECKSTEIN, I. BOZOVIC, AND G. F. VIRSHUP

**Edward H. Ginzton Research Center,
Varian Associates, Inc., Palo Alto, Ca
94304-1025**

August 16, 1993

Overview

Under this subcontract, we have performed the total of 54 depositions of superconducting Bi-Sr-Ca-Cu-O thin films -- 5 in July 1992, 10 in August 1992, 12 in September 1992, none on October 1992 (machine was serviced), 3 in December 1992, 6 in January 1993, 13 in February 1993, 5 in March 1993, and 5 in April 1993. The films that showed surfaces predominantly free of second-phase defects were processed into four terminal devices. These were tested by I-V and R(T) measurements, with and without external microwave radiation or magnetic field. The principal results are as follows.

1. Deposition procedure, including the specific shuttering sequence, has been developed which provides films with clean, defect-free surfaces over most of the wafer. A good yield of Josephson junctions has been achieved in some of the films (e.g. No. 905 and No. 908). Josephson junction character of the devices was confirmed by clear microwave-induced (Shapiro) steps observed in I-V characteristics.

2. The problem of high contact resistances has been solved. By appropriate modulation doping in the top few molecular layers of the upper Bi-Sr-Ca-Cu-O electrode, we have succeeded in reliably reducing contact resistances from as much as thousands of ohms to less than 1 Ω, over the area of about 10^{-6} cm^2.

3. A new barrier design has been developed, with which we have obtained I_cR_n = 5-7 mV. This is an order-of-magnitude improvement over what is achieved with most other types of HTSC Josephson junctions.

4. Within the limited statistics currently available with the new barrier design, we have also observed an order of magnitude decrease in variability of junction properties such as I_C or R_n.

5. In order to achieve compatibility with Hypres testing facilities, a new device layout has been developed and implemented.

1. Introduction

Trilayer (or sandwich) Josephson junctions, in which a thin insulating barrier layer separates two superconducting layers, have been fabricated quite successfully with conventional, low-T_c superconductors. The principal advantage of this geometry is that, with good epitaxy and growth control, it should provide very reproducible and uniform junctions.

The key technological problem here is to ensure that the superconductor remains undeteriorated in the vicinity of the barrier. What constitutes "vicinity" turns out to depend on the material, i.e., on its superconducting coherence length, which in low-T_c superconductors is of the order of 10-100 nm. In the case of a high-T_c superconductor such as $Bi_2Sr_2CaCu_2O_8$ (or 2212, for brevity), the coherence length is much shorter, in particular in the *c*-axis direction (i.e., perpendicular to CuO_2 planes). Actually, in this material the superconducting slabs are more or less independent, so it will largely be the last molecular monolayer, the one closest to the barrier, that has to ensure that the order parameter is conveyed across the junction.

From the materials science point of view, this means that crystalline perfection has to be maintained on an atomic scale, in particular near the barrier. Furthermore, the barrier must be made very thin - comparable, perhaps, to one molecular layer of the superconductor - and structurally as perfect itself. Indeed, this is not an easy task; one has to deposit an insulating layer, merely 2-3 nm thick, without any pinholes over large, macroscopic areas.

Nevertheless, in what follows we will show that a state-of-the-art thin film deposition technique, specifically developed for this task, makes it possible to fabricate HTSC trilayer Josephson junctions with characteristics which are excellent and actually superior to those currently achieved with simpler techniques such as step-edge, bicrystal, or biepitaxial grain-boundary junctions.

2. Atomic-Layer-by-Layer Reaction-Controlled Epitaxy

Atomic Layer-by-Layer Reaction-Controlled Epitaxy (ALL-RCE) is a technique for synthesis of oxide superconductors, developed in the last several years [1-6] in the E. H. Ginzton Research Center. The hallmark of this technique is very accurate sequencing of atoms supplied to the growing crystalline surface, in order to kinetically control the chemical reactions occurring at the surface. Under the conditions we operate, only the top molecular layer is involved in the growth chemistry. Hence, precise sequences of molecular layers can be assembled into heterostructures such as multilayers or superlattices with little or no intermixing.

A schematic diagram of the ALL-RCE system is shown in Figure 1.

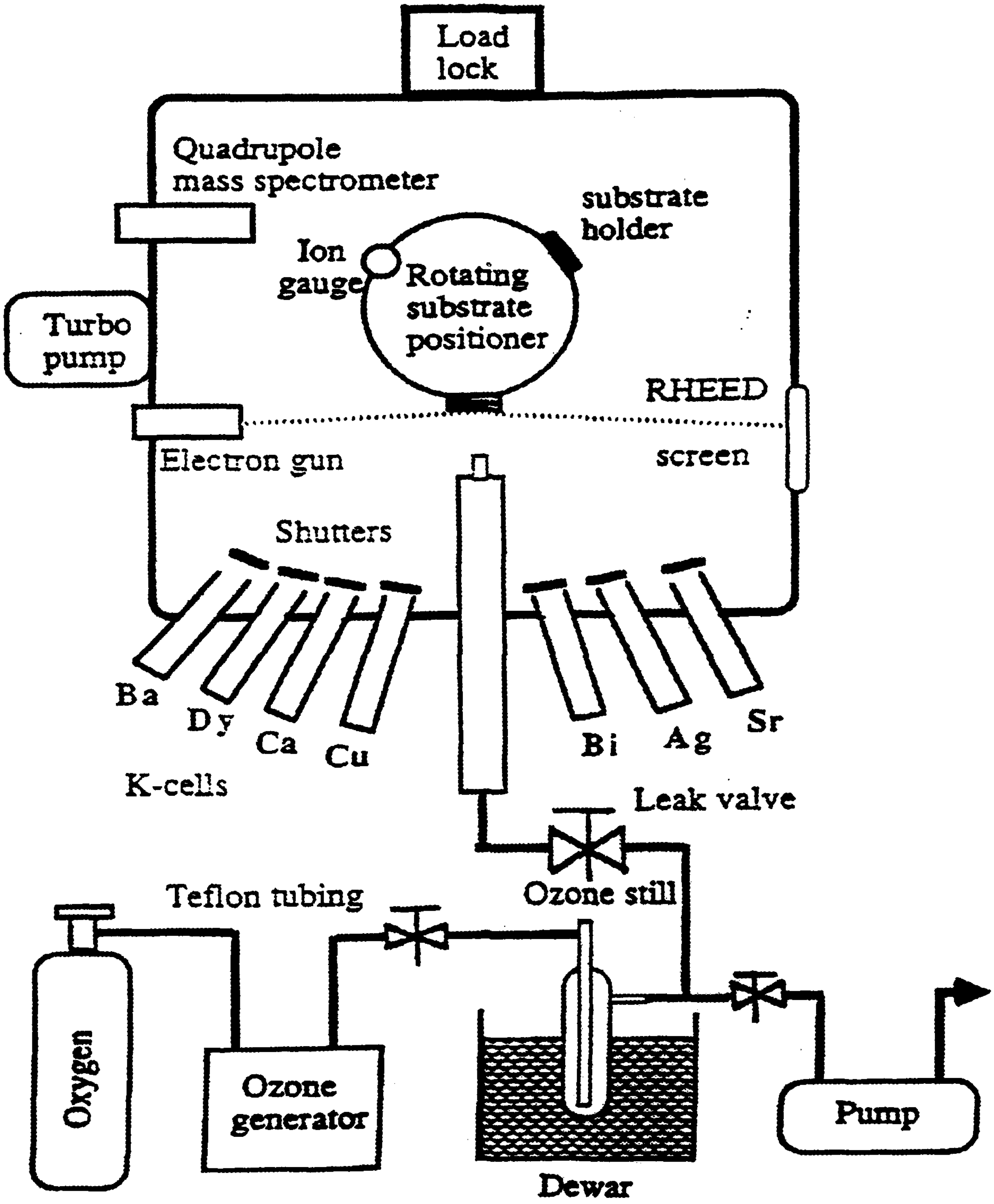


Figure 1: Schematic diagram of ALL-RCE growth system.

It contains individual Knudsen cells for each cation component of the film to be grown. These thermal evaporation sources provide atomic beam fluxes which are stable over periods of hours to within a few percent. Currently, seven sources are installed in the system: Bi, Sr, Ca, Cu, Dy, Ba, and Ag. This configuration permits the growth of both Dy-Ba-Cu-O compounds, and compounds from the Bi-Sr-Ca-Cu-O family. The Ag source is used for *insitu* deposition of electrical contacts. The sources have shutters which are computer controlled.

The atomic flux from each source is monitored to an absolute accuracy of better than one percent using a technique we have developed which is based on pseudo-double-beam atomic absorption spectroscopy [7]. In this technique, a mechanically chopped beam of light from a hollow cathode lamp passes in front of the substrate position, and is detected using a photo-multiplier tube and a lock-in amplifier. By opening and closing the shutter during the measurement, a pseudo-double-beam effect is achieved, which makes the measurement immune to drifts in reflection and absorption by the viewport windows, gain settings on amplifiers, and lamp emission intensity. The technique is sufficiently fast and accurate to detect and correct for changes in beam flux of less than one percent in real time. In order to relate the atomic absorption signal from each source to the actual beam flux, calibration films are grown under constant thermodynamic conditions, and the resulting surface density of atoms in the film is obtained by Rutherford Back Scattering (RBS) analysis.

Beam flux measurement and control by atomic absorption spectroscopy has proved critical to atomic-layer-by-layer growth of high quality HTSC thin films. In particular, we have found that atomically flat films free of second-phase defects can be obtained only if the layer composition does not deviate by more than one to three percent (depending on the specific phase and element in question) from the correct stoichiometry.

Oxidation is achieved using a pure ozone beam, which provides sufficient oxidation power even at relatively low fluxes.

The typical oxygen background pressure is about 10^{-5} Torr, which permits line-of-sight beam deposition and abrupt beam flux modulation using shutters. The temperature of the heated substrate is controlled to within 1°C using optical pyrometer.

The crystalline structure of the film surface is monitored *insitu* by reflection high energy electron diffraction (RHEED). It provides information, in real time, on the crystallographic microstructure (specifically, the lattice constants and the symmetry) of the surface, its epitaxial relation to the underlying film, and its flatness or roughness on an atomic scale.

3. Defect-free Bi-Sr-Ca-Cu-O films

We have performed several growth experiments aimed specifically at determining the optimal substrate temperature, the ozone pressure, and the stoichiometry, in order to minimize the density of second phase defects. Such defects are suspected to be one of the major causes of variations of the characteristics of trilayer Josephson junctions synthesized by ALL-RCE. We have tried to correlate the density of defects to the targeted material stoichiometry and temperature variations across the substrate surface. Further, the spatial distribution of defects relative to the orientations of the atomic beam sources has been studied. Finally, attempts were made to absorb such defects in early stage of their formation be intercalation of molecular layers of the low-T_c 2201 phase.

As the result of these studies, we were able to find a set of deposition parameters, including the specific shuttering sequence (i.e., the sequence in which different atoms are supplied to the growing surface), which provided films with clean, defect-free surfaces over most of the wafer.

4. Device Fabrication

The films were processed into four-contact vertical transport test structures by a combination of wet chemical etching (for device isolation), and ion milling. Sputtered SiO_2 was used as an isolation layer to bring the top counter-electrode layer contacts to large pads for probing. Ohmic contacts were formed by *insitu* evaporation of silver.

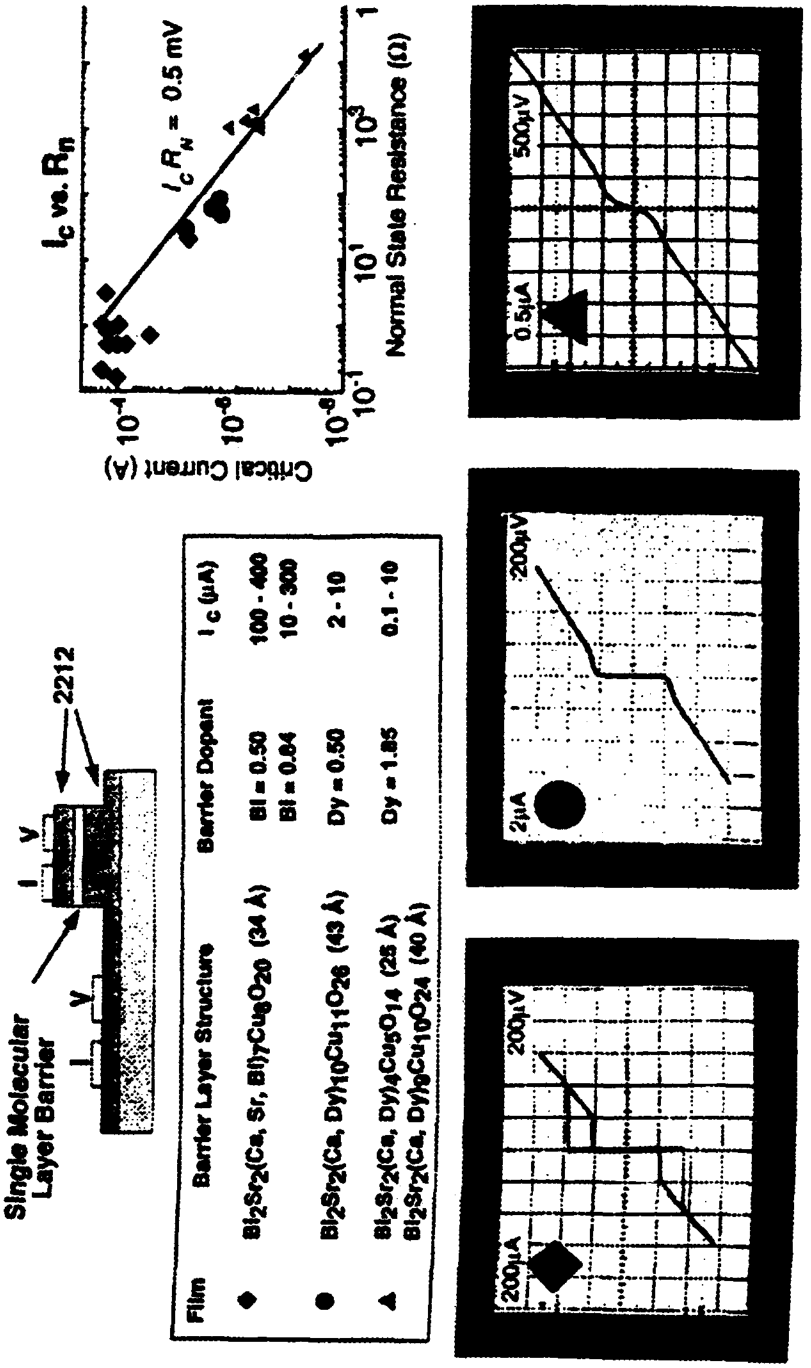
Despite this precaution, the contact resistances turned out to be unacceptably large in some of our earlier devices. We ascribed this to the loss of charge carriers in the few topmost 2212 layers, due to (i) oxygen depletion during the cool-down (notice that it is performed in vacuum - i.e., with ozone supply shut off - below about 500-600 ^{0}C), (ii) possible oxidation of silver in contact with 2212, and (iii) possible electron leakage and depletion from silver, leading to hole compensation in 2212.

To solve this problem, we have developed a method of modulation doping the few top-most layers of 2212, by omitting one Bi-O layer out of each 2212 molecular layer. In other words, we grow the last few molecular layers with 1212 stoichiometry and structure. This artificial, meta-stable compound has an increased hole density, and it provides for greatly reduced contact resistances - typically under 1 Ω, over the area of about 10^{-6} cm^2, at low temperatures.

In order to achieve compatibility with Hypres testing facilities, we have developed and implemented a new device layout. It provides simultaneous electric contacts to all the devices on a wafer and, thus, speeds up their testing considerably.

5. High T_c Trilayer Josephson Junctions

In Figure 2, we have summarized some of our data [8,9] on trilayer Josephson junctions manufactured by ALL-RCE. The top and the bottom electrodes consisted of $Bi_2Sr_2CaCu_2O_8$, with $T_c >$ 80 ^{0}K after device fabrication. The barrier consisted of a single molecular layer with the structure of $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$, with n between 5 and 11. Such phases are not thermodynamically stable, but nevertheless they can be grown by virtue of atomic layering.



Film	Barrier Layer Structure	Barrier Dopant	I_c (μA)
◆	$Bi_2Sr_2(Ca, Sr, Bi)_7Cu_8O_{20}$ (34 Å)	Bi = 0.50 Bi = 0.64	100 - 400 10 - 300
●	$Bi_2Sr_2(Ca, Dy)_{10}Cu_{11}O_{26}$ (43 Å)	Dy = 0.50	2 - 10
▲	$Bi_2Sr_2(Ca, Dy)_4Cu_5O_{14}$ (25 Å) $Bi_2Sr_2(Ca, Dy)_9Cu_{10}O_{24}$ (40 Å)	Dy = 1.85	0.1 - 10

Figure 2: Critical current versus normal state resistance for devices from nine growths. The results indicate that doping of barrier layers with Dy or Bi changes critical current and normal state resistance by over three orders of magnitude while maintaining nearly constant I_cR_n voltage.

In Figure 2, we have shown the device structure; the area of the active mesa was 30 μm by 30 μm. Few typical junction I-V characteristics are shown as well. Very clear microwave (Shapiro) steps (as seen in Figure 3) have also been observed in such devices.

In order to engineer the barrier properties, trivalent cations (Dy and Bi) were doped to selected Ca sites. ALL-RCE makes it possible to control the level doping within the barrier, to vary the local density of charge carriers and, thus, to control the barrier resistance. In this way, the junction critical current and normal state resistance were scaled by over three orders of magnitude, while their product remained nearly constant ($I_cR_n \approx 0.5$ mV).

For the most heavily doped barriers, resistance exceeded 10^5 Ω at low temperatures. This shows that there was no leakage due to pinholes or second phase inclusions, over a relatively large area of (30 μm)^2, although the barrier thickness was only 25 Å

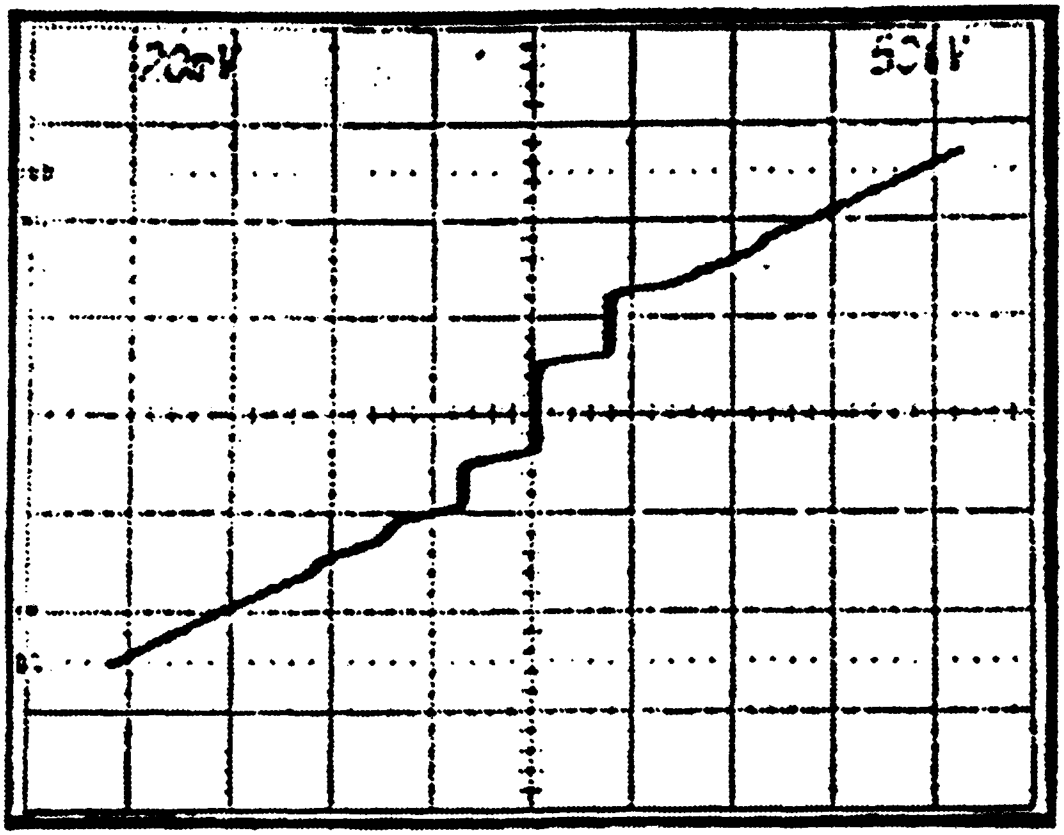


Figure 3. **Current-voltage characteristic of a trilayer Bi-Sr-Ca-Cu-O B Josephson junction illuminated with 17.9 GHz microwave radiation.**

6. New Barrier Design: Modulation Doping

Among other results, our own studies of transport in ultrathin films and dilute superlattices [10,11] have shown that 2212 superconductor is quasi-two-dimensional. We believe that the superconducting order parameter undulates along the crystallographic **c** direction, reaching its maximal value in CuO_2 planes and its minimum somewhere in the BiO layer region. In order to improve the superconducting coupling across the barrier, we have attempted to omit one out of two BiO layers on each side of the 2212 barrier. One could think of this as insertingf a 2212-1278-1212 sequence or equivalently a 2212-0278-2212 sequence, see Figure 4. Indeed, very high critical currents (ca. 10^4 A/cm^2) have been observed in such structures - over two orders of magnitude more than what we typically observe with a similar 2278 barrier. We interpret this result as the consequence of modulation doping in the barrier, which is made more metallic and actually superconducting.

In order to reduce the critical current, we have doped such 0278 barriers with Dy (substituting for 7-30 mol % of Ca). That reduces the critical current down to a usable level, say <1 mA, in devices with the cross-section of about 10^{-5} cm^2. More importantly, it provides I_cR_n = 5 -7 mV, which is an order-of-magnitude improvement over previous state-of-the-art (also our own) trilayer junctions. It is also higher than what is achieved with most other types (e.g. step-edge, bicrystal, biepitaxial, etc.) of HTSC Josephson junctions.

Another important aspect of stronger superconducting coupling across the barrier is that one would expect increased device robustness and reduced variations in device characteristic. Indeed, within the limited statistics currently available with the new barrier design, we have observed about an order of magnitude improvement in uniformity of junction properties such as I_c or R_n.

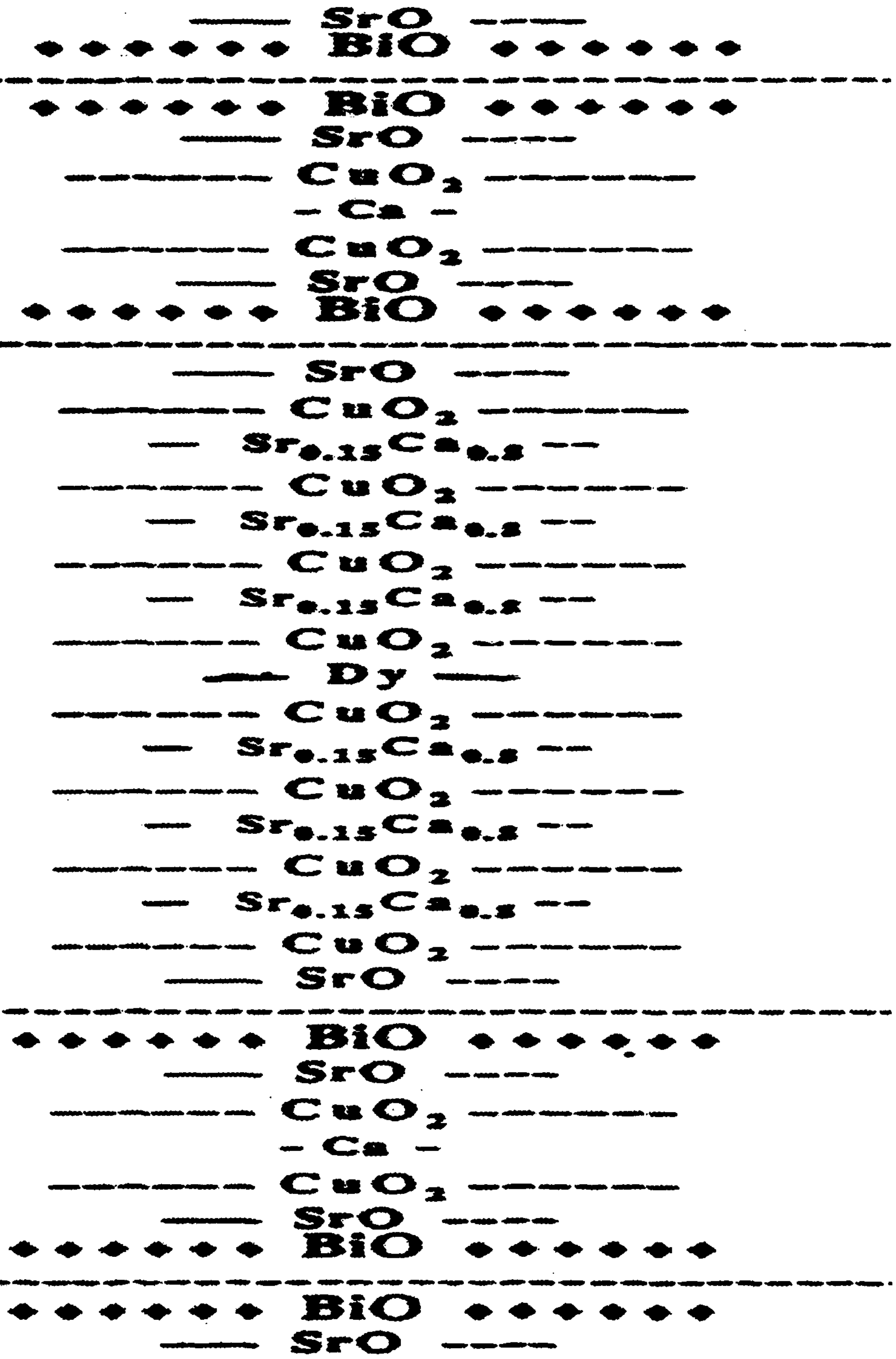


Figure 4. A schematic description of the barrier design that involves a novel metastable 0278 compound.

References:

1. J. N. Eckstein, D. G. Schlom, E. S. Hellman, K. E. von Dessonneck, *J. Vac. Sci. Technol.* **B7**, 319 (1989).

2. J. N. Eckstein, I. Bozovic, K. E. von Dessonneck, D. G. Schlom, J. S. Harris, Jr., S. M. Bauman, *Appl. Phys. Lett.* **57**, 931 (1990).

3. J. N. Eckstein, I. Bozovic, D. G. Schlom, J. S. Harris, Jr., *Appl. Phys. Lett.* **57**, 1049 (1990).

4. I. Bozovic, J. N. Eckstein, D. G. Schlom, and J. S. Harris, Jr., in *"Science and Technology of Thin Film Superconductors II"*, edited by R. McConnell and S. Wolf (Plenum, New York, 1990) p. 267.

5. J. N. Eckstein, I. Bozovic, M. E. Klausmeier-Brown, G. Virshup, and K. S. Ralls, *Materials Research Society Bulletin* **17**, 19 (1992).

6. J. N. Eckstein ,I. Bozovic, M. E. Klausmeier-Brown, G. Virshup, and K. S. Ralls, *Thin Solid Films* **216**, 8 (1992).

7. M. E. Klausmeier-Brown, J. N. Eckstein, I. Bozovic, and G. F. Vir-shup, Appl. Phys. Lett **60**, 657 (1992).

8. G. F. Virshup, M. E. Klausmeier-Brown, J. N. Eckstein, and I. Bo-zovic, Appl. Phys. Lett. **60**, 2288 (1992).

9. M. E. Klausmeier-Brown, G. F. Virshup, I. Bozovic, J. N. Eckstein, and K. S. Ralls, Appl. Phys. Lett. **61**, 2806 (1992).

10. I. Bozovic, J. N. Eckstein, M. E. Klausmeier-Brown, and G. F. Vir-shup, Journal of Superconductivity **5**, 19 (1992).

11. I. Bozovic and J. N. Eckstein, Active and Passive Elec. Comp (1993) in press.

APPENDIX B

HIGH TEMPERATURE SUPERCONDUCTOR RAPID SINGLE FLUX QUANTUM LOGIC

HTS RSFQ LOGIC

1. RSFQ Logic

Recent successful development of Rapid Single Flux Quantum (RSFQ) logic [1] demonstrates advantages of SFQ digital circuits over traditional latching logic. In the RSFQ logic family, information is stored as circulating currents in superconductive loops. A single quantum of magnetic flux, $\Phi_0 = 2.07$ pH·mA, represents a binary "1." Simple RSFQ gates dissipate less than 1 µW or 1% of the power of latching logic. The ultimate speed of RSFQ digital devices can exceed 100 GHz. From a fabrication point of view, a main advantage of RSFQ logic is that they only require nonlatching Josephson junctions with nonhysteretic I-V curve. These types of junction are being fabricated in HTS.

Our present analysis is intended to discuss the possibility of HTS implementation of RSFQ circuits based on the Varian process. As an RSFQ example we consider a typical RSFQ element, a 2:1 prescaler or toggle flip-flop with input Josephson transmission line (JTL) (Figure 1). The circuit design covers all critical parameters required for the design of arbitrary RSFQ devices.

The basic components of RSFQ circuits are Josephson junctions, inductances, and the bias resistors. For an optimized circuit, all parameters and their ratios are typically within ±30%. The range of the various critical currents of the employed Josephson junctions is $I_c$min to ~ 3 $I_c$min. For the RSFQ circuit of Figure 1, the junctions critical current ratios to Icmin are: J1 = 1.0, J2= 1.5, J3 = 2.0, J4 = 2.5, J01 = J02 = 1.5, J03 = J04 = 2.0. The circuit inductances are divided into two classes: storage and nonstorage. A storage inductance (L00) makes it possible to trap a SFQ into the superconducting loop (J03, L00, J04) and store it in the form of a persistent circulating current until it is specifically released. Nonstorage inductances (L1, L2, L3, L01, L02) are for interconnection only and should not trap SFQs under any

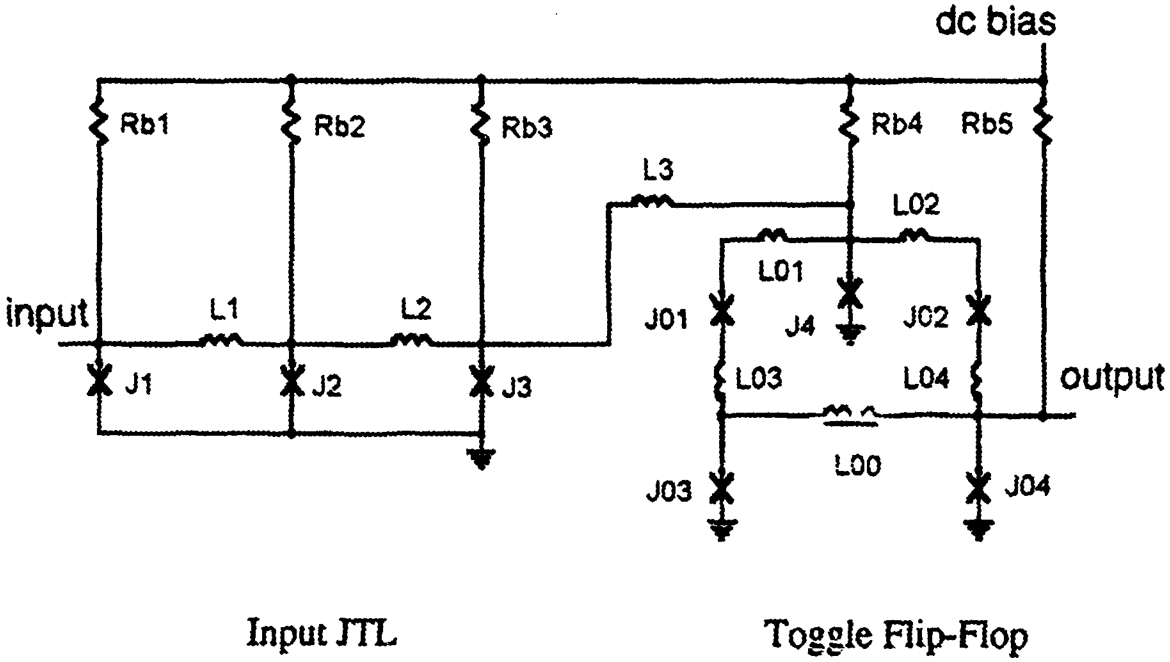


Figure 1: A 2:1 Prescaler - a toggle flip-flop with input JTL .

conditions. In addition, there is always some number of parasitic inductances (L03, L04) associated with the concrete layout and supposed to be nonstorage also. A basic parameter is the dimensionless inductance $\beta_l = 2\pi L I_c/\Phi_0$. For a storage inductance $\beta_l \approx 4$ to 7, and for nonstorage $\beta_l \approx 2$ to 4. For parasitics we should have $\beta_l \leq 1$, otherwise circuit margins will not achieve ±30%. The bias resistors (Rb1, Rb2, Rb3, Rb4, Rb5) provide the dc biases I < Ic for the corresponding junctions and should be in the same ratios as the junction critical currents. The value of resistors are 5 to 10 times higher than the junction resistance R_n. There are no extra specific requirements for resistors beyond the standard LTS implementation.

The next section describes the requirements for Josephson junctions and inductances for the projected HTS implementation.

2. Requirements for Circuit Parameters

2.1 Josephson Junctions.

The Josephson junction are characterized by the critical current I_c, the I_cR_n product and the McCumber-Stewart parameter β_c. Specific requirements for I_c will be discussed later in this section. The I_cR_n product is not very important at the initial stage of implementation. In general, it is desirable to have this parameter be higher in order to attain higher operational speed. For example for the Varian, $I_cR_n = 0.5$ mV, the expected ultimate operational frequency for the circuit of Figure 1 is ~ 160 GHz. This frequency is comparable with the operational frequencies attainable with traditional Nb superconductive circuits. The McCumber-Stewart parameter β_c should be less than 1 to preserve the nonhysteretic I-V curve.

The requirement for critical current is defined by the operational temperature $T < T_c$. This temperature sets a minimal threshold energy U_0 of the Josephson junction biased by the dc bias current $I < I_c$. To ensure low (10^{-30}) probability of false junction switching important for digital applications, the energy U_0 should satisfy the following condition:

$$U_0 > 100\ k_BT; \qquad (1)$$
$$U_0 \equiv 2/3\ I_c\Phi_0/2\pi\ [2(1-I/I_c)]^{3/2},$$

where k_B is Boltzman's constant. It is easy to see that the minimal I_c at given I/I_c is proportional to T. The value of I/I_c is determined by the circuit schematic and for most RSFQ circuits is about 0.8.

$$I_c\text{min} \approx \pi/\Phi_0\ 600\ k_BT \approx T\mu A/K. \qquad (2)$$

For liquid N_2 T = 77 K and $I_c$min = 2 mA, for moderate temperature T = 40 K - $I_c$min = 1 mA.

Another important criterion for choice of the operational temperature is the circuit tolerance to temperature variation. If T is close to T_c a small variation of temperature ΔT would cause a large variation of critical currents $\delta I_c = \Delta I_c/I_c$. At T = 77 ^{0}K and ΔT = 5 ^{0}K, $\delta I_c \approx \pm 15\%$. Such large critical current variations make the device operation at 77 ^{0}K problematic, taking into account the additional existing fabrication spread. Alternatively, at 40 ^{0}K and for the same ΔT = 5 ^{0}K, $\delta I_c \approx \pm 5\%$ that is quite acceptable for digital applications.

The feasibility of fabricating moderately complex circuits depends on the junction uniformity. According to some approximations [2], 15-junction prototype HTS chips can be made with a yield of about 50% if the standard process deviation does not exceed 12%.

2.2 Inductances.

To achieve the required range of β_l from 2 to 7 and keep parasitic inductances low enough, the development of multilayer superconductive structures including a ground plane layer is essential. The inductance L of a superconductive strip over a ground plane (microstrip line - MSL) is:

$$L = \mu_0 \Lambda_{eff} / (K(W, h) W); \tag{3a}$$

$$\Lambda_{eff} = h + \lambda_1 \, cth(d_1/\lambda_1) + \lambda_2 \, cth\,(d_2/\lambda_2), \tag{3b}$$

where $\mu_0 = 4{*}10^{-7}$ H/m, W - is the width of MSL, h - is the dielectric thickness, d_1 - is the layer thickness, d_2 - is the ground plane thickness, λ_1 - is the penetration depth of the layer, λ_2 - is the penetration depth of the ground plane layer, and K(W, h) - is a fringe factor. For our estimate we can approximate $K \approx 1$ at $W/h > 4$, $K \approx 3(h/W)^{1/2}$ at $W/h < 4$.

To lay out the circuit inductances with a reasonable accuracy, the inductance of a lithographic square (MSL with its length equals to its width W), $\beta_{ly} = \beta_l W$, should not exceed 2. From (2), (3) and approximating $\Lambda_{eff} \approx h + 2\,\lambda$

$$\beta_{ly} \approx 3{*}10^4\, T\,(h + 2\,\lambda)/\; K(W, h) \tag{4}$$

For T = 77 ^{0}K, penetration depth is large and h +2 λ is about 2 μm. It makes $\beta_{ly} \approx 4$ at usual optical lithography (K ~ 1). For 40 ^{0}K, β_{ly} becomes closer to desirable value 2. Using more advanced lithography to achieve submicron linewidth, it is possible to relax further the inductance design due to the larger than 1 fringe factor K(W, h).

3. Conclusion

Considering a typical RSFQ circuit, a toggle flip-flop with input JTL (2:1 prescaler), we have analyzed the feasibility of its HTS implementation. To implement HTS RSFQ circuits, a multilayer superconducting structure including a superconductive ground plane will be required. As a result of this analysis, the best choice is RSFQ circuit implementation for operation at 40 ^{0}K. At this temperature, the minimal critical current is to be about 1 mA. This makes the circuit stable the thermal fluctuations and enables suitable accuracy in the layout design of circuit inductances. A higher than available I_cR_n product of 0.5 mV is not important at the initial stage. However, fabrication uniformity should be better than ±15%. If these requirements are met, the RSFQ circuits with a complexity of 15 - 20 junctions (e.g., a 2:1 prescaler) can be built for a maximum input frequency of 160 GHz.

5. References:

1. K. Likharev et al., IEEE Trans. Appl. Superconductivity, **1**, 3 (1991).

2. D. Miller et al., IEEE Trans. Appl. Superconductivity, **3**, 2728 (1993).

APPENDIX C

SURFACE AND INTERFACE CHARACTERIZATIONTHROUGH X-RAY PHOTOELECTRON SPECTROSCOPY

FINAL REPORT

Development of Contacts for High Temperature Superconductors

Proposal Date: July 1, 1992 to June 30, 1993
Contract No.: F33615-90-C-1456
Subcontract No: 703767

Submitted to
Mr. Elie Track
Hypres, Inc.
175 Clearbrook Road
Elmsford, NY 10523

Professor William E. Spicer
Stanford University
Solid State Laboratory
Stanford, CA 94305-4055
Phone: 415-723-4643

The main focus of the Stanford subcontract is to study the contact metallurgy and fabrication related issues of $Bi_2Sr_2CaCu_2O_{8+\delta}$ and related materials. This study is carried out with the high-resolution angle-resolved photoemission capabilities developed and pioneered by the Stanford group. Under this contract, we have performed detailed studies of high quality materials in both thin film and crystalline form. The single crystalline work was done in collaboration with Prof. A. Kapitulnik of Stanford University and this work provides critical data base for our thin film and contact studies. The thin film work was done in close collaboration with the Varian group. In the following paragraphs, we give details of our results.

1) We have performed by far the most detailed studies of the surface superconductivity of the Bi2212 single crystals. [1, 2] The surface superconductivity information is crucial to the understanding and controlling the quality of Josephson junctions made on these materials where coherence length is very short. Our most important discovery was finding an anomalously large superconducting gap anisotropy in the a-b plan [1]. Fig.1 presents normal (open squares) and superconducting state (filled circles) ARPES spectra from a Bi2212 sample cleaved in UHV. The **k**-space locations were chosen so that the normal state peak is at the Fermi level, and the midpoint of the leading edge in the normal state coincides with the Fermi level at both points A and B. Very clear spectral changes are observed at A as the sample is cooled below T_c. The leading edge of the superconducting spectrum is pulled back to higher binding energy, reflecting the opening of the superconducting energy gap. At the same time, a "pile-up" peak and a "dip" near -80 meV appear in the data. [3] At B, only minor changes with temperature are observed, indicating that the gap is undetectable within experimental uncertainty. This striking difference at the two **k**-space locations indicates that the superconducting gap is very anisotropic.

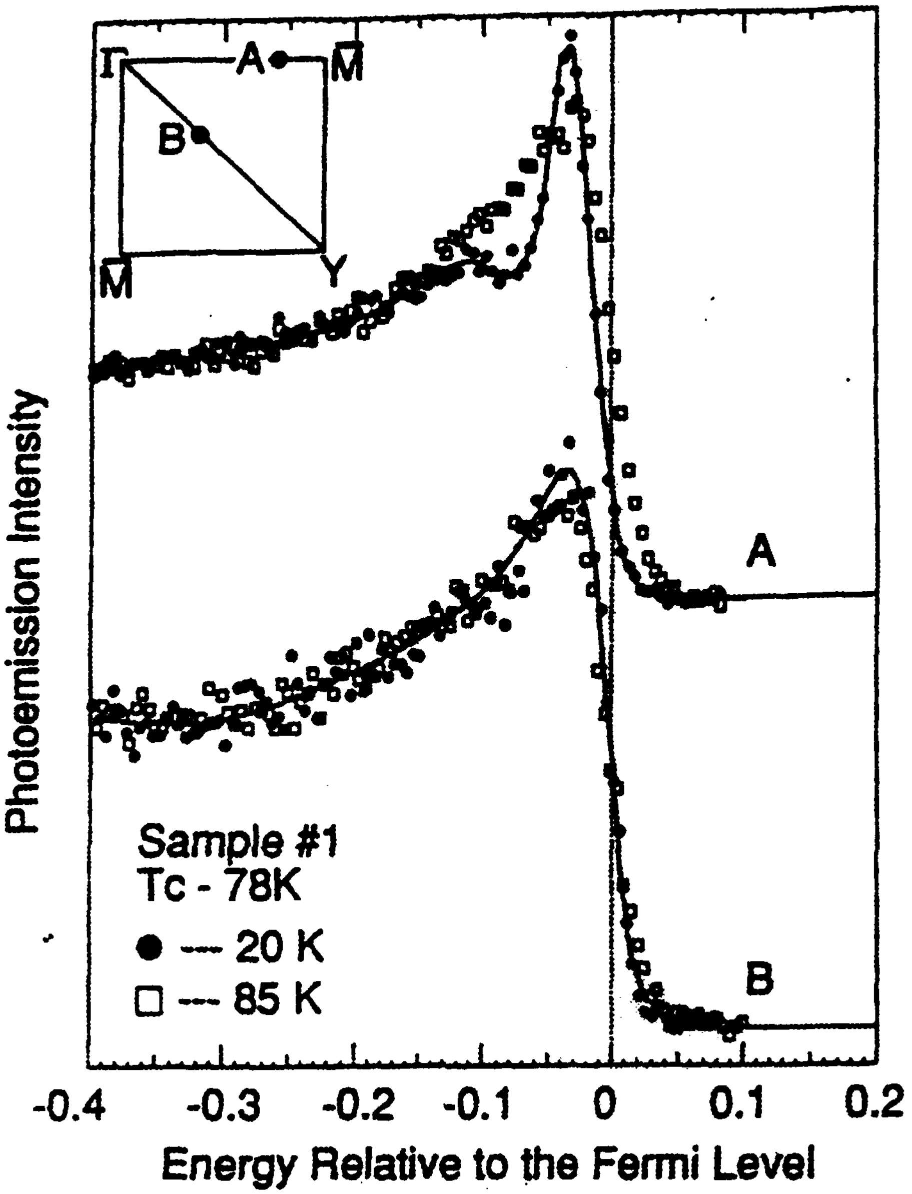


Figure 1: High resolution photoemission spectra from sample No. 1 recorded at k-space locations A and B, as illustrated in the inset. The spectra at B were measured before those at A. The spectral changes above and below T_c are caused by the opening of the superconductor gap. The change at A is quite visible, yielding a larger gap. The change at B is hardly visible, suggesting a very small or null gap.

There are several possible explanations for the observed gap anisotropy. The first is that Bi2212 has a d-wave instead of a more conventional BCS s-wave superconducting order parameter, instead of conventional s-wave BCS superconducting order parameter. This explanation is also consistent with many other experimental findings such as NMR and microwave penetration depth measurements. If this explanation is correct, we are dealing with a very different kind of superconductors than the ones we are familiar with. We will have to reevaluate the junctions and devices made with the cuprate superconductor. This d-wave scenario is also a possible explanation for the fact that no "ideal" tunneling junction has been fabricated using the cuprate superconducting material. The second possible explanation for the observed gap anisotropy is that the superconductivity in this material is caused by interlayer Josephson tunneling mechanism with very anisotropic tunneling matrix elements;[4] however, in this case, the superconducting order parameter has a s-wave symmetry. It is still too early to determine which explanation is the correct one.

2) A significant discovery is that the Bi2212 surface is much more inert than the other high Tc materials we have tested. We carried out his test in the following way. First, we studied the fresh Bi2212 surface and confirmed it to be superconducting with our surface sensitive photoemission spectroscopy. We then introduced contamination by exposing Bi2212 surface at low temperature to residue gases. We found that the surface after this exposure is no longer superconducting. We find that we can regenerate a superconducting surface by warming the sample to room temperature and blowing away the contamination gas on the surface. In contrast to Bi2212, the surface of the other cuprate superconductors cannot survive this stability test. The high surface stability of Bi2212 makes it a promising substrate for contacts because of its higher tolerance to processing conditions.

3) We have studied the growth conditions and chemical reactions when growing metal overlayers on high temperature superconductors. In addition to commonly used materials such as Au and Ag, we have also studied materials such as Bi and

metallic A_xC_{60} ("A" represents an alkali metal - we used Rb and K). The interface study between A_xC_{60} and Bi2212 is motivated by our desire to detect (or to understand the absence) of the proximity effect. This effect is crucial for the performance of SNS junction devices, and its existence between cuprate superconductors and normal metals so far remains an open issue. Although supercurrents have been observed on some SNS junctions, the I_cR_n product of these junctions remains low. Our early attempt to detect the proximity induced gap in Au and Ag has resulted in null answer.[5] A possible explanation for that finding is that the carrier density in Au and Ag is too high so that we were not able to see the proximity induced gap. The A_xC_{60} film has very low carrier density and, thus, might be a good choice for the experiment. Furthermore, A_3C_{60} are also superconducting and so additional insight can be gained through the study of the coupling of the superconductor between the two materials. So far, no proximity effect induced gap has been observed yet. However, we are still in the process of determining whether this null result is intrinsic or if this is complicated by the chemical reactions at the interface.

Figures 2 and 3 give high-resolution photoemission data recorded from interfaces of Bi/Bi2212 and A_xC_{60}/Bi2212. The spectra are presented as a function of interface thickness. The A_xC_{60} spectra show a representative 60Å overlayers compared to a thicker film. With the increase of interface thickness, the electronic structure as reflected in the photoemission spectra is severely modified. This means that, even though Bi2212 material has probably the most inert surface compared with other high temperature superconductors, its contact with Bi and A_xC_{60} is highly reactive. This work indicates that while these materials are better candidates for obtaining a proximity effect for reasons such as carrier density and carrier type, they are too reactive to produce good interface for proximity effects. As mentioned in the last paragraph, we never detected the proximity effect induced superconducting gap in the metallic overlayer. This new finding and our earlier work on Au/Bi2212 and Ag/Bi2212 interfaces suggest that the metallic contact of Bi2212 and other metals is still an unresolved problem.

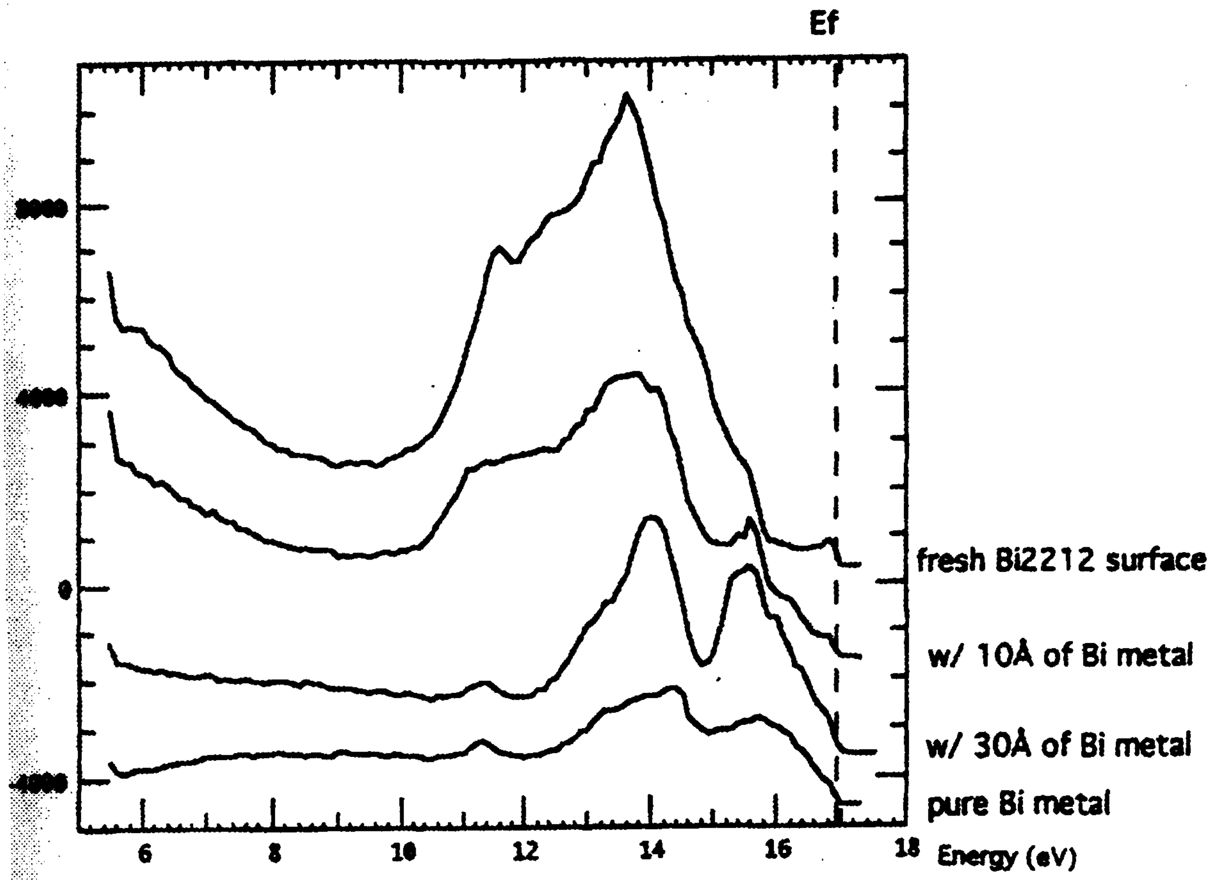


Figure 2: Bi2212 superconductor with various amounts of Bi metal. The 16 mV Fermi level gap present in the top spectrum was quenched by the addion of only 10 Å of bismuth metal. No proximity effect was observed. The Bi2212 core levels were also shifted which demonstrates that the surface reacted with the metal - an undesirable occurrence for this superconductor system.

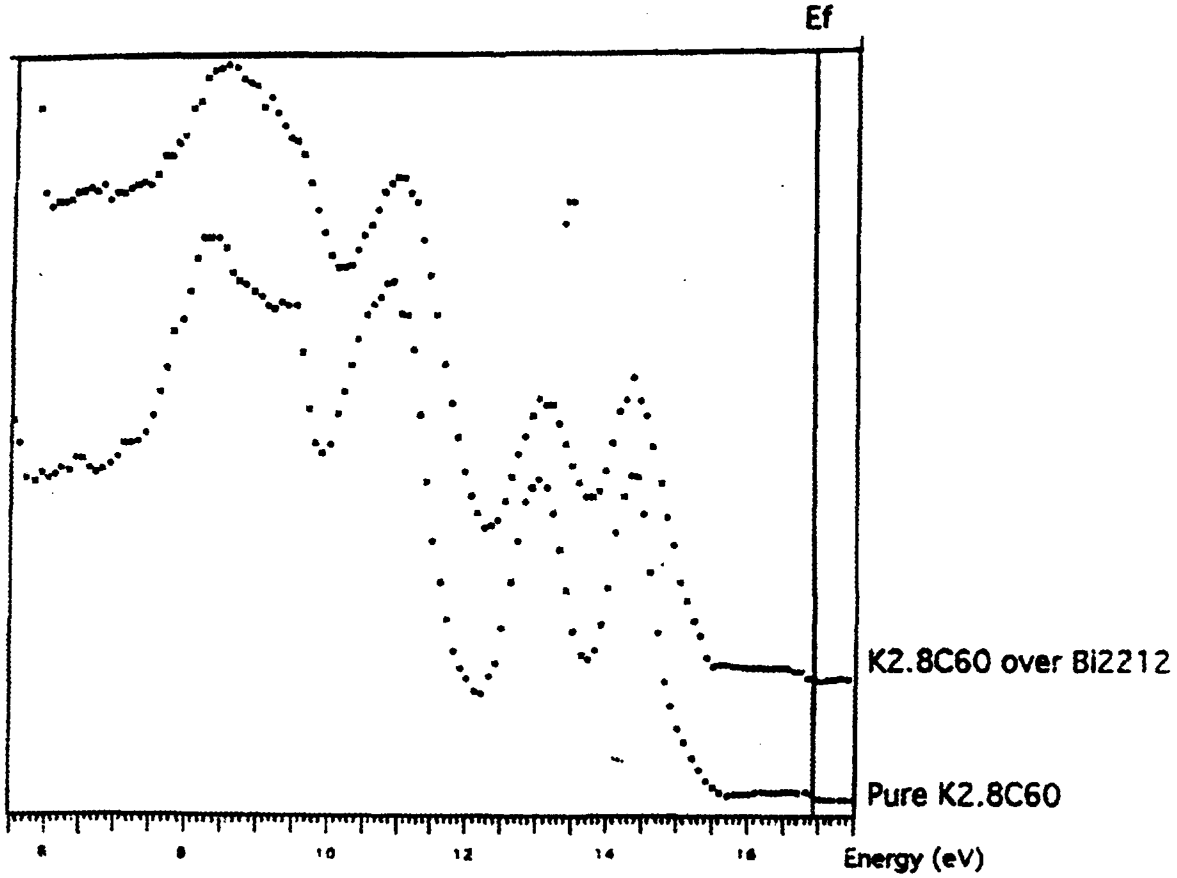


Figure 3: Bi 2212 with an overlayer of 60 Å of $K_{2.8}C_{60}$ compared with a pure $K_{2.8}C_{60}$ sample. The best overlayer results were these, where the C_{60} overlayer charged only 60 mV. All other samples charged more severely. It appears that the potassium migrates to the Bi2212 surface and then reacts with it. Previous research showed that the high reactivity of potassium on Bi2212 disrupts its superconductivity and conductivity.

4) We have performed high-resolution angle-resolved photoemission studies of the MBE grown thin films from Varian. We have successfully developed a technique to prepare a clean film surface for photoemission studies. Figure 4 shows data taken from bulk crystals and thin films below T_c. The opening of the superconducting gap below T_c is clearly visible in the data. These data represent the first observation of the superconducting gap in thin films by surface sensitive technique such as photoemission. This is a significant technical breakthrough because it means that the powerful surface sensitive characterization techniques such as photoemission are now applicable to the problems in thin films and possibly the electronic devices associated with them. Furthermore, the data taken at different parts of the **k**-space are different. This implies that the Varian MBE films are indeed single crystalline films. We have also studied the films with different oxygen treatment and found that their surface superconducting properties are very sensitive functions of the oxygen treatment. Now we have optimize our techniques, we should be able to correlate the surface and interface properties of these films with their transport properties and the related characteristics of devices made out of these films.

The projects of our subcontract is moving forward steadily and has been productive. An important reason for our performance is that we have developed a unique and powerful experimental capabilities. This project helped us to strengthen these capabilities, which will make our future effort more productive and efficient. We anticipate making other significant discoveries from a continuation of this subcontract.

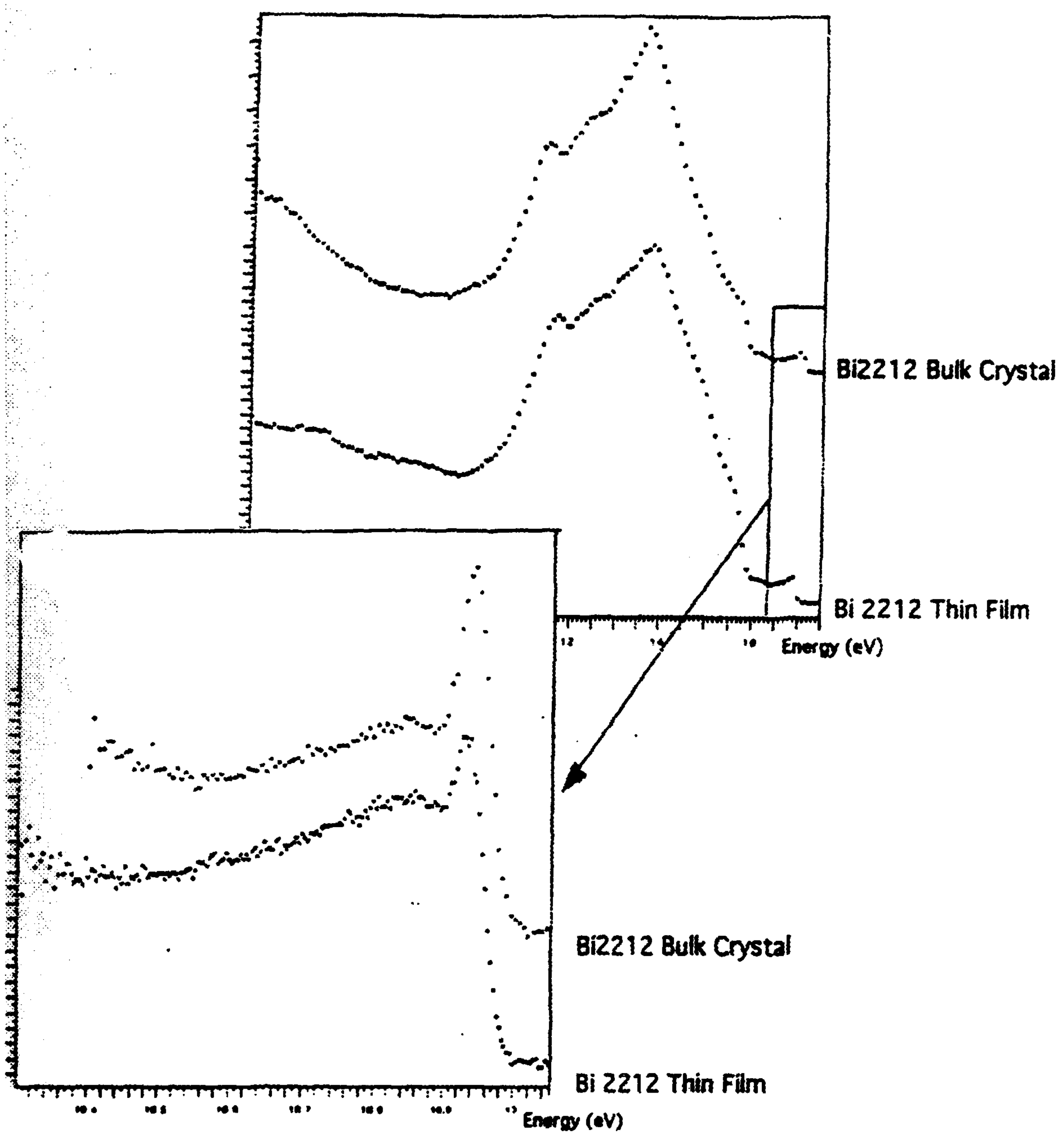


Figure 4: Thin film samples of Bi2212 compare well with bulk crystals. The sensitive, near Fermi area also compars well giving a superconducting gap of 11 mV compared to a typical bulk value of 16 mV.

Reference:

1. B.O. Wells et al., Phys. Rev. B 46, 11830 (1992)
2. Z.-X. Shen et al., Phys. Rev. Lett., 70, 1553 (1993)
3. D.S. Dessau et al., Phys. Rev. Lett., 66, 2160 (1991)
4. S. Chakravarty et al., Science, (July) 1993.
5. D.S. Dessau et al., Appl. Phy. Lett. 58, 1332 (1991).