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SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

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| 1a. RESTRICTIVE MARKINGS<br><b>UNCLASSIFIED</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |  | 1b. RESTRICTIVE MARKINGS                                                                  |  |
| 2a. SECURITY CLASSIFICATION AUTHORITY                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |  | 3. DISTRIBUTION/AVAILABILITY OF REPORT<br><br>UNLIMITED                                   |  |
| 2b. DE                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |  | 5. MONITORING ORGANIZATION REPORT NUMBER(S)<br><br>AFOSR-TR- 89-0098                      |  |
| 4. PER<br><br>AD-A204 556                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |  | 6a. NAME OF PERFORMING ORGANIZATION<br>Stanford University<br>W. W. Hansen Lab of Physics |  |
| 6b. OFFICE SYMBOL<br>(If applicable)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |  | 7a. NAME OF MONITORING ORGANIZATION<br>AFOSR/NE                                           |  |
| 6c. ADDRESS (City, State and ZIP Code)<br>Stanford CA 94305                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |  | 7b. ADDRESS (City, State and ZIP Code)<br>Bldg 410<br>Bolling AFB DC 20332-6448           |  |
| 8a. NAME OF FUNDING/SPONSORING ORGANIZATION<br>Same as 7a                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |  | 8b. OFFICE SYMBOL<br>(If applicable)                                                      |  |
| 9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER<br>F49620-88-C-0004                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |  | 10. SOURCE OF FUNDING NOS.                                                                |  |
| 8c. ADDRESS (City, State and ZIP Code)<br>Same as 7b                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |  | PROGRAM ELEMENT NO.<br>61102F                                                             |  |
| 11. TITLE (Include Security Classification)<br>Superconducting Thin Films Composites and Junctions                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |  | PROJECT NO.<br>2306                                                                       |  |
| 12. PERSONAL AUTHOR(S)<br>Professor Geballe                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |  | TASK NO.<br>C1                                                                            |  |
| 13a. TYPE OF REPORT<br>Annual Report                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |  | 13b. TIME COVERED<br>FROM 01/10/87 TO 31/10/88                                            |  |
| 14. DATE OF REPORT (Yr., Mo., Day)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |  | 15. PAGE COUNT                                                                            |  |
| 16. SUPPLEMENTARY NOTATION                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             |  |                                                                                           |  |
| 17. COSATI CODES                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |  |                                                                                           |  |
| 18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |  |                                                                                           |  |
| 19. ABSTRACT (Continue on reverse if necessary and identify by block number)<br><br>Four methods for growing thin films of high Tc superconductors have been developed and properties of the films have been investigated. In addition to the support received under this contract, the research was also supported in part by the Stanford Center for Materials Research under the NSF/MRL Program and other DOD contracts. A new superconducting structure of composition $Y_{1.8}Ba_{0.2}Cu_3O_{7-x}$ has been discovered. In a number of ways it has properties which are better than those of the $YBa_2Cu_3O_7$ structures. It is less anisotropic in the $CuO_2$ (conducting planes) and therefore has less (or no) twinning. It retains oxygen under vacuum conditions and thus may be superior for making planar Josephson Junctions. Keywords: Yttrium Compounds, Barium, Copper Compounds, Oxides, Epitaxial growth, Vapor deposition. (CW) |  |                                                                                           |  |
| 20. DISTRIBUTION/AVAILABILITY OF ABSTRACT<br>UNCLASSIFIED/UNLIMITED <input type="checkbox"/> SAME AS RPT. <input type="checkbox"/> DTIC USERS <input type="checkbox"/>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |  |                                                                                           |  |
| 21. ABSTRACT<br><b>UNCLASSIFIED</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |  |                                                                                           |  |
| 22a. NAME OF RESPONSIBLE INDIVIDUAL<br>Weinstock                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |  |                                                                                           |  |
| 22b. TELEPHONE NUMBER<br>(Include Area Code)<br>(202) 767-4933                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         |  |                                                                                           |  |
| 22c. OFFICE SYMBOL<br>NE                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |  |                                                                                           |  |

ANNUAL REPORT  
F49620-88-C-0004  
10/1/87-10/31/88

### **ABSTRACT**

Four methods for growing thin films of high-Tc superconductors have been developed and properties of the films have been investigated. In addition to the support received under this contract, the research was also supported in part by the Stanford Center for Materials Research under the NSF/MRL Program and other DOD contracts.

- Superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  films of thickness ranging from 500Å to 5µm have been successfully made by the reactive magnetron sputtering technique. Effects of composition, annealing condition and thickness of an epitaxial film on its orientation are discussed. Films show anisotropic resistivities and critical current densities that are orientation dependent.

- Conditions required for in situ growth of  $\text{YBaCuO}$  thin films by reactive electron-beam evaporation have been explored. Three sources of activated oxygen (atomic oxygen from microwave discharge, a plasma generated by electron beams, and an ion beam) were compared. The best results so far have been obtained with atomic oxygen. Epitaxial films with high critical currents have been grown on  $\text{SrTiO}_3$  <100> and <110>,  $\text{Al}_2\text{O}_3$  <1102> and  $\text{MgO}$  <100> at 600°C. Evaporation rates were controlled with a rate monitor using atomic absorption.

- Thin films of  $\text{Bi-Sr-Ca-Cu-O}$  have been produced on substrates including (100)  $\text{MgO}$  and (100)  $\text{SrTiO}_3$  using pulsed laser ablation of a variety of targets. The films have been characterized using surface roughness, resistivity, x-ray diffraction, scanning electron microscopy and transmission electron microscopy as criteria.

- Superconducting thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  have been produced by metallorganic chemical vapor deposition (MOCVD). Volatile metallorganic precursors of the oxide components,  $\beta$ -diketonates of Y, Ba, and Cu, are thermally decomposed on hot substrates to form crystalline films. The Ba compound was evaporated in the presence of externally added vapors of the  $\beta$ -diketonate to obtain steady evaporative behavior. Superconducting films were obtained on  $\text{SrTiO}_3$  substrates at temperatures above 800°C. The best films have onset temperatures of 90K and loss of resistance as high as 68K. Epitaxial growth was obtained.

A new superconducting structure of composition  $\text{Y}_2\text{Ba}_4\text{Cu}_8\text{O}_{20}$  has been discovered. In a number of ways it has properties which are better than those of the  $\text{YBa}_2\text{Cu}_3\text{O}_7$  structure. It is less anisotropic in the  $\text{CuO}_2$  (conducting planes) and therefore has less (or no) twinning. It retains oxygen under vacuum conditions and thus may be superior for making planar Josephson Junctions.

The upper critical field  $H_{c2}$  and fluctuation conductivity have been measured for highly oriented thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ . The  $H_{c2}$  results demonstrate the intrinsic anisotropy in this layered superconductor. The broadening of the resistive transition under fields is interpreted in terms of critical fluctuations. The fluctuation conductivity shows

dimensional crossover as expected for quasi-two-dimensional material. Using these data, the intralayer and interlayer coherence lengths  $\xi_c(0)$  and  $\xi_{ab}(0)$  have been determined.

Optical transmission and reflection spectra (mid ir through uv) and Raman spectra of superior-quality 90-, 180-, 400-, and 1000-nm-thick superconducting Y-Ba-Cu-O films have been measured. Characteristic excitonic bands, and in particular the absorption band at  $\approx 0.37$  eV were not observed, making it unlikely that the high- $T_c$  superconductivity in cuprates arises from exciton-mediated electron pairing.

A scanning tunneling microscope has been used to image the a-b plane of a single crystal of  $\text{Bi}_2(\text{Ca},\text{Sr})_3\text{Cu}_2\text{O}_{8+\delta}$  ( $T_c \sim 85\text{K}$ ) and a thin film grown epitaxially on a (100)  $\text{SrTiO}_3$  substrate. Images taken at room temperature in air of a freshly cleaved bulk sample clearly show the one-dimensional superstructure ( $27.2 \pm 0.7 \text{\AA}$  periodicity) which is common to this phase.

A new film deposition system (with funding from the DOD-sponsored University Research Instrumentation Program) has been assembled at the vendor's site and has been undergoing tests prior to acceptance. The growth and the load lock/processing chamber have been tested and are leak-tight. The substrate transfer arm and the associated support and transport assemblies have been assembled and tested. A critical test of the concept of using sliding teflon seals has successfully passed its initial vacuum test. The design of the six emitter, ten pocket electron-beam source has been completed. The magnetic field configuration is a new concept and should permit the close proximity of the sources without serious interaction between the sources which is highly desirable.

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## TALKS & SEMINARS

- MRL Directors Meeting, Washington, DC, November 3, 1987
- MRS, Boston, November 29-30, 1987
- 1988 Condensed Matter Physics Conf., Aspen, CO, January 17-23, 1988
- University of Utah, February 22
- M<sup>2</sup>S, Interlaken, Switzerland, February 26-March 5
- APS, New Orleans, March 20-25
- MRS, Reno, Nevada, April 5-9
- Los Alamos, April 11-12
- Case University, Cleveland, April 21
- Cornell University, May 10-12
- Chevron, Richmond, CA, May 26
- Crystal Growth, Sierra Camp, June 7-9
- ASC Conference, San Francisco, August 21-25
- Tokai University Workshop, Tokyo, Japan, November 12-17

# DEPT. OF APPLIED PHYSICS - LIST OF VISITORS

December 1987

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Soviet Delegation for Condensed Matter Theory Workshop  
A. F. Andreev, S. A. Brazovski, I. E. Dzyaloshinskii, L. P. Gor'kov, I. M. Khalatnikov, Yu. A. Ossipyan, V. L. Pokrovskii, M. V. Sadovskii, I. F. Schegolev, V. B. Timofeev

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K. Togano, National Research Institute for Metals, Ibaraki, Japan

January 1988

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Kiyotaka Wasa, Matsushita Electric Industrial Co, Ltd.

February

-17/19

Alex Mueller, IBM Zurich Research Lab

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A. Ozeki, Science News Section, Asahi Shimbun, Tokyo

March

-30/31

Dr. Simon Capelin, Cambridge University Press

April

(13)

Dr. Tsuchiya, Mitsubishi Chemical

(27)

Michael Gurvitch, AT&T

?

Masasi Inoue, Dept. of Materials Science, Hiroshima University

May

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King-Ning Tu, IBM Watson Research Center

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Peter Dickin

March

-14/16

Masatoshi Sato, Institute for Molecular Science

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Rudy Peierls

Jim Harper, IBM

April

-27/28

Harold Weinstock, AFOSR

June

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Edward Fenton, nat. Res. Council, Ottawa, Canada

July

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Dr. Schultz, Siemens Central Research Lab

August

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STAG

September

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H. Adrian, Phys. Institut, Erlangen, FRG

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Dwight Duston, SDI, Washington, DC

October

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Rick Scott, et al. TRW vitists Stanford

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Bill Nellis

November

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J. Geerk, Inst. f. Nuk. Festkorperphysik

**Publications Funded by Air Force**  
**Contract F49620-88-C-0004**  
**October 1987-88**

1. "Optical Measurements on Oriented Thin  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  Films: Evidence Against Excitonic Superconductivity?" by I. Bozovic, D. Kirolov, A. Kapitulnik, K. Char, M. R. Beasley, T. H. Geballe, Y. H. Kim, and A. J. Heeger, *Phys. Rev. Lett.* 59, 19, 11-9-87 2219-21.
2. "High  $T_c$  Thin Film Superconductivity: Science and Technology (invited)," by T. H. Geballe, *Journal of App. Physics* 63 (8), 4-15-88, 4003-4.
3. "Photoemission Study of the Oxidation States and the Electronic Structure of Copper Oxide Superconducting Thin Film," by Z. -X. Shen, J.-J. Yeh, I. Lindau, W. E. Spicer, J.Z. Sun, K. Char, N. Missert, A. Kapitulnik, T.H. Geballe, M.R. Beasley, *SPIE Symp. Proc. Vol. 948-10*, 1987.
4. "Superconductivity - the State that Came in from the Cold," by T. H. Geballe and J. K. Hulm, *Science* 239 367-375 1-22-88.
5. "High-Temperature Superconducting Perovskites - Science and Technology," by T. H. Geballe, talk given at Fermilab; published in *Fermilab Industrial Affiliates Seventh Annual Meeting*, May 21 and 22, 1987.
6. "Ordered Defect Structure in Epitaxial  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  Thin Films," by A. F. Marshall, R. W. Barton, K. Char, A. Kapitulnik, B. Oh and R. H. Hammond, *Phys. Rev. B* 37 16 9353-9358, 6-1-88.
7. "Synthesis and Properties of Thin Film High  $T_c$  Oxide Superconductors, by M. R. Beasley presented at Yamada Conference XVIII on Superconductivity in Highly Correlated Fermion Systems (invited paper), Sendai, Japan, 8-31 to 9-3-87, *Physica* 148B North Holland 1987, pp. 191-195.
8. "Upper Critical Field, Fluctuation Conductivity, and Dimensionality of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$ ," by B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, *Phys. Rev. B* 37, 13 7861-7864, 5-1-88.
9. "Properties of Y-Ba-Cu-O Thin Films with Ordered Defect Structure:  $\text{Y}_2\text{Ba}_4\text{Cu}_8\text{O}_{20-x}$ ," by K. Char, Mark Lee, R. W. Barton, A. F. Marshall, I. Bozovic, R. H. Hammond, M. R. Beasley, T. H. Geballe and A. Kapitulnik; *Phys. Rev. B Rapid Communications* 38 1, 834-837, 7-1-88.
10. "Scanning Tunneling Microscopy of the A-B planes of  $\text{Bi}_2(\text{Ca},\text{Sr})_3\text{Cu}_2\text{O}_{8+\delta}$  Single Crystal and Thin Film" by M. D. Kirk, C. B. Eom, B. Oh, S. Spielman, M. R. Beasley, A. Kapitulnik, T. H. Geballe and C. F. Quate, *Appl. Phys. Lett.* 52 (24) 6-13-88, 2071-2073.
11. "Site Selective Doping and Superconductivity in  $(\text{La}_{1-y}\text{Pr}_y)(\text{Ba}_{2-x}\text{La}_x)\text{Cu}_3)_{7-\delta}$ ," by D. B. Mitzi, P. T. Feffer, J. M. Newsam, D. J. Webb, P. Klavins, A. J. Jacobson and A. Kapitulnik, *Phys. Rev. B* 38, 6667-76 (1988).

12. "Optical Anisotropy of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ " by I. Bozovic, K. Char, S. J. B. Yoo, A. Kapitulnik, M. R. Beasley and T. H. Geballe, Phys. Rev. B Rapid Communications 38 7 (9-1-88) pg. 5077-5080.
13. "Properties of Films of High-Tc Perovskite Superconductors" by A. Kapitulnik. Proceedings of the International Conference in Interlaken, Switzerland, Physica B-C special issue.
14. "Fabrication and Properties of Bi-Sr-Ca-Cu-O Thin Films Made Using Pulsed Laser Deposition," by D. K. Fork, T. H. Geballe, J. B. Boyce, F. Ponce and R. I. Johnson. Presented at the Applied Superconductivity Conference, San Francisco, California August 21-25, 1988, to be published in Proc. IEEE.
15. "Growth and Properties of Sputtered High-Tc Oxide Thin Films," by K. Char, M. R. Hahn, T. Hylton, I. Bozovic, M. R. Beasley, T. H. Geballe and A. Kapitulnik. Presented at the Applied Superconductivity Conference, San Francisco, California August 21-25, 1988, to be published in Proc. IEEE.
16. "In Situ Growth of Superconducting  $\text{YBaCuO}$  Using Reactive Electron Beam Coevaporation," by N. Missert, P. Rosenthal, R. Barton, M. R. Beasley, T. H. Geballe, R. H. Hammond, A. Kapitulnik, J. Mooij, V. Matijasevic, E. Garwin and C. Lu. Presented at the Applied Superconductivity Conference, San Francisco, California August 21-25, 1988, to be published in Proc. IEEE.
17. "Photoemission Study of the Surface Chemistry and the Electronic Structure of Copper Oxide Superconducting Thin Films," by Z.-X. Shen, J.-J. Yeh, I. Lindau, W. E. Spicer, J. Z. Sun, K. Char, N. Missert, A. Kapitulnik, T. H. Geballe and M. R. Beasley, SPIE Vol. 948 - High-Tc Superconductivity: Thin Films and Devices (1988); pp. 41-48.