

AD-A268 016



DOCUMENTATION PAGE

Form Approved
OMB No 0704-0188

This report is estimated to average 1 hour per response, including the time for review of instructions, searching existing data sources, gathering and reviewing the collection of information, Send comments regarding this burden estimate or any other aspect of this collection of information, including this burden estimate, to Washington Headquarters Service, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

2. REPORT DATE
14 July 1993

3. REPORT TYPE AND DATES COVERED
Annual 15 June 1993 - 14 June 1993

4. TITLE AND SUBTITLE
Investigations of YBaCuO Materials with Near-Room-Temperature Resistivity Transitions

5. FUNDING NUMBERS
AFOSR-91-0319

6. AUTHOR(S)
Lowell E. Wenger
J.T. Chen

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Wayne State University
Department of Physics
666 W. Hancock
Detroit, MI 48202

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U.S. Air Force
AFOSR/NE
Building 410
Bolling AFB, DC 20332-6448

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
2305/GS

DTIC
ELECTE
AUG 12 1993
S A D

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION / AVAILABILITY STATEMENT
UNCLASSIFIED/UNLIMITED

93-18875

13. ABSTRACT (Maximum 200 words)
This annual technical report describes the continual superconductivity research on the materials and structural phases responsible for the above 200-K superconducting transitions, their synthesis conditions, and the study of the physical properties associated with these transitions. Superconducting resistive transitions and magnetic flux trapping phenomena have been observed in mixed-phase ceramic samples of YBaCuO (e.g. Y₅Ba₆Cu₁₁O_x) and on the surfaces of YBa₂Cu₃O₇ single crystals in the temperature range 230 K to 270 K. Near room-temperature oxidation studies show that the properties can change, including a large resistance decrease over a 3-month period. This behavior is consistent with the formation of a different oxide phase on the surface of the grains. Structural analyses by x-ray diffraction and high-resolution transmission electron microscopy indicate these samples have surfaces with large amounts of defect structures as well. Correspondingly these surface phases/structures may be responsible for the above 200-K transitions.

This document has been approved for public release and sale; its distribution is unlimited.

14. SUBJECT TERMS
Superconductivity
Room-temperature superconductivity
YBaCuO superconductors

15. NUMBER OF PAGES
5

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

Contract Number: AFOSR-91-0319

Project Title: Investigations of YBaCuO Materials with Near-Room-Temperature Resistivity Transitions

PIs: Lowell E. Wenger
J.T. Chen
Wayne State University

Technical Report for 1992 - 1993:

The main objectives of our superconductivity research are two-fold: (1) to understand the nature of the anomalous transitions with transition temperatures T_c above 200 K observed in some mixed-phase cuprate samples and (2) to improve the sample fabrication conditions so that samples showing the anomalies can be synthesized more consistently. To attain the first objective, measurements of the electrical resistivity, the current-voltage characteristics, the microwave effects, and the magnetization have been continually investigated on samples exhibiting anomalies above 200 K. To improve upon the fabrication conditions, the effects of the reaction temperature, cooling rate, and oxidation conditions have been studied. A brief summary of the progress made to date is briefly described below.

Electrical Measurements

As we have reported previously, the resistive transitions and current-voltage characteristics on several ceramic as well as single-crystal platelet samples of Y-Ba-Cu-O materials have exhibited superconducting-like transitions in the temperature range of 230 K to 270 K.[1-3] Other researchers[4] have made similar observations and provide some confirmation of our results.

Magnetic Measurements

Temperature-dependent magnetization measurements on several ceramic and single-crystal platelet samples of Y-Ba-Cu-O have shown hysteresis between the zero-field-cooled (ZFC) and the magnetic field-cooled (FC) curves with the onset of the hysteresis occurring above 200 K. This hysteretic behavior can be interpreted as superconducting flux trapping inside an inhomogeneous superconductor. ac susceptibility measurements on other ceramic samples have shown small diamagnetic signals equivalent to 1% or less of the diamagnetic signal resulting from the 90-K superconducting transition. C.Y. Huang has independently performed ac susceptibility measurements on some our samples and has observed a diamagnetic shift below 230 K.[5] More recently, we have observed transitions at 336 K on three different

DTIC QUALITY INSPECTED 3

Dist	Special
A-1	

batches of Y-Ba-Cu-O samples. While the ZFC magnetization is featureless in the vicinity of 336 K, the FC magnetization shows a sudden increase below 336 K. Although this transition is detectable by the positive increase in the FC magnetization, this increase could be associated with a small Josephson screening current and the trapping of a large amount of magnetic flux. This type of "positive magnetization" in the FC magnetization data has been previously observed for BiSrCaCuO samples[6-8] as well as for a superconducting Nb disk.[9] Further characterizations are presently being pursued in order to elucidate the exact nature of this magnetic transition.

Microwave Measurements

In addition to the ac susceptibility measurements, Dr. Huang has also performed microwave experiments on the same sample which exhibited the diamagnetic shift at 230 K. A cavity resonance frequency shift has been observed at the same temperature of 230 K which is indicative of an inductance change associated with a superconducting transition.[5]

Material Synthesis and Characterization

The most successful route for synthesizing samples with T_c above 200 K has been by mixing an appropriate ratio of Y_2O_3 , $BaCO_3$, and CuO for a nominal sample composition of $Y_5Ba_6Cu_{11}O_x$ and multiple heatings to about $950^\circ C$ with thorough grindings of the powder between heatings. The powder is subsequently cold-pressed into a pellet and annealed at $950^\circ C$ followed by cutting the pellet into small pieces which are reannealed at $950^\circ C$ in O_2 gas and rapidly cooled back to room temperature. X-ray diffraction patterns show that these pieces are multi-phase with $YBa_2Cu_3O_{7.8}$ and Y_2BaCuO_5 being the predominate structures. High-resolution transmission electron microscopy studies revealed numerous defect structures in the CuO_2 planes. Energy dispersive x-ray spectroscopy indicate a deficiency of Cu near the surface, yielding roughly $YBa_2Cu_{2.7\pm 0.2}O_x$. It should be noted that the Zürich group[10] has reported observing diamagnetism at 120 K in $YBa_2Cu_{2.78}O_7$ single crystal samples. This suggests that defects in Y-Ba-Cu-O materials may play an important role in enhancing T_c .

Oxidation Studies

One critical condition for observing and reproducing a zero-resistance transition having a T_c above 200 K is to keep the sample in an O_2 environment continuously during the measurements and thermal cyclings. This effect has been confirmed by a group at the University of Regensburg.[4] However in other experiments, such as microwave and magnetization measurements, the anomalous high- T_c phenomena can be observed even when the samples are not kept in oxygen environment. One possible explanation for these different experimental observation conditions is that the diffusion of oxygen molecules into a solid at

low temperatures is limited. Thus this results in only the material near the surface of grains being affected by diffusion of oxygen and the formation of granular shells of the higher T_c material. In microwave and magnetization measurements, disconnected granular shells can still contribute to an observable signal even though flux trapping will dominate the magnetization response. However, the observations of zero-resistance and bulk-like Meissner effect require a continuous intergranular path of these "good" surfaces, i.e., good connectivity between granular shells of the higher- T_c material. In order to determine the effect of low-temperature oxygen diffusion (equivalent to near room-temperature annealing) on the Y-Ba-Cu-O materials, the resistivity of one sample has been monitored over a three-month period which has been continuously kept in O_2 gas at 2 atm and at 350 K or below. Not only did the resistance decrease by an order of magnitude at room temperature during this time, but the temperature dependence changed from a linear to a concave upwards behavior, a clear indication of a material change caused by low-temperature oxidation. Furthermore the decrease in the resistance over time can be understood in terms of the growth of the shell thickness covering the grain. Using a simple model calculation involving three geometric parameters (the radius of the grain, the shell thickness, and the intergranular contact depth) and an exponential diffusion of oxygen with time (i.e., the growth of the shell having zero resistance), the calculated results agree qualitatively with the experimental data. This further supports our speculation that a different oxide phase on the surfaces of grains can be formed near room temperature and may be the phase responsible for the anomalously high- T_c superconductivity.

Future Objectives

During the next year, our plans are to continue the near-room-temperature oxidation studies and to expand the studies to include higher O_2 pressures, electrochemical oxidation, and ozone treatments in order to determine the effect upon the material properties above 200 K and ultimately to find better synthesis conditions for reproducing samples which exhibit higher superconducting transition temperatures. In addition to our electrical and magnetic measurements, we will continue to utilize DTA/TG measurements, x-ray diffraction studies, and optical and electron microscopy studies to assist in the material characterizations.

REFERENCES

- [1] J.T. Chen, L-X. Qian, L-Q. Wang, L.E. Wenger, and E.M. Logothetis, *Modn. Phys. Lett.* **3**, 1197 (1989).
- [2] L-Q. Wang, G-H. Chen, L.E. Wenger, and J.T. Chen, *Physica C* **185-189**, 607 (1991).
- [3] J.T. Chen, L.E. Wenger, L-Q. Wang, and G-H. Chen, *AIP Conference Proceedings* (Superconductivity and Its Applications), edited by Y.H. Kao, A.E. Kaloyeros, & H.S. Kwok (AIP, New York, 1991), p. 459.
- [4] R. Schönberger, H.H. Otto, B. Brunner, and K.F. Renk, *Physica C* **173**, 159 (1991).
- [5] C.Y. Huang, private communication.

- [6] P. Svedlingh, K. Niskanen, P. Norling, P. Nordblad, L. Lundgren, B. Lönnberg, and T. Lungström, *Physica C* 162-164, 1365 (1989).
- [7] W. Braunisch, N. Knauf, V. Kataev, S. Neuhausen, A. Grütz, A. Kock, B. Roden, D. Khomskii, and D. Wohlleben, *Phys. Rev. Lett.* 68, 1908 (1992).
- [8] B. Schliepe, M. Stindtman, I. Nikolic, and K. Baberschke, *Phys. Rev. B* 47, 8331 (1993).
- [9] M.S.M. Minhaj, L.E. Wenger, and J.T. Chen, unpublished work; *Bull. Amer. Phys. Soc.* 37, 749 (1992).
- [10] E. Kaldis, J. Karpinski, S. Rusiecki, B. Bucher, K. Conder, and E.Jilek, *Physica C* 185-189, 190 (1991).

Papers and Abstracts:

L-Q. Wang, M.S.M. Minhaj, J.T. Chen, and L.E. Wenger, "Construction of a simple low-field solenoid for the Quantum Design[®] SQUID magnetometer", Rev. Sci. Instrum., accepted.

M.S.M. Minhaj, S. Meepagala, J.T. Chen, and L.E. Wenger, "Thickness dependence on the superconducting properties of thin Nb films", Phys. Rev. B, submitted.

Talks:

J.T. Chen, L.E. Wenger, L-Q. Wang, C.H. Chen, and D.J. Werder, "Measurements, Processings, and Structures of Y-Ba-Cu-O Materials with Anomalous Transitions near 260 K", invited talk, Symposium on Processing and Applications of High T_c Superconductors, Ben Franklin Superconductivity Center, Philadelphia, PA, 29 June 1992.

L.E. Wenger, "Search for Room-Temperature Superconductivity", seminar, Miami University, OH, 7 October 1992.

L.E. Wenger and J.T. Chen, "Experimental Evidence for Superconductivity above 200 K", Near-Room-Temperature Workshop, Bodega Bay, CA, 19 October 1992.

Personnel:

PIs:

Lowell E. Wenger, Professor of Physics
J.T. Chen, Professor of Physics

Graduate Research Assistants: (supported during summer term)

Dah-Chin Ling
Mohamedo Minhaj
Jonathan Obien
David Thompson