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During the period of this grant, October 15, 1997—April 14, 2001, characterization has been carried out in order to understand microwave properties of grain boundaries. Measure- ments in an externally applied dc magnetic field in addition to the rf field of the stripline resonator. Intermodulation distortion measurements have also been carried out as a function of the grain boundary angle. The measurements in external magnetic fields require the use of a suspended microstrip geometry without superconductivity ground planes so that the shielding effects of the ground planes could be avoided. The film parameters which give the best device characterization have been established.					
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Studies of High-T_c Superconducting Films for High-Power Microwave Applications

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OBJECTIVES

- Measurement and modeling of the nonlinear microwave properties of thin films of high-T_C materials in order to understand the loss mechanisms and the nonlinear mechanisms.
- Measurement of the nonlinear properties of grain boundaries as a function of angle in order to relate to films with defects.
- Measurement of the linear and nonlinear microwave impedance of the grain boundaries in an applied dc magnetic field in order to understand the vortex dynamics in the grain boundaries.
- Measurements and modeling of the intermodulation distortion (IMD) in grain boundaries
- Comparison between patterned and unpatterned films to understand the role of patterning in the measured nonlinearities.
- Overall objective is better films for microwave components for application in communication and radar systems

STATUS OF EFFORT

• We have completed the measurements of the nonlinear impedance of grain boundaries fabricated on bicrystal substrates with angles from 2 to 24 degrees to investigate the role of small angle grain boundaries on the nonlinear impedance of thin films. This was the subject of Joe Habib's Ph.D. thesis.

- The effect of the patterning process on the nonlinearity of the microwave surface resistance R_S of YBCO thin films has been completed. With the use of a sapphire dielectric resonator and a stripline resonator, the microwave R_S was measured before and after the patterning process, as a function of temperature and the rf peak magnetic field in the film. Experimental and modeled results show that the patterning has no observable effect on the microwave residual R_S or on the power dependence of R_S . This was in part the subject of Hao Xin's Ph. D. thesis
- Measurements of the microwave intermodulation distortion in engineered bicrystal high temperature superconducting (HTS) YBCO grain boundaries have been completed. The two-tone intermodulation of thin YBCO plain films (without engineered grain boundary junctions) and a series of films with engineered bicrystal grain boundary junctions of misorientation angles between 2 and 24 degrees were measured. This was in part the subject of Hao Xin's Ph. D. thesis.
- We have begun the measurements of the nonlinear surface impedance of YBCO films doped with Zn Ni and Ca impurities. This is in collaboration with the group of Prof. Gad Koren at the Technion in Israel.
- We have also begun measurements of the nonlinear microwave properties of films with varying oxygen content. This is in collaboration with the group of Prof. Richard Humphreys at DERA UK and Dr. Matthias Hein of the University of Wuppertal in Germany.

ACCOMPLISHMENTS/NEW FINDINGS

The research in microwave power handling of high-Tc thin films has focused on four areas: 1. Measurements of the nonlinear impedance of and intermodulation distortion in grain boundaries as a function of grain boundary angle. 2. Effect of patterning on nonlinearities. 3. Systematic measurements of nonlinear response in films as a function of oxygen doping. 4. Measurements of the nonlinearities as a function of impurity doping with Zn, Ni, and Ca doping.

1. Measurements of the nonlinear impedance of and intermodulation distortion in grain boundaries as a function of grain boundary angle

Microwave-frequency power-dependence measurements have been performed on thin-film YBa₂Cu₃O_{7- δ} grain boundaries engineered on sapphire bicrystal substrates with misorientation angles of $\theta = 2, 5, 10$, and 24°. A stripline-resonator measurement technique is employed. The rf results are compared to dc measurements performed on a four-point test structure on the same substrate as the resonator. The measurements demonstrate that low-angle grain boundaries ($\theta < 10^\circ$) have little effect on the rf power handling, while the high-angle grain boundaries ($\theta = 24^\circ$) cause large nonlinear losses due to Josephson vortices created by rf currents.

These are the first direct measurements of the microwave properties of high- T_c engineered grain boundaries. Understanding the microwave nonlinear behavior of grain

2

boundaries is necessary to understanding the power dependence of thin films, since the defects are most likely to include grain boundaries and stoichiometric defects that behave like junctions. Knowledge of the microwave properties of the Josephson junctions is also of great importance for the application the junctions in microwave devices or high-speed digital circuits.

The measurements of engineered grain boundaries have demonstrated that the lowangle grain boundaries are not weak links at microwave frequencies and therefore not the source of the nonlinearities in high-quality epitaxial thin films that have been shown from X-ray measurements to contain grain boundaries of less than 1°. For angles less than 5° no reduction of rf critical current was observed, indicating that the critical currents of the low-angle grain boundaries are indistinguishable from the rest of the film. This work was the subject of Joe Habib's Ph. D. thesis

We have also measured the intermodulation distortion in the same series of grain boundaries. We found, in like manner to the impedance measurements, that low angle grain boundaries do not contribute to the IMD observed in high-quality films. The IMD work was a part of Hao Xin's Ph. D. thesis.

2. Effect of patterning on nonlinearities.

A proposed source of nonlinear behavior is flux penetration. This source has been thought to be especially relevant to patterned films in stripline and coplanar kinds of geometries where the edge of the film possibly contains a damaged region, and because of the current distribution in such structures, the rf magnetic field is enhanced and perpendicular to the plane of the film at the patterned edge. This confluence of effects has been proposed as a possible mechanism. This source of nonlinear behavior is also amenable to experimental verification.

We have investigated the effects of patterning on the nonlinear surface impedance by measuring YBCO films, first in the unpatterned state in a dielectric resonator and subsequently, after photolithographic patterning, in a stripline resonator. We found no significant difference between the two sets of measurements, leading to the conclusion that patterning damage is inconsequential for the nonlinear behavior. This result also argues strongly against flux penetration as the cause of the nonlinear behavior, since the rf magnetic field distributions in the two resonators are very substantially different. In the dielectric resonator, the current flows circularly, and the rf magnetic field lies in the plane of the film and points in the radial direction, presumably a direction that is unfavorable for flux penetration. In contrast, the current flows along the length direction of the stripline and the magnetic fields wrap around the strip, parallel to the film plane at the center but perpendicular at the edges, a direction favoring flux penetration. The agreement of the two sets of measurements in spite of the completely different field orientations strongly indicates that flux penetration is not the source of the low-field nonlinearity. We were able to fit the data for the two experimental geometries with a current-dependent nonlinear conductivity, indicating that the nonlinear behavior is current driven rather than magnetic field driven. It is still possible that at high values of the rf power and magnetic field, flux penetration still plays a significant role. This work was a part of Hao Xin's Ph. D. thesis.

3. Systematic measurements of nonlinear response in films as a function of oxygen doping

Another avenue of exploration is the effect of oxygen doping. We have measured a series of films with varying oxygen content. Epitaxial YBCO films were prepared by electronbeam coevaporation on MgO substrates at DERA in the U.K. The films were deposited within a few percent of the 1:2:3 cation stoichiometry as judged from their morphology and energy dispersive X-ray analysis. Oxygen content and ordering were adjusted by repeated plasma annealing in argon or activated oxygen. The final oxygenation level was judged from the critical temperature T_c , critical current density J_c and flux creep rate derived from DC magnetization measurements, and from the c-axis lattice parameter obtained from X-ray diffraction. We have investigated films prepared in three batches with oxygen contents ranging from highly underdoped to slightly overdoped. We have found that underdoping increases the nonlinear R_s but affects the low power linear surface impedance very little if at all. The optimally doped film and the overdoped film exhibit nearly the same nonlinearity, but the overdoped film shows a slight indication of better performance at high power.

4. Measurements of the nonlinearities as a function of impurity doping with Zn, Ni and Ca doping

The effects of impurities on the nonlinear behavior are now being investigated. It has always been considered that nonlinearities can result from regions of suppressed order parameter. Doping with impurities might introduce regions either with normal state conduction or with suppressed order parameter. In single crystals of BSCCO, zinc and nickel impurities have been shown to suppress the order parameter over several atomic sites. Furthermore, Ni is magnetic, and thus shows the role of magnetic impurities. Ni and Zn have been shown to substitute for the Cu in the CuO planes and chains of YBCO. Doping of YBCO with calcium is also of interest because Ca is believed to substitute for the Y and provide more carriers giving the same result as overdoping with oxygen. In addition, Ca has been shown to improve the properties of grain boundaries.

To carry out these investigations we have obtained films of YBCO deposited at the Technion in Israel by laser ablation, doped with two atomic % Ni and Zn, i.e. YBa₂Ni_{0.06}Cu_{2.94}O_{7- δ} and YBa₂Zn_{0.06}Cu_{2.94}O_{7- δ}, and films with 30% Ca doping Y_{0.7}Ca_{0.3}Ba₂Cu_{2.94}O_{7- δ}, on LaAlO₃ substrates. Films without any impurity doping and with optimum oxygen doping were also deposited at the same time for comparison purposes.

The impurity-doped films show slightly lower R_s than the pure film. Overall, however, the reduction is relatively small and there is not a dramatic effect from the inclusion of the impurities in the films. The Zn-and Ni-doped films have a very substantial increase in nonlinearity relative to the reference pure film while the Ca-doped film shows only a very small increase, although the Ca doping is much higher than the Ni and Zn doping. The $R_s(I_{\rm ff})$ of Ca-doped film is essentially unchanged from that of the pure reference film. While the magnitude of the nonlinear R_s is different, the shape of the Ni and Zn curves is

similar to that of the Ca and pure films. Similar results have been obtained with two separate sets of films, indicating that this is reproducible. Intermodulation distortion (IMD) measurements also indicate that the Zn- and Ni-doped films have larger IMD than the pure film.

These results are true for the entire range of temperatures. Since the Zn and Ni are believed to substitute for the Cu and the Ca for the Y, it seems that the nonlinearities are most likely generated by the disorder and spatial variations of the order parameter in the CuO planes. The small effect from the Ca doping also suggests that cation disorder plays a very small role in the generation of nonlinear effects. It also seems that magnetic effects do not influence the nonlinearities significantly. The Ni ion has a magnetic moment while the Zn ion has zero moment yet the effects of the two species are indistinguishable.

Doping experiments will continue and will be a part of Sang-Hoon Park's Ph. D. thesis.

PERSONNEL SUPPORTED

- Dr. Daniel Oates Visiting Scientist and Lincoln Laboratory Staff
- Faculty: Professor Mildred Dresselhaus
- Graduate Students: Hao Xin, Ph. D. Student, Sang-Hoon Park Ph.D. Student
- Dr. Gene Dresselhaus Research Staff

Theses

Youssef Habib, "Measurements and modeling of the microwave impedance in high- T_c grain boundary Josephson junctions: Josephson fluxon generation and vortex dynamics", Massachusetts Institute of Technology Ph. D Thesis, Department of Physics, February, 1999.

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N. Belk, D. E. Oates, D. A. Feld, G. Dresselhaus, and M. S. Dresselhaus, "Linear and Nonlinear Microwave Dynamics of Vortices in YBa₂Cu₃O_{7-x} Thin Films," *Phys. Rev.* B56, pp. 11966-11978 (1997).

Invited Conference Papers

.

"Nonlinear Surface Impedance, Intermodulation Distortion, and Harmonic Generation" D. E. Oates

Sixth Symposium on High-Temperature Superconductors in High-Frequency Fields, May, 2000 Capri Italy

"Nonlinear Behavior of Superconducting Devices," D. E. Oates NATO Advanced Study Institute, Millau France, Sept, 1999

"Measurements and Modeling of Microwave Impedance of High-*T*_c Grain-Boundaries," D. E. Oates, Y. M. Habib, C. J. Lehner, L. R. Vale, R .H. Ono G. Dresselhaus, and M. S. Dresselhaus Applied Superconductivity Conference, Palm Desert CA Sept 13-18, 1998

"Flux dynamics" D. E .Oates,

Fifth Symposium on High-Temperature Superconductors in High-Frequency Fields, June 22-25 1998, Stockholm Sweden

Contributed Conference Papers and Posters

"Microwave intermodulation distortion in bicrystal YBCO grain boundary junctions," H. Xin, D. E. Oates, G. Dresselhaus, and M. S. Dresselhaus, Oral Presentation, Materials Research Society, Boston MA, December 2000.

"Nonlinear microwave response of YBaCuO films of varying oxygen content before and after patterning,"

M.A. Hein, A. Porch, A. V. Velichko, R. G. Humphreys, P. J. Hirst, and D. E. Oates Oral Presentation Applied Superconductivity Conference Virginia Beach, VA Sept 17-22, 2000

"Observation of Individual Josephson Vortices in YBCO Bicrystal Grain-Boundary Junctions"

D. E. Oates, H. Xin, G. Dresselhaus, and M. S. Dresselhaus

Oral Presentation Applied Superconductivity Conference Virginia Beach, VA Sept 17-22, 2000

"Nonlinear Surface Impedance and Intermodulation Products: Relationship to Oxygen Content,".

D. E. Oates, M. A. Hein, A. Porch, A. V. Velichko, R. G. Humphreys, and P. J. Hirst Poster presentation, VI Symposium High Temperature Superconductors in High Frequency Fields, Capri Italy May 24-27, 2000

"Observation and Dynamics of Individual Josephson Vortices in YBCO Grain-Boundary Junction,",

H. Xin, D. E. Oates, G. F. Dresselhaus, and M. S. Dresselhaus Oral Presentation, American Physical Society 2000 March Meeting. Minneapolis MN.

"Microwave and dc Fluxon Dynamics in Bicrystal YBCO Grain-Boundary Josephson Junctions,"

H. Xin, D. E. Oates, G. F. Dresselhaus, and M. S. Dresselhaus Poster presentation, Materials Research Society Symposium Boston MA. Nov. 30-Dec. 2 1999

"Measurements and Modeling of the Microwave Impedance in High- T_c Grain Boundary Josephson Junctions: Josephson Fluxon Generation and Vortex Dynamics," Y. M Habib, C. J. Lehner, D. E. Oates, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus,

Oral Presentation, Materials Research Society, Boston MA, December 1997.

"Properties of Grain-Boundary Josephson Junctions at Microwave Frequencies,"

Y. M Habib, C. J. Lehner, D. E. Oates, L. R. Vale, R. H. Ono, G. Dresselhaus, and M. S. Dresselhaus,

Oral Presentation, American Physical Society March Meeting, Los Angeles, CA, March 1998.

"Coupled-Grain/RSJ Series Array for Modeling of Nonlinear Microwave Surface Impedance of YBCO Thin Films"

J. Herd, H. Xin, and D. E. Oates

Applied Superconductivity Conference, Palm Desert CA Sept 13-18, 1998

"Comparison of the Microwave Frequency Power Dependence of Surface Resistance of Unpatterned and Patterned YBCO Thin Films,"

H. Xin, D. E. Oates, and A. C. Anderson

Oral Presentation Materials Research Society Fall Meeting Boston MA, Nov. 30,-Dec. 4, 1998.

APPENDIX

Project Title: "Studies of High Tc Superconducting Films High Power Microwave Applications" Subcontract to AFOSR Grant # : F49620-98-1-0021

Final report for period 10/15/98-4/14/01

P.I.: Prof. S. Sridhar, Northeastern University

OBJECTIVES

Investigate the nonlinear microwave response of superconducting films and crystals, characterize and understand the low and high power response of superconductors, particularly high temperature superconducting cuprates, with a view to improving the performance of superconducting microwave devices.

ACCOMPLISHMENTS

This project explored the phenomenology and experimental aspects of high-frequency vortex dynamics and electromagnetic response in a variety of superconductors. The materials probed range from single crystals of low-TC magnetic superconductors, such as the Boro-carbides, which are principally of pedagogical interest, to thin films of high-TC oxide superconductors, which have the potential to radically alter the state of the art of electronics technology.

A two-pronged attack involving experimental investigation and numerical modeling was used to understand the phenomenology of microwave nonlinearity in high temperature superconductors, and to circumvent it for device applications. Measurements in presence of a weak DC magnetic field revealed a new phenomenon, now known as the "magnetic recovery effect", that the applied field can actually serve to decrease both surface resistance and nonlinearity.

Measurements of harmonic generation, a quantity of most direct relevance to passive circuit design, were carried out on a resonant structure. A methodology was developed that is particularly suited for single-tone measurements on high-Q structures and also gives information on other quantities such as the penetration depth. It was found that the data can be quantitatively explained by taking into account the power dependence of the insertion loss and the contribution of higher order terms of the nonlinear impedance, without having to resort to any additional adhoc assumptions. Measurement of harmonic power on a suspended stripline microwave resonator patterned out of thin film YBa2Cu3O7-8 (YBCO) was carried out as a function of temperature and microwave power. The third harmonic power P3 shows a subtle nonlinear dependence on the fundamental power P1 on a log-log scale. Fit to a straight-line yields slopes of P₃ vs. P₁ between 1.5 to 3, contrary to elementary calculations that predict a slope of exactly 3. It is shown that third harmonic power generated from higher order terms in the nonlinear impedance could account for the discrepancy quantitatively without resorting to any additional adhoc assumptions. Results of measurement of fifth harmonic power were carried out as a confirmation. A small second harmonic power was also observed that does not show anomalous power dependence within the experimental accuracy.

Although the critical state model has been proven to provide the most accurate picture of nonlinear response of superconductors as shown from surface impedance and harmonic generation measurements, so far the model could be solved only for highly idealized geometries. A numerical paradigm, with physical assumptions equivalent to those of the critical state, was implemented that yields results that quantitatively agree with the analytical calculations in the appropriate limits but is not restricted to the unphysical idealizations. This lays the foundation of a

computer aided design framework for passive microwave devices that is explicitly developed for superconducting materials.

We showed that a common feature of temperature-dependent microwave absorption is the presence of absorption peaks. ac loss peaks can arise when the internal T-dependent magnetic relaxation time crosses the measurement frequency. These features are observed in the insulating (Sr_xCa_{14-x} Cu₂₄O₄₁, La_{5/3}Sr_{1/3}NiO₄ and YBa₂Cu₃O_{6.0}), pseudo-gap (T > T_c in underdoped YBa₂Cu₃O_{7.8}, Hg : 1223 and Hg : 1201) and superconducting (T < T c) states of the oxides. The commonality of these features suggests a microwave loss mechanism arising from inhomogeneous charge distribution, rather than a quasiparticle origin, for the so-called "conductivity" peaks observed in the cuprate superconductors.

The nonlinear dependence on applied ac field (b_{ω}) or current (i_{ω}) of the microwave (ac) impedance $R_{\omega}+iX_{\omega}$ of both short and long Josephson junctions is calculated under a variety of excitation conditions. The dependence on the junction width is studied, for both field symmetric (current antisymmetric) and field anti-symmetric (current symmetric) excitation configurations. The resistance shows steplike features every time a fluxon (soliton) enters the junction, with a corresponding phase slip seen in the reactance. For finite widths the interference of fluxons leads to some interesting effects which are described. Many of these calculated results are observed in microwave impedance measurements on intrinsic and fabricated Josephson junctions in the high temperature superconductors. When a dc field (b_{dc}) or current (i_{dc}) is applied, interesting phase locking effects are observed in the ac impedance Z_w . In particular an almost periodic dependence on the dc bias is seen similar to that observed in microwave experiments at very low dc field bias. These results are generic to all systems with a $\cos(\phi)$ potential in the overdamped limit and subjected to an ac drive.

PUBLICATIONS

1. "Non-linear Microwave Impedance of Short and Long Josephson Junction", Z. Zhai, Patanjali V. Parimi, and S. Sridhar, Phys. Rev. B, **59**, 9573 (1999).

2. "Observation of Individual Josephson Vortices in YBCO Bicrystal Grain-boundary Junctions", H.Xin, D.E.Oates, S.Sridhar, G.Dresselhaus and M.S.Dresselhaus, Phys. Rev. B. (Rapid Comm.), 61, R14952 (2000)

3. "Computer-aided modeling of superconducting striplines with ground planes using critical state models", T.Dasgupta, J.S.Derov and S.Sridhar, J. of Applied Physics, (accepted for publication, 2001).

4. D.P.Choudhury, "High-frequency electrodynamic response of novel superconductors: fundamentals and applications", Ph.D. Thesis, Northeastern University (1999).

INVITED PRESENTATIONS AT CONFERENCES

- 1. "Quantum Chaos: Theory and Applications", Cocoyoc, Mexico, July 8-13, 2001.
- 2. 84th Statistical Mechanics Conf, Rutgers University, Dec. 17-19, 2000.
- 3. "Collosal Magneto resistance and , Telluride Workshop, Aug , 2000.
- 4. Nobel Symposium on "Quantum Chaos Y2K", Backaskogs Castle, Sweden, Jun 13-16, 2000.
- 5. Int. Conf. On "HTSC in High Frequency Fields", Capri, Italy, May 2000.
- 6. Invited talk at American Physical Society, March Meeting, Minneapolis, MN, Mar. 20-25.
- 7. ONR Workshop on "Stripes and Antiferromagnetism in HTS Electronics", March 4-5, 2000.
- Institute for Nuclear Theory Workshop on "Complex Systems and Quantum Chaos", U. of Washington, Seattle, Mar. 12-16, 2000.
- 9. M2S-HTSC-VI, Houston, Texas, Feb. 20-25, 2000.
- 10. 5 Lectures at the Summer School on "Quantum Chaos", Canberrra, Australia, Jan.11 to Feb.22, 1999.

11. "Electrodynamics of Spin and Charge in Cuprates and Nickelates", International Conference on High Temperature Superconductivity, Miami, FL, Jan.7 to 12, 1999.

INVITED PRESENTATIONS AT ACADEMIC AND RESEARCH INSTITUTIONS

1. TIFR, Mumbai, 7/3/2001.

2. Raman Research Institute, 6/26/2001

- 3. IISc, Bangalore, 6/21/2001
- 4. TRW, Albuquerque, May 23, 2001
- 5. MACOM, Nov. 2000
- 6. ISSP, Chernogolovka, Russia, June 2000.
- 7. IISC, Bangalore, June 2000
- 8. TIFR, Bombay, May 2000
- 9. Brandeis University, Oct. 5, 1999.
- 10. Tufts University, Sept 1999.
- 11. Naval Research Laboratories, June 2, 1999.
- 12. Los Alamos National Labs, Feb. 10, 1999
- 13. U. of Florida, Gainesville, April '99
- 14. U. of NSW, Sydney, Australia, Jan. 25, 1999.
- 15. U. of Miami, Nov.17, 1998.