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New techniques for growing epitaxial thin films of high temperature superconducting cuprates have been developed. The electrical and superconducting properties of the thin films have been investigated by transport, magnetic, optical and tunneling measurements. Insights into the basic nature of the superconducting interaction and the physics of some superconducting devices have been gained.

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**STANFORD CENTER FOR RESEARCH ON
SUPERCONDUCTIVITY
AND
SUPERCONDUCTIVE ELECTRONICS**

The impetus for the formation of a Center for research on Superconductivity and superconducting electronics at Stanford was provided by the open ended potential of the newly discovered high Tc copper oxide superconductors for electronic applications. The purpose of the Center was to enhance the ongoing work of the programs of Professors Kapitulnik, Geballe and Beasley in superconductivity and superconducting electronics, including further collaborations with other faculty members at Stanford, with other universities, and with government and industrial laboratories.

The goals envisioned at the start of the center were as follows.

--to determine conclusively the physical mechanism(s) underlying the superconductivity of these new materials, and on the basis of this knowledge to seek improved superconductors, possibly even with higher transition temperatures.

--to develop an understanding of the physical properties of these new superconductors with an eye toward developing generically new, as well as known, superconducting devices; and to produce prototype devices as proofs of principle.

--to synthesize, characterize and optimize thin films and multilayer thin film structures of the new superconductors suitable for physical study and for electronic applications.

--to be prepared to capitalize on any further breakthroughs in new superconducting materials.

--to produce a cadre of young scientists and engineers knowledgeable and experienced in the science and technology of the new superconducting materials.

These goals have been fully achieved in some cases and partly achieved in others as can be seen in the list of accomplishments which are summarized below in sections 2, 3, 4 and 5 and are documented in the publications listed in section 6. Those who have received PhD's or post doctoral support under our program are listed in section 7, along with present location.

II. Accomplishments in thin film synthesis and processing

(a) We have developed the 90° off axis sputtering process for growing films from composite targets in which the composition of the target is preserved in the films and in which the desired structure is formed directly from the vapor- i.e., *in situ* growth. The *in situ* feature is desirable because no solid state reactions with accompanying changes in volume, morphology, etc., that degrade the quality of the film are involved. This technology has been transferred to a large number of university and industrial laboratories in the United States by the Stanford Center by means of publications, students who have received PhD's and gone on to other organizations, and visitor exchange. The argon-oxygen sputtering pressure is high enough so that the transport through the plasma is mainly diffusive. The mobility of the species at the surface during the growth is critical so that careful control of the surface temperature is required.

(b) We have found, contrary to our own and the expectations of others that the surfaces and the crystal perfection of a-axis films can be superior to c-axis films. The a-axis films grow at lower temperatures, and form better interfaces. A-axis films are thus excellent candidates for making good sandwich-type Josephson junctions, both of the tunneling and proximity effect varieties. There are no twins in the a-axis films but there are lots of grain boundaries, because the in-plane b and c/3 cell lengths are almost the same. In contrast, in the c-axis films, the in-plane a and b axes differ by $\sim 2\%$, resulting in heavy twinning.

(c) The relatively good square lattice matching between superconducting YBCO and insulating PrBCO makes it possible to grow superlattices, which as judged by the ion channeling experiments, are as perfect as single crystals of either compound, even though the channeling ions must traverse 20 interfaces. TEM cross sections show the superlattice by an out-of-focus image which is sensitive to the electron density. The in-focus image of the atomic rows shows that the structure maintains itself right through the interfaces. The smoothness of the surface is such that no features can be imaged with our most sensitive SEM (resolution $\sim 100\text{\AA}$). The atomic force microscope shows undulations of $40\text{\AA}$ over distances thousands of $\text{\AA}$. X-ray diffraction scans indicating superlattice satellite peaks also demonstrate high crystalline perfection. The grain boundaries between the b and c in-plane orientations in the a-axis films cause excess resistance. Critical currents for a-axis films are reduced by about two orders of

magnitude from the c-axis films because of these grain boundaries. This should not degrade vertical transport, however, and thus should not be a problem in making sandwich type junctions.

(d) Earlier attempts to produce high quality thin films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) on Si substrates were thwarted by interface reactions and thermal expansion mismatch. In a collaboration with Xerox several epitaxial buffer layer systems including yttria-stabilized zirconia and MgO which allow the epitaxial growth of YBCO on Si without chemical reaction have been found. All of the materials may be grown in a single process by laser deposition. Pulsed laser deposition has been found to be a rapid and versatile technique for exploring new epitaxial systems on a variety of substrates; this work has provided the first extension of pulsed laser deposition to epitaxy on semiconductor substrates. Although direct epitaxy of YBCO on silicon may not be possible, buffer layers permit the growth of device quality YBCO films with critical current densities in excess of 10^6 A/cm^2 at 77 K. Methods have been developed to pattern these films in-situ by removing the buffer layer in selected areas to produce isolated superconducting mesas. The complications of thermally induced tensile stress can be avoided through the use of silicon-on-sapphire instead of silicon. This work has revealed methods for controlling the in-plane epitaxy of YBCO films on zirconia via homoepitaxy and nucleation-initiating monolayers. Ongoing research has demonstrated that some of the epitaxial growth processes developed for Si can be adapted to produce similar results on GaAs substrates.

(e) The deposition temperature and pressure for thin films of $\text{YBa}_2\text{Cu}_3\text{O}_7$ made by different processes have been compared. Several anomalous results are found. For O_2 pressure $\leq 10 \text{ mTorr}$, films with average composition substantially off the 1:2:3 stoichiometry have more bulk-like properties, including higher T_c 's, compared to films made on-stoichiometry. Films made at lower oxygen pressure also have c -axis lattice constant expanded compared to the films made at higher pressures ($> 100 \text{ mTorr}$). The films show evidence that they are excess hole-doped compared to the ideal material. Altogether, the results strongly suggest that metal-atom point-like defects are quenched into the films. A model based on the presence of Ba-for-Y substitution has been developed and found to be consistent with the experimental results. We suggest that these defects might be a generic property of *in-situ* films.

(f) The CuYO_2 delafossite-type structure has been identified as a second phase in thin films of Cu- and Y-rich $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (123) made by electron beam co-evaporation at conditions of low oxygen pressure; this phase occurs as extremely small, densely distributed, highly oriented precipitates within the 123 matrix. The smallest (sub-100 Å) [001] oriented precipitates of this hexagonal phase are strained to give a better lattice match to the surrounding 123 structure. The results suggest that, for thin film deposition conditions of sufficiently low temperature and pressure, there may be a tie line between $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and

CuYO₂ in the equilibrium phase diagram. The size and distribution of the precipitates is such as would be expected to influence structurally sensitive superconducting transport properties, e.g. critical currents.

(g) Highly oriented, high-quality YBCO thin films on MgO substrates were examined and found to have a high density of planar defects such as out-of-phase boundaries and low-angle grain boundaries. The origin of a large number of these boundaries is found to be imperfections in the substrate surface morphology. In addition, it was found that by varying film thickness and film oxygenation temperature, the final twin microstructure could be strongly affected, without any accompanying change in superconducting properties. For extremely high critical current material, twin boundaries do not play a dominant role in determining superconducting properties.

III. Accomplishments with the MBS Facility

At the present time the favored ways of making thin films of the high T_c materials is off-axis sputtering (perfected at Stanford) and laser ablation. It is our feeling that if a quantum improvement in the quality of the films is to be made, more powerful approaches must be brought to bear. Toward this end we have been studying an MBE-like approach in which the metallic elements are thermally evaporated in the presence of oxygen. The goal is to achieve MBE-like deposition with all its power and in situ characterization capability.

(a) A program to characterize existing polished substrates for HTSC film growth and then to modify them to improve their smoothness was begun under this program. We have used RHEED and LEED for in situ characterization, and AFM for ex situ examination. Homo-epitaxial growth of electron-beam evaporated MgO and Al₂O₃ resulted in improved atomic surface ordering compared to polished crystals and similar to that of high temperature annealed crystals, as evidenced by RHEED and LEED patterns. However, AFM scans revealed an array of in-plane ordered rectangular shaped (~ .1mm on a side) mesas (~ 100Å high). We are exploring the effect of growth temperature, surface preparation, and the presence of ion beams during growth on the surface morphology. These preliminary results demonstrate that surface characterization using RHEED and LEED, while indicating good atomic ordering of the surface, does not indicate the true long range smoothness. Published studies by others have used electron diffraction (RHEED and LEED) and x-ray diffraction to claim epitaxial growth and imply surface smoothness. Our results show that additional characterization, such as the AFM, is needed.

(b) We have grown films of the so-called "infinite lattice" copper oxide compound (CuO₂)_n (Sr)_n, using electron beam codeposition of the metals and an atomic oxygen beam from a modified ECR source. The structure was identified

by x-ray diffraction. As expected, these undoped films were insulating, although in some cases self-doping due to Sr vacancies resulted in semiconducting and metallic temperature dependencies of the resistivity. Recent work by others suggests this material can be doped so as to yield a 100K superconductor.

(c) An important, and on-going, aspect of our MBS work has been the development of rate monitoring for the electron beam feed back control that is compatible with the required high oxygen background pressures. We have been using Atomic Absorption (AA) over the past few years, but we are now extending the technology to new elements (Sr and Nd), to lower rates (goal is $\sim .1$ Å/sec), and to higher accuracies than ever achieved before (the goal is 1%). We are finding that the hollow cathode lamp sources for these elements are not sufficiently stable. With the help of the manufacturer feedback circuits are being developed to stabilize these lamp sources.

(d) A movable mutual inductance probe mounted from the wall of the MBS Loadlock/Process chamber was developed and tested. This has permitted the in situ measurement of the T_c of a thin film mounted on the recently developed liquid helium cooled substrate fixture. This mutual inductance probe should be ideal for those materials where either contacts on the substrate are not possible or desirable, and where it is not possible to remove the thin film from the vacuum for measurement. Furthermore it will permit in situ complex conductance measurements on 2-D thin film systems.

IV. Physical Property Measurements

(a) Using the off-axis sputtering deposition techniques in collaboration with Hewlett-Packard we have carried out a careful, quantitative study of the role of large-angle grain boundaries in the high residual rf losses universally observed in c-axis YBCO thin films on MgO. MgO is a good substrate material for microwave devices because it has a low dielectric constant and is cubic. The latter feature insures that the dielectric properties are isotropic in the plane and makes the design of devices simpler.

(b) The lowest accurate bound on the surface resistance, $16 \pm 3 \mu\Omega$ at 10 GHz and 4.2K, reported to date has been found for in-situ grown $YBa_2Cu_3O_7$ thin films. The increase in loss with increasing deviations of the films from the characteristics of ideal, equilibrium single crystals makes clear that values above this level are representative of extrinsic loss mechanisms. Very small quantities of c-axis material misaligned in the surface plane can contribute significant losses. Four percent volume fractions of highly misaligned c-axis grains lead to losses above $500 \mu\Omega$ at 10GHz and 4.2 K. Our data also show that, for these samples, it is unlikely that the $16\text{-}\mu\Omega$ value is dominated by loss mechanisms associated with the misaligned material. Losses of a different origin dominate. For the lower-loss films R_s is correlated to the dc transport characteristics.

(c) We have measured the amount of nonreciprocal circular birefringence of 50 to 800 Å YBa₂Cu₃O₇ films in transmission with a 15-μm beam diameter. A novel instrument with a sensitivity of 2 μrad for nonreciprocal phase shifts was developed by modifying a fiber-optic gyroscope. It is insensitive to reciprocal phase shifts. We observed no nonreciprocal phase shifts in any samples. This result casts strong doubt upon recent reports that optical nonreciprocal effects were observed in reflection. The latter, if correct would be strong evidence for an exciting theory of the superconductivity of the cuprates, the anyon theory.

(d) We have studied the relation between the magnetic shielding current in epitaxial thin films of YBa₂Cu₃O₇ and the shape of the current-voltage (J-E) characteristic of the superconductor in its critical state. The weak temperature dependence of the relaxation implies a temperature-insensitive J-E characteristic that resembles that of conventional type-II superconductors when a spatial variation of critical current density (J_c) is present. We have shown such a distribution of J_c can be used as an explanation for the apparently large and temperature-insensitive relaxation of the critical states observed in YBa₂Cu₃O_{7-δ}. We also have demonstrated the presence of edge pinning in narrow microbridges of YBaCuO.

(e) We have found a strong substrate dependence of the temperature dependence of the magnetic penetration depth $\lambda(T)$ at low temperatures. For the best samples the low temperature dependence is exponential, excluding the possibility of nodes in the energy gap in YBCO. Unlike conventional strongly-coupled superconductors, however, the inferred temperature dependence of $\lambda(T)$ is not characterized by a single-gap (scaled, isotropic) BCS temperature dependence. The observed temperature dependence can be fit with either a parallel superconductor model involving the planes and chains or a series weak link model, both with a larger gap of $2\Delta(0)/k_B T_c = 4.5$. However, other interpretations are not completely ruled out.

(f) We have used experiments on very thin films to compare and isolate two dimensional effects in the cuprates. By comparing to multilayer systems with similar parameters, we can identify the decoupling of the superconducting planes in magnetic fields at temperatures much above the irreversibility line. These results show that if the irreversibility line is to be considered a melting transition line, it implies melting of the solid state into a liquid of three dimensional flux lines.

(g) Monte Carlo simulations of a model of weakly coupled 2D superconducting planes have been used to identify two melting lines corresponding to the disappearance of the translational and the hexatic order of the vortex lattice in the planes. The Lindemann number corresponding to the translational order-disorder transition is found to be about 0.2 but to deviate from this value in the extreme density limits. We find a significant effect of cutting/reconnection of the vortices on the in-plane stiffness and the mean-square

deviation from equilibrium positions, which should have a profound effect on the dynamics of the system.

(h) We have found that in a-axis superlattices of $\text{YBa}_2\text{Cu}_3\text{O}_7/\text{PrBa}_2\text{Cu}_3\text{O}_7$ the CuO_2 planes can be divided into alternating 1D-like superconducting/insulating strips. When a magnetic field is applied in the plane of the substrate (perpendicular to the CuO_2 planes) we find that the resistive transitions of superlattices with modulation wavelengths of $96\text{\AA}$ ($48\text{\AA}$ - $\text{YBa}_2\text{Cu}_3\text{O}_7/48\text{\AA}$ - $\text{PrBa}_2\text{Cu}_3\text{O}_7$), $72\text{\AA}$ ($48\text{\AA}$ - $\text{YBa}_2\text{Cu}_3\text{O}_7/24\text{\AA}$ - $\text{PrBa}_2\text{Cu}_3\text{O}_7$), and $48\text{\AA}$ ($24\text{\AA}$ - $\text{YBa}_2\text{Cu}_3\text{O}_7/24\text{\AA}$ - $\text{PrBa}_2\text{Cu}_3\text{O}_7$) show a crossover with the upper part of the transition insensitive to magnetic fields up to 8T. The crossover temperature is controlled by the PBCO thickness. These results show that in the temperature range where we observe this insensitivity the structure behaves as 1D superconducting-insulating channels whose width is determined by the $\text{YBa}_2\text{Cu}_3\text{O}_7$ and $\text{PrBa}_2\text{Cu}_3\text{O}_7$ thicknesses. By comparing these results to those obtained on c-axis superlattices we estimated that $100\text{\AA}$ a-axis PBCO should produce the complete decoupling of the YBCO channels. The channel dimensions and the perfection of the superlattices are such that the study of quasi 1-D transport is feasible.

(i) Extensive theoretical studies of the "chiral spin liquid" state, which is the starting point for calculations of high- T_c superconductors based on the anyon pairing idea, have been carried out. This has led to the development of a gauge theory formalism that has made realistic calculations possible for the first time. The optical conductivity of the t-J model, thought by many theorists to be an appropriate idealization of the high- T_c problem, has been calculated using this formalism, and shown to agree well with both exact cluster calculations of the model and the mid-infrared conductivity seen in real high- T_c materials. Through this work the way has been opened which should permit calculation of accessible experimental quantities, such as the photoemission spectrum, the a.c. spin susceptibility, pairing susceptibility, and the energy gap.

V. Device Physics Studies

(a) We have investigated the anisotropy of the Josephson proximity coupling of conventional low T_c superconductors to YBCO thin films. Devices that couple to YBCO along the c-axis and along the a-axis have been prepared and their properties compared. The superconducting coherence length is very different along these two directions, the a-axis coherence length being much longer than that along the c-axis. The results show Josephson proximity coupling to a low temperature superconductor (Pb) can be achieved on an a-axis film but not on a c-axis film, despite the fact that a state-of-the-art low contact resistance was achieved.

(b) Studies of the proximity coupling of a-axis $\text{YBa}_2\text{Cu}_3\text{O}_7$ through Ag to a BCS superconductor have shown that the $I_c R$ product is systematically much smaller than the BCS limit of $\sim 5\text{mV}$. The observed $I_c R$ values range from 10 to $100\ \mu\text{V}$ in the $\text{YBa}_2\text{Cu}_3\text{O}_7$ a-direction to apparently zero in the c-direction. The reduced $I_c R$ in the a-direction can be understood as arising from a large boundary resistance, consistent with the mismatch of carrier densities across the $\text{YBa}_2\text{Cu}_3\text{O}_7$ -Ag interface. However, the same analysis gives an $I_c R$ in the c-oriented junctions that is at least three orders of magnitude larger than observed.

VI. List of publications in which the research was fully or partially supported by the Center in the form of stipends, and/or research expenditures, and/or facilities.

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