



Final Technical Report  
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ADVANCED SUPERCONDUCTING MATERIALS  
  
AND DEVICE CONCEPTS  
  
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## **Goal of the Program**

The goal of this program was to carry out studies of the high-temperature superconductors and related materials in support of the applications of superconductivity relevant to the Air Force. More specifically we aimed 1) to identify materials and physical issues fundamental to the understanding of the high temperature superconductors relevant to superconducting electronics, 2) to learn how to grow and characterize these materials in thin film and/or single crystal form, 3) to study their relevant physical properties and 4) to connect our results to issues of practical importance.

## **Accomplishments**

Below we list our accomplishments organized around the materials studied. In each case we summarize both what was learned and how it relates to superconducting electronic applications. Detailed accounts of how these materials were prepared and the physical property studies can be found in the indicated references.

### **1) 123 YBaCuO .**

123 YBaCuO is the leading material of choice in all high-temperature superconducting electronic applications. There is also some applications work on 2223 TlBaCaCuO, which has a higher superconducting transition temperature. In this program we grew and studied single crystals of 123 YBCO. Some collaborative measurements were also made on crystals grown by the British Columbia group headed by Walter Hardy. The most important result was our study of the field dependence of the heat capacity in the presence of an applied magnetic field. By applying a field it was possible to extract cleanly the electronic part of the heat capacity, free of contributions from lattice excitations, which are not field dependent [1, 2, 3]. The observed  $H^{1/2}$  dependence of the heat capacity is a consequence of the novel d-wave symmetry of the pair wavefunction in the high-temperature superconductors. This work is one of the major bricks in the foundation of the case for d-wave pairing in the cuprate superconductors.

We have also been among the leaders in understanding the important implications of the d-wave superconductivity for superconducting electronics. For example, it is necessary to make Josephson junctions along particular crystallographic directions, if strong Josephson coupling is to be obtained. Also, d-wave symmetry produces nodes in the single-particle excitation spectrum of the superconductor, suggesting that excited (normal like) electrons may relax very much faster in a d-wave superconductor than in a conventional s-wave superconductor. This fact is important in understanding and assessing the utility of various non equilibrium superconducting devices. Finally, under DoD URI support we conceived a of a new circuit family of complementary Josephson

junctions (the analog of CMOS) that uses the d-wave symmetry of the high-temperature superconductors as one means of creating a complementary Josephson junction [4].

Finally, we have also studied the thermal boundary resistance between 123 YBaCuO thin films and the substrates on which they are deposited [5]. Our results confirm earlier results claiming that this thermal boundary resistance is larger than expected on the basis of the well known acoustic mismatch model. The thermal boundary resistance is of obvious relevance to questions of self heating in superconducting devices made from the high temperature superconductors. In other work we developed an atomic oxygen annealing process capable of cleaning the surface of YBCO thin films, so as to provide low electrical boundary resistances [6].

## **2) 2212 BiSrCaCuO.**

2212 BiSrCaCuO has been the most important material for studying spectroscopically the anisotropy of the energy gap in the high temperature superconductors. This is because this compound is extremely two dimensional and cleaves naturally between charge neutral planes, leading to very clean surfaces for photoemission spectroscopy. We have developed procedures for growing some of the best available single crystals of 2212 BiSrCaCuO [7]. These have been used in a collaboration with Z.X. Shen to first demonstrate anisotropy in the energy gap of the high-temperature superconductors consistent with d-wave superconductivity [8]. This work of Shen based on our materials is another important brick in the foundations of d-wave pairing in the cuprate superconductors.

We also demonstrated that there was no Josephson coupling along the c-axis (perpendicular to the CuO<sub>2</sub> planes) between 2212 BiSrCaCuO and a low temperature conventional s-wave superconductor (Pb), whereas very good Josephson coupling could be obtained to the ab-axes (along the CuO<sub>2</sub> planes) [9]. These results provide further evidence for the d-wave nature of the pairing state in 2212 BiSrCaCuO. We have also extended the theory of magnetic diffraction characteristics of Josephson junctions to include junctions with extreme type-II superconductors, such as BSCCO, when the junction is made along the a-axis [10].

The importance of the pairing symmetry to superconducting electronics was discussed in 1) above.

We have also studied the vortex phases in 2212 BiSrCaCuO [11]. Our results along with those of others have definitively established the existence of various phases associated with the tendency of the vortex lattice to fluctuate dramatically even at very low temperatures in such highly anisotropic materials. This fact is the origin of the very restricted range of utility of this material as a high-current, high-field superconductor.

These deficiencies of 2212 BiSrCaCuO have led us investigate under EPRI support the use of biaxially textured buffer layers for the growth of YBCO films on engineering substrates as an alternative. This work was initiated under AFOSR support in an earlier grant period and was subsequently transitioned to EPRI support.

### 3) $SrRuO_3$ .

$SrRuO_3$  is unusual in being a conducting oxide and an itinerant ferromagnet. Having the perovskite crystals structure and lattice constants like those of the high temperature superconductors, it is an obvious candidate for the normal barrier material in superconducting/normal metal/superconducting (SNS) Josephson junctions. We have grown both single crystals and thin films of this material for physical study. Also, in collaboration with Kookrin Char of Conductus we have examined its potential as the N-barrier material in 123 YBCO SNS junctions [12].

On the physical property side, we have found that while the magnetism of this material appears understandable in terms of established ideas, this is certainly not the case for electrical transport. In fact we have established that this material belongs in a new class of materials known as bad metals [13,14]. Bad metals are those in which the temperature dependence of the resistivity is temperature dependent in a quasi linear fashion to temperatures well above that where the electron mean free path is small compared to the Fermi wavelength of the conduction electrons. Put more simply and directly, beyond the range in which Boltzmann transport theory can be expected to apply. This result raises new and possible deep theoretical questions. It also obviously relates to the use of such materials as the “N” material in an SNS proximity junction.

More recently, in collaboration with Schlessinger at the University of California at Santa Cruz, we have observed that the infrared conductivity of this material cannot be understood in terms of the conventional Drude theory and is more like that seen in the high temperature superconductors than that of a conventional metal. We have also measured directly the ferromagnetic domain wall resistance in this material and find it to be anomalously large compared with conventional elemental ferromagnets [15]. This result suggests that there may be a very high degree of spin polarization present in the conduction band.

On the applications side, we demonstrated with Char that Josephson coupling could be achieved using  $SrRuO_3$  as the normal barrier in SNS proximity effect barriers, but that a large interface resistance between the  $SrRuO_3$  and the 123 YBCO films made it less attractive as a practical N material compared with alternative barrier materials developed later, such as Co-doped YBCO [16]. The origins of this large interface resistance are not definitively understood.

We continue to be interested in proximity effect with this material because theory predicts that the superconductivity induced into a ferromagnetic by the proximity effect should have a novel oscillatory decay, as opposed to the conventional exponential decay. Under DoD URI support we have conceived of a new superconducting memory device based on this effect, which operates very analogously to the so-called giant magnetoresistance devices that are important in magnetic memory and sensor applications [17]. The physical principle are completely different in our device, however.

Further results with SrRuO<sub>3</sub> are discussed just below in our discussion of ferroelectric field effect doping.

#### 4) SrCuO<sub>2</sub>

SrCuO<sub>2</sub> is the so-called infinite layer material of high temperature superconductivity. The name derives from the fact that its structure is the three dimensional extension of the inner superconducting CuO<sub>2</sub> layers found in the crystal structure of the high temperature cuprate superconductors. It is the doping of these CuO<sub>2</sub> layers that leads to high temperature superconductivity in these materials. Obviously there has been great interest in trying to grow this material and then somehow dope it. Most researchers tried to achieve doping by chemical substitution, others by the use of multilayering to achieve an analog of the naturally occurring structure.

In our work we took a unique approach and introduced the notion of depositing this material epitaxially onto a ferroelectric (the well known PZT) and tried to induce doping by a field effect of the ferroelectric. Using this technique we were able to change the resistivity of a very thin layer of SrCuO<sub>2</sub> epitaxially deposited onto PZT [18]. We were not, however, able to induce superconductivity into the SrCuO<sub>2</sub>.

We also demonstrated that we could change substantially the resistivity of a very thin layer of SrRuO<sub>3</sub> deposited epitaxially on PZT [19]. We further were able to write submicron ferroelectric domains into the PZT using a scanning probe and read out the polarization using the field effect on SrRuO<sub>3</sub> [20]. In principle, these results provide a demonstration of all the elements needed to make a very high density ferroelectric memory.

#### 5) Manganites

Manganites with perovskite-related structures exhibit a rich variety of transitions and magnetic phenomena. These highly-correlated electronic systems have been intensively studied recently by virtue of the “colossal negative magnetoresistance” observed in La<sub>0.67</sub>Ca<sub>0.33</sub>MnO<sub>3</sub> thin films upon cooling through the insulator-metal transition, which occurs in the vicinity of the ferromagnetic transition. Application of a magnetic field favors the metallic state.

In collaboration with Hiskes and coworkers at Hewlett Packard, we showed that the observed very large magnetoresistance was due to extrinsic effects (i.e., grain boundaries, etc. ) which suppressed the insulator-metal transition, moving it to lower temperatures [21]. The consequent exponentially large increase in the resistance of the insulator is due to this lower temperature, and thus is not of technological interest for a room temperature technology. We also found that with annealing at successively higher temperatures, the insulator-metal transition temperature could be raised and the negative establish similar, almost intrinsic behavior (Curie temperature near 265K) for  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ , whether in the form of thin films, bulk ceramics or single crystals. Studies of the conductivity in the insulating state over a wide range of temperature up to 1200K were found to favor transport by adiabatic small polarons,  $\sigma = A/T \exp(-E_a/kT)$ , over other models such as variable range hopping or simple semiconduction [22].

These studies were also extended to other concentrations of Ca from pure  $\text{LaMnO}_3$  to  $\text{CaMnO}_3$ . Adiabatic small polaron behavior was found to obtain throughout the entire doping range. Furthermore, the concentration dependence of the pre-exponential factor A explicitly shows the effects of onsite Coulomb repulsion. Unlike  $\text{SrRuO}_3$ , the ferromagnetic ordering could not be fit by classical exponents (Heisenberg, Ising) at least to within 0.01  $T_c$  as  $T_c$  is approached from above. Other unusual behavior, including a positive third order (in field) susceptibility, are evidence of the existence of ferromagnetic clusters which from neutron and muon results of others are likely to be dynamic.

In a related study, also with the Hewlett Packard group, ceramic, single crystal, and thin films of  $\text{Gd}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$  were found to be ferrimagnetic ( $T_c$  between 50 and 80K) with a compensation temperature of 15K [23]. A molecular field model with the Gd sub-lattice antiferromagnetically coupled to the ferromagnetic Mn lattice, explains the results qualitatively. A large high-field susceptibility is found at 5K indicative of sub-lattice rotation. Like  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ , the temperature dependence of the resistivity up to 1100K indicates the transport is by adiabatic small polaron hopping.

## 6) Other studies.

Toward the end of this grant period, we also studied two other problems important to the applications of superconductivity. First we measured the nonlinear microwave response of thin films of the superconducting amorphous Mo-Ge thin films. The point of this work was to clarify through the use of a model system the physical nature of the mechanisms seen in the high temperature superconductors. We demonstrated both a linear J and quadratic  $J^2$  term in the surface resistance and inductance of these films. These current dependencies are also seen in the high temperature superconducting thin films depending on the power level. The interpretation of these results is still underway

We also carried out a preliminary study using numerical simulations of the step response of series arrays of Josephson junctions acting as high voltage output driver amplifier for Josephson digital circuits communicating with semiconducting circuits. This is a nontrivial problem in nonlinear dynamics due to coupling of the Josephson oscillations of the individual junctions in the array. We established some general rules for how this coupling affects the dynamics of the array acting as a switch. Two clearly important principles are that the symmetry of the on (finite voltage) state must be the same as the off (zero voltage) state. Only then can transients associated with changes in the nature of the dynamical states be avoided, and very rapid switching obtained. Similarly unwanted transients can be associated with the presence of significant disorder in either the magnitude of the critical currents of the junctions in the array or the timing of the input pulsed to the individual junctions in the array. This work is being prepared for publication.

Finally, we have just received the new Floating Zone Furnace for single crystal growth of oxides and in particular high-Tc-related systems. The high temperature-high pressure capabilities of the furnace will be used in particular to explore new insulating phases as candidates for substrates of high-Tc films.

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Charles H. S. Ahn, September 1986  
Growth and Properties of Epitaxial Complex Oxides

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Surface Studies on YBCO and Transient Dynamics of Series  
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## Publications

November 1, 1994 – October 31, 1997

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