Critical Parameters of Superconducting Materials and Structures

M.J. Fluss
R.H. Howell
P.A. Sterne
J.W. Dykes
W.D. Mosley
A. Chaiken
K. Ralls
H. Radousky

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Critical Parameters of Superconducting Materials and Structures

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ABSTRACT

We report here the completion of a one year project to investigate the synthesis, electronic structure, defect structure, and physical transport properties of high temperature superconducting oxide materials. During the course of this project we produced some of the finest samples of single crystal detwinned $\text{YBa}_2\text{Cu}_3\text{O}_7$, and stoichiometrically perfect $(\text{Ba},\text{K})\text{BiO}_3$. We deduced the Fermi surface of $\text{YBa}_2\text{Cu}_3\text{O}_7$, $(\text{La},\text{Sr})_2\text{CuO}_4$, and $(\text{Ba},\text{K})\text{BiO}_3$ through the recording of the electron momentum density in these materials as measured by positron annihilation spectroscopy and angle resolved photoemission. We also performed extensive studies on Pr substituted $(\text{Y},\text{Pr})\text{Ba}_2\text{Cu}_3\text{O}_7$ so as to further understand the origin of the electron pairing leading to superconductivity.

INTRODUCTION

As practical applications of high temperature superconducting materials grow nearer there remain significant gaps in our understanding of the superconducting phenomenon. We have investigated a variety of electronic and molecular structure issues as to how they relate to the basic physical phenomena/properties in several oxide superconducting systems. The research was enhanced with collaborators in industry and the UC system.

The electronic structure of superconducting materials has been a topic of fundamental importance in understanding the superconducting mechanism ever since the discovery of the high temperature superconducting phenomena. At LLNL we have performed a series of unique benchmark experiments to determine the details of the Fermi surface in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ inaccessible by other techniques by using angular correlation of the annihilation radiation of electron-positron pairs (ACAR) as well as the more well known synchrotron based angle resolved photo emission spectroscopy (ARPES). The ACAR data for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ first established the conductivity of the copper oxygen chains and remains the most convincing experimental evidence of this feature which confirms predictions of detailed band structure calculations at LLNL and NRL. Not all the high temperature superconductors have this interesting chain structure, but most have a common Cu-O plane from which the dominant high temperature superconductivity is believed to arise. As a result of this reasoning we set out to observe the evolution of the electronic structure of the first high temperature superconductor $(\text{La},\text{Sr})_2\text{CuO}_4$ which has only one Cu-O plane in the unit cell so as to compare to models treating the basic issues of electron correlation in superconducting oxides.

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FERMI SURFACE vs. DOPING

We have measured the shape and location of the Fermi surface in $(\text{La,Sr})_2\text{CuO}_4$ using the positron ACAR for Sr dopings of 0.1, 0.13, and 0.2 and observed the evolution of the Fermi surface as the hole doping in the system is increased. These measurements were analyzed in coordination with detailed calculations of the ACAR measurement in the $(\text{La,Sr})_2\text{CuO}_4$ system using the Linear Muffin Tin Orbital method. Samples were single crystals of $(\text{La,Sr})_2\text{CuO}_4$ grown by the "spinning crucible" method obtained from collaborators at the University of Tokyo. The details of the changes in the Fermi surfaces observed as a function of Sr doping are not reproduced by our theory leading us to the conclusion, after detailed analysis for systematic errors and model limitations, that the Fermi surface is distorted or smeared as doping is increased.

We have explored the possible causes of the Fermi surface smearing in this system. Some such as Sr disorder, or effects due to changes in the positron sampling function (the overlap of the positron wave function with the occupied electron states) have been found to be highly unlikely. The likelihood that the observed Fermi surface smearing effects are due to electron correlations is growing and is the subject of many new research initiatives.

STOICHIOMETRIC DEFECTS

$(\text{Ba,K})\text{BiO}_3$ is a copper-free superconducting oxide with a transition temperature above 31K. Due to the lack of magnetic ions in the structure and its high onset temperature, it is believed hold much promise in technological applications. In collaboration with Dave Mosley, Peter Klavins (both graduate student employees in C&MS), and Robert Shelton at the UC Davis physics department, we have investigated the phase diagrams and the influence of cation defects and oxygen stoichiometry on the magnetic and transport properties of high-quality electrodeposited crystals of the material. By combining positron annihilation measurements and calculations, we have determined that unlike the melt textured material, electrodeposited $(\text{Ba,K})\text{BiO}_3$ is free of cation defects. More importantly, it was also determined that these samples lack full oxygen stoichiometry. When the samples were annealed in an oxygen rich atmosphere, oxygen uptake was observed by an amount which corresponded well to theoretical calculations. Oxygen uptake has been found to lower the normal state resistivity and to improve the metallic dependence of resistance versus temperature measurements. Oxygenating $(\text{Ba,K})\text{BiO}_3$ also effects the superconducting properties by increasing the onset temperature and sharpening the transition.

We have also determined the Fermi surface in well oxygenated, large single crystals of $(\text{Ba,K})\text{BiO}_3$. The shape of the Fermi surface agrees well with calculations predicting surface regions capable of "nesting behavior" which is usually associated with BCS superconductivity.

POISONING of HIGH TEMPERATURE SUPERCONDUCTIVITY

Pr is the only rare earth that fails to give a superconducting system in the 123 oxide structure. Extensive doping and transport studies were performed on ceramic and single crystal samples of the $(\text{Y,Pr})\text{Ba}_2\text{Cu}_3\text{O}_7$ system have resulted in a good

understanding of the relative role of Pr in changing the charge distribution and the spin distribution in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

STRUCTURE-PROPERTY RELATIONSHIPS in HIGH T_C PLANAR JUNCTIONS

We have collaborated with Varian Research Corporation to study the physical properties of transport in unique superconductor-insulator-superconductor junctions. Films of superconducting $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and related compounds are being fabricated by atomic layer by atomic layer using molecular beam epitaxy at Varian Research Corporation. This fabrication technique has resulted in new compounds and phases that would not form in bulk processing techniques. Metastable structures such as superlattices of successive $\text{Bi}(2212)$, and $\text{Bi}(2201)$ unit cells, unit cells containing only one layer of bismuth oxide rather than the usual two, and unit cells containing an arbitrary number of copper-oxygen layers have all been fabricated. Two layers of superconducting $\text{Bi}(2212)$ separated by an insulating layer containing seven copper-oxygen and calcium layers doped with dysprosium have been deposited. After depositing electrode material these three component films are patterned into complete tunneling devices to form Superconductor-Insulator-Superconductor Josephson Junctions.

These devices form good Josephson junctions at low temperature and have sharp Shapiro steps under microwave illumination. These are the only tunnel junctions that depend on transport along the c axis perpendicular to the copper-oxygen plane and they have unique properties when compared with junctions formed by in-plane transport. They are the first high temperature Josephson junctions to show hysteresis in the current-voltage characteristics and they can be fabricated so that the product of critical current and normal state junction resistance is constant while the critical current varies by decades. Details of the temperature dependence of the current flow through the insulating layer have identified a hopping mechanism for the charge transport in some insulating phases. Other SIS combinations lack these characteristics as the properties of the Josephson junctions are correspondingly modified.

This collaboration has continued past the end of the LDRD project through a CRADA with Varian and funding from the Technology Transfer Initiative. This work has resulted in a better scientific understanding of both the superconducting mechanism and the film deposition process.

End

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