DOE/ER/45113--T2

DE91 004236

**Performance Report
Superconducting Materials
September 1, 1990 - February 28, 1991
Grant No. DEFG05-84ER45113**

**J. Ruvalds
Department of Physics
University of Virginia
Charlottesville, VA 22901**

Prepared for the Department of Energy under Grant No. DEFG05-84ER45113

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER
JRS

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Table of Contents

	<u>Page</u>
Paper Published During Grant Period	1
Papers Submitted and Invited Talks During Grant Period	2
Personnel and Students	3
Time Devoted to Project by Principal Investigator	5
Research Projects	6
I. Fermi Liquid Nesting in High Temperature Superconductors	8
II. Optical Properties of High Temperature Superconductors	11
III. Hall Effect in Superconducting $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$	15
IV. Source of High Transition Temperatures	18
V. Prospects for new Superconductors	19
References	21

Papers Published During Grant Period

1. "Hall Effect and Electronic Structure of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ", J. Ruvalds and A. Virosztek, Phys. Rev. B42, No. 1, 399 (1990).
2. "Infrared Reflectivity of High T_C Superconductors", J. Ruvalds and A. Virosztek, Physica B165, 1267 (1990).
3. "Nested Fermi Liquid Theory", A. Virosztek and J. Ruvalds, Phys. Rev. B42, No. 7, 4064 (1990).
4. "Fermi Liquid Reflectivity of TiS_2 ", C. Julien, J. Ruvalds, A. Virosztek, and O. Gorochoy, Sol. St. Comm. (to appear).

papers removed and cited separately -

Papers Submitted During Grant Period

1. "Oxygen Content, Plasmons, and Superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_x$," Y. Ishii and J. Ruvalds, Phys. Rev. (pending).
2. "Optical Properties and Fermi Surface Nesting in Superconducting Oxides," J. Ruvalds and A. Virosztek, Phys. Rev. B (pending).

Lectures and Invited Talks in 1990

IBM Thomas J. Watson Research Center

Gordon Conference on Correlated Systems, Brewster, MA

Université Pierre et Marie Curie, Paris

Institut Laue-Langevin, Grenoble, France

Kernforschungszentrum, Karlsruhe, Germany

Walther-Meissner Institut, Garching, Germany

University of California at Irvine

University of California at Los Angeles

Personnel and Students

Research Associate Jian Zhang joined our group on September 1, 1990 after earning his PhD from Ohio University.

Dr. A. Virosztek completed his postdoctoral appointment in the summer of 1990 and continuous to work with our group on research projects involving high temperature superconductors. He has accepted a permanent job in Budapest, Hungary.

During the last two years our efforts to discover new superconductors involved the enthusiastic participation of students with various backgrounds and interests. In addition to graduate student Tom Sutto, the following undergraduate students contributed to our research: Gregory Ashe, (now grad student at U. Michigan); Hyun B. Shin (now grad student at U. Virginia); Mike Rilee (now grad student at Cornell); and current undergraduates Sarah Woldehana, Clark Allen, Mike Diener, Peter Owen, Cynthia Vinion, and Vera Siregar.

We look forward to involvement of undergraduates as well as graduate students in the future.

Synopsis of Research by Personnel and Students

Dr. A. Virosztek joined our team as a research associate in 1988 after completing a postdoctoral term with Prof. K. Maki at USC. Dr. Virosztek's recent results on the Hall Effect of superconducting oxides are particularly important for understanding many fundamental issues, such as the actual density and mobility of charge carriers in these unusual superconductors. Furthermore, his new results on Fermi Liquid nesting in superconducting oxides have provided a physical explanation for anomalies in the quasiparticle damping. Nesting of the Fermi surface was found to produce a resistivity which increases linearly with temperature in accord with experiment. Furthermore, the corresponding linear frequency variation of the damping may produce unusual spectral features in the dielectric function which may be probed by optical measurements and independently by electron loss spectroscopy.

Dr. Jian Zhang joined our group just two months ago, but already has produced a solution of the Boltzmann transport equations in the case of Fermi surface nesting which refers to nearly parallel sections of the electron trajectory in metals such as the high temperature superconductors. He has applied these results to examine the optical conductivity and reflectivity as a function of frequency up to roughly 1eV. The correspondance of the nested Fermi Liquid analysis to experimental data on $\text{YBa}_2\text{Cu}_3\text{O}_7$ and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ is quite good. These studies reveal the importance of electron-electron scattering and provide information on the charge carrier density in these superconducting oxides.

Recently we have initiated a search for new superconducting materials with the aid of several students and colleagues. Guided by our theoretical basis, we have synthesized various oxide superconductors and established their transition temperatures using a SQUID magnetometer apparatus in the chemistry laboratory of Prof. E. Sinn. Also, we are examining certain sulfides which have a planar electronic structure, with the goal of intercalating specific atoms to achieve novel high temperature superconductors.

The chemistry students involved in our search are undergraduates Clark Allen, Mike Diener, Peter Owen, and graduate student Tom Sutto, whereas physics undergraduates working on the project were Vera Siregar and Cynthia Vinion.

Time Devoted to Project by Principal Investigator

1. 50% of time during the academic year period September 1, 1990 - June 1, 1991.
2. 100% of time June 1, 1991 - August 31, 1991.
3. 50% of time during the forthcoming academic year period Sept. 1, 1991 - Feb. 28, 1992.

Research Projects

Our research on high temperature superconductors has recently produced novel insights for the normal state properties of the copper oxides that have been discovered in the last few years. Advances in materials preparation have produced single crystal samples, and sophisticated surface cleavage techniques have unveiled truly metallic behavior in many respects. Thus, the very recent discovery of a Fermi surface in several cuprate superconductors by photoemission spectroscopy has aroused interest in experimental features which heretofore were in apparent contrast to the expectations for a conventional Fermi Liquid.

Standard Fermi Liquid properties are based on a well defined quasiparticle whose damping is small at the Fermi energy E_F . As shown by Luttinger 30 years ago, electron-electron collisions for particles within an effective mass approximation have a DC damping $\Gamma \sim T^2$ which becomes $\Gamma \sim \omega^2$ for a frequency $\omega = E - E_F$. The conventional damping results from the momentum and energy conservation requirement in three dimensions, and there are modifications in systems of reduced dimensionality. Ordinary metals such as copper are notable for very weak electron-electron contributions to the damping which are barely detectable in most cases.

However, the normal state transport and optical properties of the superconducting cuprates exhibit very unusual variations as a function of temperature and frequency. For example, the resistivity is found to be linear in temperature over a wide range in $\text{YBa}_2\text{Cu}_3\text{O}_7$ and several other oxides. The corresponding optical conductivity exhibits an anomalous contribution at high frequencies. Both features differ from conventional metals, and thus indicate an anomalous strong damping.

Our group has shown that the strong on-site Coulomb coupling U of the copper oxide systems yields a novel quasiparticle damping rate in cases where the Fermi surface has nearly parallel trajectories referred to as "nesting". Such a nested Fermi liquid (NFL) is found to have a DC damping rate $\Gamma_{\text{NFL}} = \alpha T$ where α is a dimensionless measure of U/W , in terms of the bandwidth W . At finite frequency $\omega > T$, the damping becomes $\Gamma_{\text{NFL}} = \alpha \omega$. The latter behavior has recently been observed in photoemission experiments on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ with a magnitude of α in the expected range.

Transport properties like the resistivity and optical conductivity require careful analysis beyond the more direct damping calculation. We have been able to derive the conductivity in the case of a nested Fermi surface and find good agreement with the infrared measurements from several laboratories. Our analysis of the dielectric response predicts a broad peak in the structure factor near the plasma frequency. The peak position and width are in accord with recent Electron Loss data on $\text{Bi}_2\text{Sr}_2\text{CaCuO}_8$. The latter analysis provides key information regarding the charge carrier concentrations in superconducting oxides.

Hall coefficient measurements revealed mysterious variations as a function of chemical composition and temperature which are difficult to interpret in terms of a single group of carriers. We have shown that the mobility of electrons in a nearly half-filled band is very low in the presence of strong electron-electron coupling, and the associated scattering cross section is very sensitive to the alloy composition. By means of a many-body calculation for carriers in two electronic bands appropriate to the cuprates, we have explained the unusual Hall effect in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ including the fact that an increase of Sr content from $x = 0.1$ to 0.3 results in a decrease of the Hall constant by more than two orders of magnitude.

One possible source of high superconducting transition temperatures T_C is the existence of low energy acoustic plasmon modes in addition to the usual majority carrier oscillations. Historically this mechanism has been baffled by the requirements for formation of the acoustic modes as well-defined excitations, and the lack of evidence for their existence. Our calculations for a simple tight-binding model reveal that the two-dimensional electronic nature emanating from the copper oxide structures favors the formation of weakly damped acoustic plasmons in a two-component carrier system. The conditions for the existence of these plasmons are sensitive to the position of the Fermi energy determined by the oxygen content for example. A qualitative correspondence of the plasmon frequency and T_C is found for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The first direct evidence for an electronic excitation near 45 meV has just been discovered at IBM with electron loss spectroscopy.

Finally we mention our experimental efforts to synthesize new superconducting alloys.

I. Fermi Liquid Nesting in High Temperature Superconductors

A fundamental issue for high temperature superconductors is the apparent inadequacy of a conventional^{1,2} Fermi liquid description for the transport data, such as the resistivity, optical properties, and other features. Yet photoemission measurements³ on freshly cleaved single crystals provide convincing evidence for the existence of a Fermi surface and electronic bands with considerable dispersion.

We have discovered⁴ a possible resolution of the Fermi Liquid dilemma as a consequence of "nesting" which refers to Fermi surfaces with sections that are nearly parallel in momentum space. Our calculations rely on a conventional Hamiltonian

$$H = \sum_{\vec{k}\sigma} E(\vec{k}) c_{\vec{k}\sigma}^\dagger c_{\vec{k}\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \quad (1)$$

where $U > 0$ denotes the on-site Coulomb repulsion, $c_{\vec{k}\sigma}^\dagger$ ($c_{\vec{k}\sigma}$) are the creation (destruction) operators for an itinerant electron or hole within a band $E(\vec{k})$ of width W .

Electron scattering is limited by the Pauli exclusion principle to a narrow region of space for a fixed temperature T and distance from the Fermi energy E_F . Hence the topology of the electron orbit is a key determinant in the damping rate Γ caused by the electron-electron coupling.

A conventional Fermi liquid refers to an energy dispersion $E(\vec{k}) = k^2/2m^*$ with an effective mass m^* . In three dimensions, the standard damping² is

$$\Gamma_{FL} = \frac{\pi \bar{g}^2}{2W} [\pi^2 T^2 + \omega^2], \quad (2)$$

where T and W are in energy units and the dimensionless coupling is $\bar{g} = U/W$. This Born approximation result is justified because the damping is quite small in ordinary metals because the bandwidth is much larger than the typical temperatures of interest. Physically the variation $\Gamma_{FL} \sim T^2$ follows from the Pauli exclusion requirement that limits the states available for the incoming electron

to a shell of thickness T , and another factor of T comes from the restriction on the target electron. It is reasonable, though not certain, that the transport properties will be influenced in a similar manner, which would yield a resistivity $\rho_{FL} \propto T^2$ providing that a mechanism is available to transfer momentum to the lattice.

Nesting of the Fermi surface refers to a quasiparticle orbit with nearly parallel sections separated by a nesting wave vector Q . An example of a nested orbit is shown in Fig. 1(b); this example reverts to perfect nesting for a square in the case of a half-filled tight-binding band in two dimensions. This situation is especially germane to the Cu $d_{x^2-y^2}$ orbitals in the Cu-O planes structures found in high temperature superconductors.

Fig. 1. Phase-space restrictions for the electron-electron cross section at finite temperature T in (a) conventional electron gas, and (b) a Fermi liquid with nesting wave vector Q . The incoming quasiparticle momenta are k and k' , with a scattering momentum transfer q . Shaded regions are allowed by the Pauli exclusion principle. The nesting broadens the range of momenta available to the scattering electron to a width proportional to W in comparison to the free particle width proportional to T . In the NFL case $q=Q$. The solid curves represent the Fermi surface.

Our calculations invoke the approximate nesting condition $E(\vec{k} + \vec{Q}) + E(\vec{k}) \approx 0$,

which yields an unusual damping rate caused by the dominance of scattering processes with momentum $\vec{Q}$. The details of the analysis have just been published⁴, and yield the following features for a nested Fermi Liquid (NFL). The solution for the quasiparticle damping is:

$$\Gamma_{\text{NFL}}(\omega \leq T) \approx \alpha T, \quad (3)$$

and $\Gamma_{\text{NFL}}(\omega \geq T) \approx \alpha \omega, \quad (4)$

where the coupling α is fixed by the self-consistency requirement

$$\alpha = \bar{g}^2 \tan^{-1}\left(\frac{1}{\alpha}\right), \quad (5)$$

and $\bar{g} = U/W$ is the dimensionless electron-electron coupling.

The magnitude of the coupling is constrained to $\alpha < 1$ because otherwise a transition to a spin density wave state is expected.

Direct experimental evidence for the frequency variation $\Gamma_{\text{NFL}} = \alpha \omega$ has been discovered by photoemission spectroscopy on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ superconductors with $\alpha \approx 0.6$. This value turns out to be compatible with independent optical reflectivity experiments which we discuss next.

II. OPTICAL PROPERTIES OF HIGH TEMPERATURE SUPERCONDUCTORS

Infrared reflectivity $R(\omega)$ measurements on single crystal specimens reveal a frequency variation in cuprates that is quite different from the classical Drude behavior found in ordinary metals like silver. A typical example of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ is shown in Fig. 2 with two sets of experimental points.^{5,6}

Fig. 2. Optical reflectivity data from the Florida (Ref. 6) and Stanford (Ref. 5) groups in comparison to the Drude behavior of the dashed curve. The solid curve shows the result of our NFL analysis.

We have calculated the dielectric function in the presence of Fermi surface nesting which takes the form

$$\epsilon_{\text{NFL}}(\omega) = \epsilon_{\infty} - \frac{\omega_p^2}{\omega \left[\omega \frac{m_{\text{NFL}}}{m_0} + \frac{i}{\tau_{\text{NFL}}} \right]} \quad (6)$$

where consideration of self energy and vertex corrections yield a transport relaxation time τ_{NFL} given

by
$$\frac{1}{\tau_{\text{NFL}}} (\omega < 2T) \approx 3.3\alpha T \quad (7)$$

and
$$\frac{1}{\tau_{\text{NFL}}} (\omega > 2T) = \alpha \omega \quad (8)$$

which are somewhat different from the Born Approximation result for the quasiparticle damping in Eqs. 3 & 4. To satisfy causality, the effective mass becomes frequency and temperature dependent:

$$m_{\text{NFL}}^* = m_0 \left[1 + \frac{2\alpha}{\pi} \ln \frac{\omega_c}{\text{Max}(2T, \omega)} \right], \quad (9)$$

where ω_c is a cut-off frequency constrained to be $\omega_c \leq W/(1+\alpha)$.

Drude relaxation is described by a constant mass and relaxation time. However, the Drude function is not in accord with the reflectivity as seen in Fig. 2. Further discrepancies are found for the optical conductivity where the Drude form is normally used to fit the low frequency peak.

Although the NFL analysis yields a good description of the reflectivity with $\alpha = 0.6$, $\epsilon_\infty = 3$, $\omega_{\text{pl}} = 1.1$ eV and $\omega_c = 0.9$ eV, it is desirable to test the analysis in relation to the optical conductivity $\sigma(\omega)$. The data for $\sigma(\omega)$ is shown in Fig. 3, along with the Drude behavior (dashed curve) and the NFL solid curve. The good description of the higher frequency region provides additional evidence for damping that is linear in ω .

An additional test of the theory is the low frequency region of $\sigma(\omega)$ which is capable of checking the predicted temperature variation of the damping $1/\tau$. Extrapolating the conductivity to the DC limit $\omega=0$ yields a resistivity that is linear in temperature in agreement with the observations.

Figure 3. Optical conductivity data on $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_2\text{O}_8$ in comparison to standard Drude behavior and our NFL calculations.

A principal result of our analysis is information regarding the charge carrier density n which appears in the plasma frequency $\omega_p^2 = 4\pi ne^2/m_0$. To pin down this quantity more precisely we have obtained the structure factor

$$S_{\text{NFL}}(q=0, \omega) = \text{Im} \left[\frac{-1}{\epsilon_{\text{NFL}}(\omega)} \right]. \quad (10)$$

The computed structure shows in Figure 4 the Drude prediction as a narrow peak at low frequency determined by the fit to the $\sigma(\omega)$ data. By contrast the nested case yields a much broader peak at a higher frequency, and it is gratifying that the electron loss measurements by the Karlsruhe group⁸ reveal the broad features predicted by the NFL analysis using the same α and ω_p from the optical studies.

Figure 4. Electron loss spectroscopy data in comparison to the structure factor in the Drude model and the solid curve of our NFL analysis.

On the basis of the optical and electron loss spectra, the plasma frequency is evidently close to $\omega_p^2/\epsilon_\infty = 1.1\text{eV}$. Given a value of $\epsilon_\infty = 3$ in accord with published accounts of the high frequency dielectric constant, we obtain a carrier density $n \sim 2 \times 10^{22} \text{ cm}^{-3}$. This concentration appears to be at odds with measurements of the Hall coefficient R_H of representative cuprate superconductors. However, the observed temperature variation of $R_H(T)$ makes a conclusive single carrier analysis doubtful. Thus we have examined the Hall coefficient by considering the carrier mobility influenced by nesting, and we report these results next.

III. HALL EFFECT IN SUPERCONDUCTING $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

A fundamental critique of band structure calculations of superconducting cuprates is the problem of estimating the charge carrier density. Recent support for the validity of the computed electronic structure⁹ in the metallic phases is provided by photoemission experiments¹⁰ which yield a Fermi surface encompassing a nearly half-filled tight binding band. This situation appears to be in conflict with Hall coefficient R_H data on some oxides (such as $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$)¹¹ that appear to indicate a much smaller number of hole carriers.

The complexity of the issue is further aggravated by significant temperature variations of in virtually all of the metallic cuprates. The case of $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ is especially intriguing since $R_H(T)$ is found¹² to change sign as a function of temperature. These results naturally suggest the presence of two types of charge carriers with contributions to R_H that are determined by the relative mobilities of electron and holes for example.

To investigate the Hall conductivity we chose a simple electronic structure that incorporates the qualitative features of the band structure results⁹, but then we add a consideration of the electron mobility in the presence of strong Coulomb scattering. The band model is shown in Fig. 5.

Fig. 5. Electronic structure of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with the Fermi energy E_F relative to the tight-binding bands E_I (originating from the $\text{Cu}d_{x^2-y^2}$ orbital) and the narrow E_h band which may correspond to the $\text{Cu}d_{3z^2-r^2}$ orbital. Both Cu levels are hybridized with oxygen p states.

We found ¹³ that multiple scattering terms beyond the Born approximation need to be included and yield a damping rate for the E_g electrons

$$\Gamma_g = \frac{\pi^3 U^2 N_c^2(E_F) T}{8[1 - U\chi'(Q,0)]^2}, \quad (11)$$

where the real part of the susceptibility is

$$\chi'(Q,0) = \frac{1}{(2\pi)^2 t_g} \ln^2 \left(\frac{16t_g}{E_F} \right), \quad (12)$$

where $8t_g$ is the bandwidth of the E_g band.

This example of a nearly half-filled two-dimensional band is a special case of Fermi surface "nesting". Earlier it was noted that the electron-electron scattering in such a band would yield a metal-insulator transition ¹⁴ to a spin density wave state whenever $1 = U\chi'(Q)$, and also provides an explanation ¹⁵ of the linear temperature variation of the resistivity of the cuprates. Our interest was focused on the influence of alloy composition on the electron mobility that is associated with a straightforward shift of the Fermi energy E_F . As Sr is substituted for the higher valence La atoms, E_F is lowered and more holes are created in the E_h band of Fig. 5 which is held rigid. However, the electrons in the E_g band exhibit a severe change in mobility as shown in Figure 6.

The reduced electron mobility at low Sr content allows the minority holes to dominate the Hall coefficient as shown in Figure 7. Since the electrons and holes mobilities presumable exhibit different temperature dependences, R_H is expected to vary with T , even though a quantitative analysis of the hole mobility is not feasible at this stage. Nevertheless, the total calculated R_H shown by the solid curve in Fig. 7 explains the anomalous drop in R_H by orders of magnitude with relatively small changes in Sr content.

In conclusion, the influence of electron-electron scattering on the quasiparticle mobility appears to reconcile the general features of a well-defined Fermi surface of a nearly half-filled band with anomalies in the Hall data.

Figure 6. Ratio of lifetimes τ_l/τ_h (which is proportional to the electron mobility) is shown as a function of Sr doping x . Within the Born approximation of the dashed curve, the electron-electron scattering yields an enhanced electron lifetime τ_l with the addition of Sr because the Fermi energy is shifted to a region of lower density of states. Multiple scattering strongly suppresses the electron mobility at low x as shown by the solid curve for intermediate Coulomb coupling $U=0.7$ eV. The corresponding metal-insulator transition to an SDW state is expected at $x \approx 0.06$ where the electron mobility vanishes.

Fig. 7. Hall coefficient R_H is shown as a function of Sr content. Data for T=77 and 300 K is from single-crystal thin films (Suzuki, Ref. 4). The dashed line shows the $1/X$ behavior expected for a single band in the effective-mass approximation. The solid curve follows from our two-band analysis with $U \approx 0.7$ eV.

IV. SOURCE OF HIGH TRANSITION TEMPERATURES

One possible mechanism of high temperature superconductivity that has been debated for decades is the possible pairing of electrons by the exchange of electronic modes such as acoustic plasmons. The advantages of a two-dimensional structure of the type found in the copper oxides have been proposed ¹⁶, but the key issue regarding the existence of acoustic plasmons as well-defined excitations remains controversial.

We have computed ¹⁷ the dielectric response for a simple 2-dimensional electronic model similar to the one shown in Fig. 5 and found two sets of plasmon oscillations. The ordinary oscillations of the majority E_g band carriers yield an optical plasmon branch near 1 eV in accord with our analysis of infrared reflectivity and electron loss data for $\text{YBa}_2\text{Cu}_3\text{O}_7$. An acoustic branch of the E_g band is also generated by the 2-dimensional constraint on the dynamics, but the single band plasmons are not considered capable of generating superconducting pairs.

On the other hand, the low energy acoustic branch extending from $\omega_h \sim 0$ at long wavelengths to roughly 0.1 eV at the zone boundary is found to exist as a well defined excitation with small damping.¹⁷ This latter mode may be a candidate for a pairing force if it indeed can be proven to exist.

High resolution electron loss measurements¹⁸ on $\text{YBa}_2\text{Cu}_3\text{O}_7$ at IBM provide the first direct evidence for the lower energy plasmon near 0.045 eV. Also neutron scattering data¹⁹ on single crystal $\text{YBa}_2\text{Cu}_3\text{O}_7$ as well as $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ show an extra peak near 0.05 eV in addition to the phonon modes. Hybridization²⁰ of phonons with acoustic plasmons may account for the extra neutron scattering peak structure.

V. PROSPECTS FOR NEW SUPERCONDUCTORS

Encouraged by our theoretical results, we have synthesized a variety of oxide and sulfide alloys with promising electronic structures for high temperature superconductivity. The remarkable enthusiasm and dedicated work of ten undergraduate students that joined our group during the last two years have provided a superb educational experience in an ambiance of pure discovery. Our student team included three women, including two minority students.

Laboratory facilities for our synthesis and sample characterization studies were provided by Dr. E. Sinn in our Chemistry department and by Prof. B. Farmer in our Materials Science department.

Most of the sample preparation was by vapor transport methods at high temperatures for the sulfide. Oxide preparation followed standard techniques reported in recent literature. Single phase properties were determined by x-ray diffraction and the onset of superconductivity was measured using a SQUID magnetometer which also yields the fraction of the sample that is superconducting from the measured Meissner effect.

A high point of our students' experience was the discovery by chemistry graduate student Tom Sutto of a new superconducting alloy with composition $\text{Ba}_4\text{Tl}_{2-8}\text{Bi}_x\text{Pb}_2\text{O}_{12-8}$. Although the maximum observed superconducting temperature was only $T_C = 10.7$ for $x = 1.0$, the concentration dependence of T_C is unusual. Measurements of the Pauli susceptibility of these alloys shows that T_C did not increase in correspondence to the electronic density of states. Powder x-ray studies reveal an orthorhombic distortion in superconducting samples, whereas a tetragonal structure with alternating MO_2 layers ($M = \text{Tl, Bi, Pb}$) interspersed by Ba is evident for $x < 0.7$ and $x > 1.2$. Confirmation of the superconductivity was provided by resistance measurements with the aid of J. Poon's group in the Physics Department.

This interdisciplinary effort to synthesize novel alloys has been particularly interesting and valuable for student morale and experience. Previous experiments on a wide variety of sulfides were able to determine superconductivity at relatively low temperatures, and variations in composition of such alloys found by our group verified changes in the transition temperature that may be expected for systems with a single band that comply with the BCS theory. However, our ultimate quest is to

discover an appropriate chemical substitution in a layered sulfide compound that generates the partial filling of two electronic states. Such a situation would provide a test of our theoretical basis for high T_C superconductivity and may result in new superconducting materials that are ductile and easier to use in commercial applications.

References

1. J.M. Luttinger, Phys. Rev. 121, 942 (1961).
2. "The Theory of Quantum Liquids," D. Pines and P. Nozieres, (Benjamin, NY, 1966).
3. C.G. Olson, R. Liu, D.W. Lynch, R.S. List, A.J. Arko, B.W. Veal, Y.C. Chang, P.Z. Jiang and A.P. Paulikas, Phys. Rev. B (pending).
4. A. Virosztek and J. Ruvalds, Phys. Rev. B42, 4064 (1990).
5. J.H. Kim, I. Bozovic, D.B. Mitzi, A. Kapitulnik, and J.S. Harris, Jr., Phys. Rev. B41, 7251 (1990).
6. K. Kamarás, S.L. Herr, J.S. Kim, C.D. Porter, G.R. Stewart, D.B. Tanner and T.T. Imusk (preprint).
7. J. Ruvalds and A. Virosztek, Physica B165 & 166, 1267 (1990).
8. N. Nücker *et al*, Phys. Rev. B39, 12379 (1989).
9. J. Yu, A.J. Freeman, and J.H. Xu, Phys. Rev. Lett. 58, 1035 (1987).
10. J.C. Campuzano *et al*, Phys. Rev. Lett. 64, No. 19, 2308 (1990).
11. M. Suzuki, Phys. Rev. B39, 2312 (1989).
12. J.M. Tarascon *et al*, Phys. Rev. B40, 4494 (1989).
13. J. Ruvalds and A. Virosztek, Phys. Rev. B42, 399 (1990).
14. J.E. Hirsch and D.J. Scalapino, Phys. Rev. Lett. 56, 2732 (1986).
15. P.A. Lee and N. Read, Phys. Rev. Lett. 58, 2691 (1987).
16. J. Ruvalds, Phys. Rev. B35, 8869 (1987).
17. Y. Ishii and J. Ruvalds, Phys. Rev. B (pending).
18. J.E. Demuth, B.N.J. Persson, F. Holtzberg and C.V.Chandrasekhar, Phys. Rev. Lett. 64, 603 (1990).
19. L. Pintschovius (preprint).
20. A historical review of acoustic plasmon research is in: J. Ruvalds, *Advances in Physics* 30, 677 (1981).

END

DATE FILMED

12 / 11 / 90