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Performance Report
Superconducting Materials
May 1,1992 - February 28, 1993
Grant No. DEFG05-84er45113

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Reprints removed

Papers Published During Grant Period

1. " Hall Effect and Electronic Structure of $La_{2-x}Sr_xCuO_4$ ", J.Ruvalds and A. Virosztek, Phys . Rev. B42, 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, 4064 (1990)

4. " Optical Properties and Fermi Surface Nesting in Superconducting Oxides "' J. Ruvalds and A. Virosztek, Phys. Rev. B43, 5498 (1991)

5. " Superconductivity in $Ba_4BiPb_2TlO_{12}$ ", T.E.Sutto, B.A.Averill and J. Ruvalds, Chemistry of Materials 3, 209 (1991)

6. " Raman Scattering by a Nested Fermi Surface ", A. Virosztek and J. Ruvalds, Phys. Rev. B. 67, 1657 (1991)

7 " Raman Spectrum of Superconducting Oxides ", A. Virosztek and J. Ruvalds, Phys. Rev.B45, 347 (1992)

8. " Fermi Surface Nesting in High Temperature Superconductors ", J. Ruvalds, J. Zhang, and A. Virosztek, J. Phys. Chem. Solids 52, 1371 (1991)

9. " Spin Susceptibility Scaling in High-Temperature Superconductors ", J. Ruvalds, J.Zhang, and A. Virosztek, Science 256, 1664 (1992)

Invited Talks

1990

IBM Thomas J. Watson Research Center

Gordon Conference on Correlated Systems, Brewster, MA

Universite Pierre et Marie Curie, Paris

Institut Laue-Langevin, Grenoble, France

Kernforschungszentrum, Karlsruhe, Germany

Walther-Meissner Institut, Garching, Germany

University of California, Irvine

University of California, Los Angeles

1991

Workshop on Fermiology of High T_c Superconductors, Argonne, IL

University of Toronto

Los Alamos National Lab

Southeastern Section Meeting of the American Physical Society, Durham, NC

Stanford University

1992

University of Illinois Spin Workshop, Urbana, IL

Los Alamos National Lab

Gordon Conference on Superconductivity, Oxnard, CA (Jan., 1993)

Graduate Students and Personnel

Jeffrey Thoma joined our group to perform his PhD thesis research in 1991. His undergraduate honor thesis at the University of Delaware provided a fine background in computational techniques which have been useful for analysis of the heat capacity on high temperature superconductors. Jeffrey's research emphasizes the influence of the cuprate composition on anomalous temperature variations of the heat capacity as well as the magnetic susceptibility.

Dr. Carsten T. Rieck became a Research Associate in our group on September 1, 1992 after earning a PhD degree at Hamburg University in Germany. His exceptionally strong background in theoretical physics enabled him to master formal techniques as well as computer analysis which resulted in our analysis of neutron spectra of high temperature superconductors.

Dr. J. Zhang completed a one year term as a Research Associate and then accepted a position with an environment studies company.

Dr. A. Virosztek completed his postdoctoral appointment in our group in the summer of 1990 and continues to interact with our group from his home base at the Central Research Institute in Budapest. His research at Virginia resulted in an original explanation for several anomalous features of high temperature superconductors, and the predictions of our theory are being tested by various experimental groups.

Our efforts to synthesize promising candidates for superconductivity involved the enthusiastic participation of several undergraduate students as well as chemistry graduate student Tom Sutto. The undergraduates who went on to graduate schools are: Gregory Ashe (Michigan), Hyun B. Shin (Virginia), Mike Rilee (Cornell). Other participating students were; Sarah Woldehana, Mike Diener, Clark Allen, Peter Owen, Cynthia Vinion, and Vera Siregar.

Time Devoted to Project by Principal Investigator

1. 50% of time during academic year period May 1,1992- May 31, 1992

2. 100% of time June 1, 1992 - February 28, 1993

The 100% effort during the 1992-93 academic year was made possible by a Sabbatical visit at Stanford University under the auspices of the University of Virginia Sesquicentennial Associate program.

Synopsis of Research

We have recently discovered an unusual frequency and temperature variation of the spin susceptibility for electrons or holes whose Fermi surfaces exhibit " nesting " in the sense of nearly parallel orbit trajectories. Our original analysis invoked this type of response to explain the unconventional electronic transport properties of high temperature superconductors, including the optical reflectivity and Raman spectroscopy data.

Direct evidence for the predicted scaling of the susceptibility as a function of frequency divided by the temperature has now been detected by neutron scattering experiments on two series of cuprate superconductors at several laboratories, with the MIT - Brookhaven collaboration providing precise details of the momentum variation of the scattering. Thus we have extended our calculations to electronic structure models that are capable of generating specific lineshape details that may be tested experimentally. Good agreement with the temperature and frequency variation of the susceptibilty measured by neutrons is achieved in the regime where the nesting analysis is appropriate.

These findings are relevant to the anomalous linear temperature variation of the electrical resistivity which is a puzzling characteristic of the high temperature superconductors. Electron-electron scattering processes which are nearly negligible in standard metals become a dominant influence on nested surfaces providing that the Coulomb interaction between charges is of intermediate strength.

Thus our nesting analysis provides insight into the fundamental nature of the charge and spin dynamics of the unconventional superconductors.

I. Scaling of the Spin Susceptibility in High Temperature Superconductors

Our analysis is based on the concept of a nested Fermi surface which is illustrated in Figure 1. If sections of the electron trajectory are nearly parallel, then the spin susceptibility may exhibit a scaling in frequency divided by the temperature in sharp contrast to conventional metals where the susceptibility is nearly temperature independent.

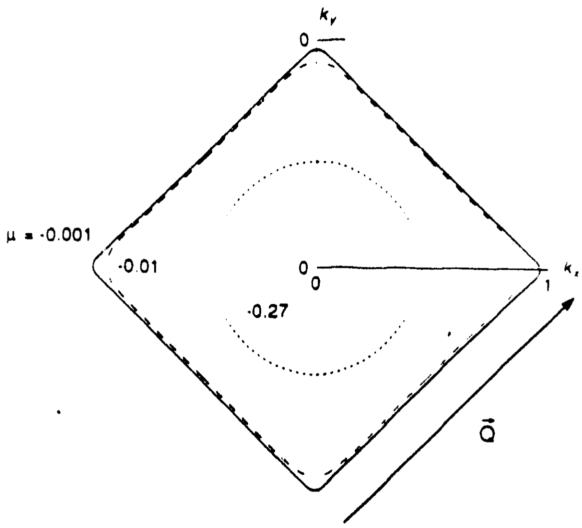

Fig.1. Example of a Fermi surface of a tight-binding energy band in two dimensions which exhibits nesting when the Fermi energy is close to half-filling(e.g. - 0.008t), where 8t is the bandwidth). The anomalous scaling response is found to be dominant near a nesting momentum vector Q.

An analytic derivation of the scaling phenomena for a nested Fermi Liquid (NFL) [1,2] follows from the definition of the susceptibility

$$\chi_0''(\vec{q},\omega) = -\frac{1}{4\pi}\int d^2k\{f[E(\vec{k}+\vec{q})]-f[E(\vec{k})]\}\,\delta\,[\omega - E(\vec{k}+\vec{q}) + E(\vec{k})], \tag{1}$$

where f(x) is the Fermi function. Using the nesting approximation for the electron energy $E(k+Q) = E(k)$, and defining a variable $x = E(k)$ allows a straightforward evaluation of the integral in Eq.1 to obtain

$$\chi_{NFL,0}''(\vec{Q},\omega) = \frac{\pi N(O)}{2}\tanh\left(\frac{\omega}{4T}\right). \tag{2}$$

This NFL result is quite different from the conventional Fermi Liquid behavior [3] of a nearly temperature independent response that characterizes ordinary metals.

Recent neutron scattering evidence [4,5] for the scaling in cuprate superconductors motivated us to proceed further and compute the response for a more realistic tight- binding energy band model with the energy dispersion

$$E(\vec{k}) = -2t\left[\cos(ak_x) + \cos(ak_y)\right] - E_F. \qquad (3)$$

This band exhibits perfect nesting in the form of a square orbit for Fermi energy $E_F = 0$ at the nesting vector Q shown in Fig.1. That special case of a half-filled band yields a singular density of states and is unstable toward the formation of a spin density wave for arbitrarily weak Coulomb coupling. As the Fermi energy is lowered, the singularity is removed even though the nesting may persist, and these features are supported by sophisticated band structure calculations for the cuprate superconductors [6,7], as well as by photoemission experiments [8,9].

The remarkable scaling behavior of the spin response calculated by our group is compared to neutron scattering data in Fig.2.

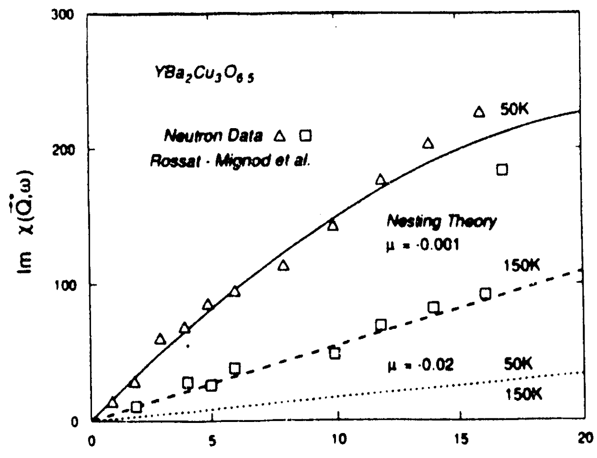

Fig.2. The unconventional scaling of the spin susceptibility calculated for the band model is seen as a strong temperature dependence for a small Fermi energy (-0.008t) that exhibits nesting. The neutron scattering data points from Ref. 4 show the scaling in $YBa_2Cu_3O_{6.5}$. Ordinary Fermi Liquid behavior is shown for a lower Fermi energy (-0.16t) which removes nesting.

A further test of our theory is the momentum variation of the susceptibility which is shown in Fig.3 for a Fermi energy that supports nesting. Although neutron scattering cross sections need to be analyzed for strong background scattering, the lineshape for one large single crystal cuprate measured by the MIT group is in reasonable agreement with the calculated spectrum when self energy corrections are included (details are available in Ref. 2).

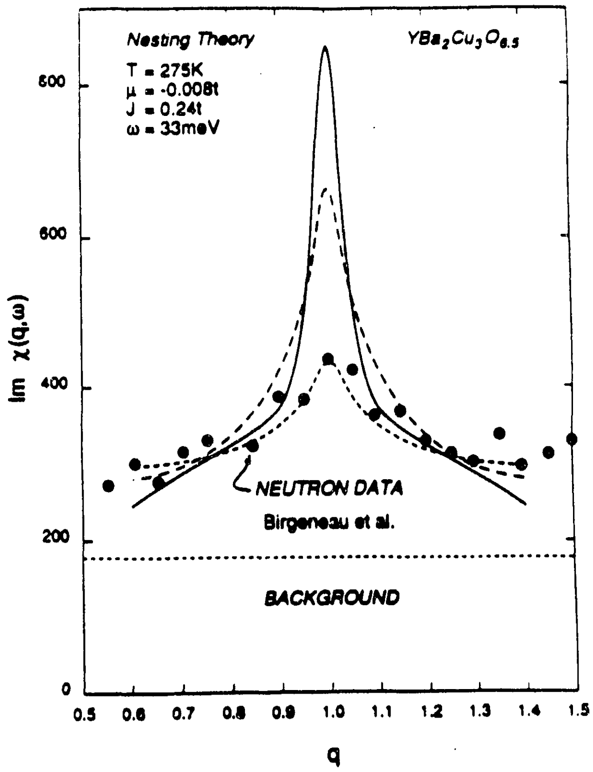

Fig.3.The neutron data of Ref. 5 reveals a lineshape as a function of momentum q for $YBa_2Cu_3O_{6.5}$ that resembles the calculated susceptibility using a Fermi energy of -0.008t . The dashed curve shows the lowest order response, while the solio curve demonstrates the influence of RPA corrections. The dotted curve includes self energy correction which are found to be significant at the frequency used in this measurement.

These scaling features produce a remarkable influence on the electron-electron scattering cross section which determines electrical transport properties. In particular, the induced frequency variation of the damping provides an explanation for the anomalous infrared reflectivity and Raman spectra as discussed in the next sections.

II. OPTICAL REFLECTIVITY AND ELECTRON LOSS SPECTRA

Infrared reflectivity data on cuprate single crystals reveal a frequency variation that is quite different from conventional Drude behavior found in ordinary metals like copper.The comparison of data in Figure 4 exhibits the high and smooth reflectance of Cu in contrast to the anomalous decreasing shapes for $Bi_2Sr_2CaCu_2O_8$ superconductor data (10) and untwinned $YBa_2Cu_3O_7$ spectra (11). Also shown is the unusual response of metallic chromium as a function of frequency.

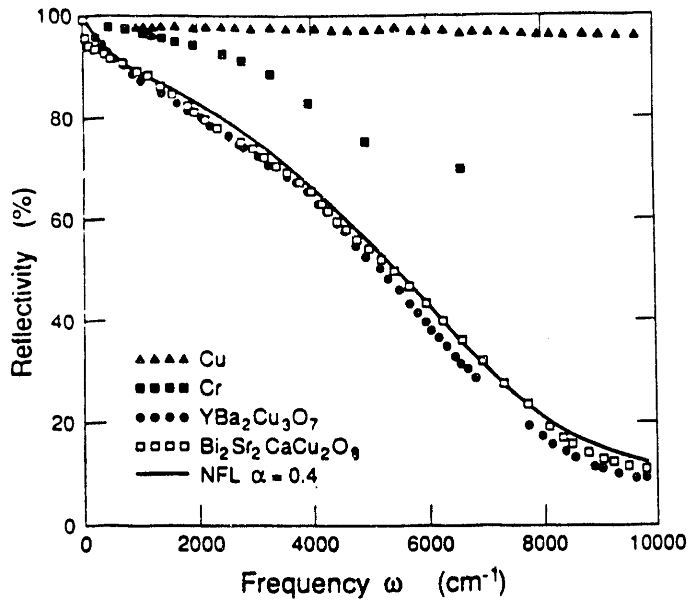

Figure 4.Comparison of optical reflectivity data up to 1.2 eV for two high temperature superconductors from references 10 & 11 with the conventional Drude behavior of copper.The solid curve is a fit of our theory based on Fermi surface nesting with an intermediate electron-electron coupling strength as discussed in Ref.13 and summarized below.

The origin of the frequency dependence of the damping can be traced to the scaling properties of the electron susceptibility which enter in the definition of the electron self energy

$$\Sigma'' \approx 2\alpha^2 N(E_F) \int d\omega' \frac{\chi''(q,\omega')}{\sinh\left(\frac{\omega'}{T}\right)} \qquad (4)$$

The conventional Fermi Liquid response is a linear in frequency and insensitive to temperature because T is presumably much smaller that the Fermi energy. In that case, a simple variable change to y= ω' / T readily reveals that the self energy is proportional to T .

The exceptional behavior of the nested NFL response follows from the observation that scaling of the susceptibility allows a similar variable change that yields a linear T variation of the self energy, and this feature is preserved when higher order corrections are included in the theory (1). Similarly, a linear frequency variation of the damping follows from the NFL analysis, and this variation provides a key to understanding the optical properties. It should be noted that the transport relaxation rate is not simply related to the self energy for anisotropic Fermi surfaces, but the ideal nesting situation reveals a mechanism for scattering that modifies the current-current correlation function to give a modified dielectric function

$$\epsilon_{NFL}(\omega) = \epsilon_{\infty} - \frac{\omega_p^2}{\omega\left[\omega\frac{m^*_{NFL}}{m_0} + \frac{i}{\tau_{NFL}}\right]} \quad (5)$$

where consideration of vertex corrections is a vital ingredient (13). The relaxation time is

$$\frac{1}{\tau_{NFL}}(\omega<2T) = 3.3\alpha T \quad (6)$$

$$\frac{1}{\tau_{NFL}}(\omega>2T) = \alpha\omega \quad (7)$$

where α is the Coulomb repulsion divided by the bandwidth. Causality requires that

$$m^*_{NFL} = m_0\left[1+\frac{2\alpha}{\pi}\ln\frac{\omega_c}{\mathrm{Max}(2T,|\omega|)}\right], \quad (8)$$

describes the frequency dependence of the effective mass up to a cutoff frequency.

An important result of our analysis is the prospect of measuring the charge density n which appears in the plasma frequency that may be inferred from the reflectivity . Thus we proceeded to check the predicted plasma frequency peak in the structure factor

(9)

The comparison of the computed structure factor to independent optical and electron loss (14) spectroscopy probes is shown in Figure 5. Unlike the standard Drude model which would predict a narrow peak whose width is proportional to temperature, our NFL results yield a broad plasmon peak whose width is almost temperature independent and a value of the "bare" plasmon frequency near 3 eV that is in accord with band structure calculations (7).

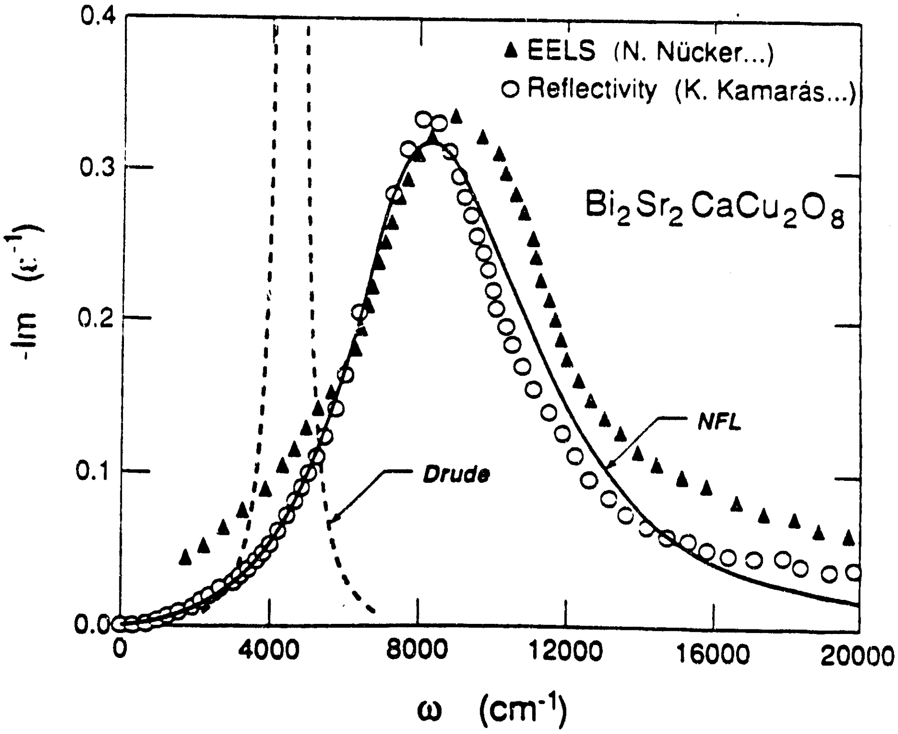

Figure 5. The solid curve shows our computed structure factor from Ref.13 in comparison to optical data of Ref.10 and independent electron loss spectra (EELS) of Ref.14. The broad peak represents a plasmon whose bare energy of 3 eV is shifted to a considerably lower value by the background dielectric function which is measured independently. The width of the peak indicates that electron-electron collisions of intermediate strength yield the unusual frequency variation of the damping.

Our theory satisfies the conductivity and structure factor sum rules that are required by charge conservation.

III. ELECTRONIC RAMAN FEATURES IN SUPERCONDUCTORS

The discovery of broad electronic spectra in high temperature superconductors by various experimental groups (15,16,17) generated great interest in part because normal metals are expected to exhibit only weak light scattering. A recent example is shown in Figure 6, along with the solid curve that is a result of our analysis (18)

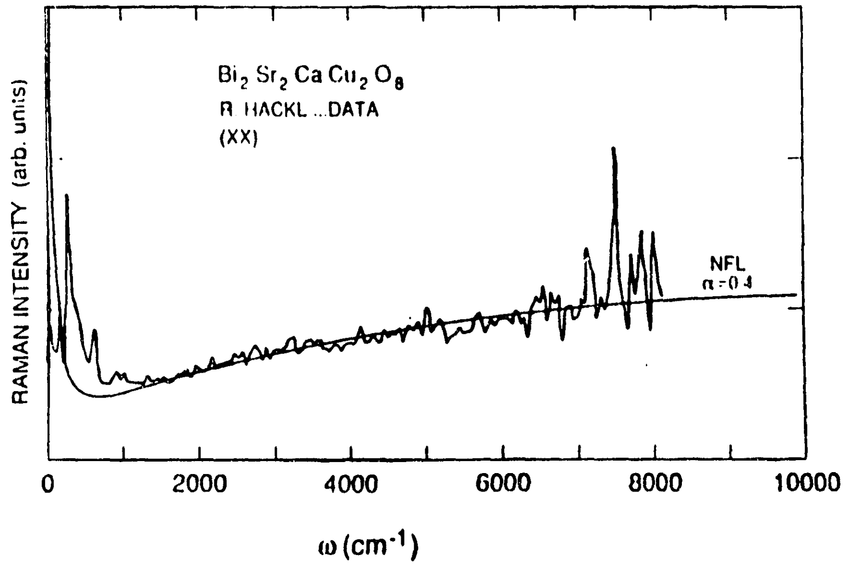

Figure 6. Raman spectrum of a high temperature superconductor from Ref.17 is shown in comparison to our calculated NFL lineshape from Ref.18 using the same values of the plasma frequency and electron-electron coupling that were obtained from our fit to the infrared reflectivity data of the metal.

The key point in our theory of the Raman spectrum is the NFL concept applied to an anisotropic electronic energy band that generates light coupling to energy density fluctuations. Earlier theories of the energy density mechanism include studies of doped semiconductors (19) and other metals (20,21). A formal feature of interest is a relationship between the optical conductivity and the Raman lineshape that was derived on general grounds (22), and also appears in our specific calculations for a nested Fermi surface.

Experiments are underway at the University of Illinois and other laboratories to test detailed predictions of our theoretical results. The details of our calculations have just been published in Reference 18.

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