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JAN 08 1990

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LA-UR--89-4190

DE90 004857

TITLE Normal State Electronic Structure and the Superconducting Energy Gap in HTSC's as Determined from Photoemission Spectroscopy

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SUBMITTED TO 2nd International Symposium on Superconductivity, Tsukuba, Ibaraki, Japan, November 14-17, 1989

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SUMMARY

Normal State Electronic Structure and the Superconducting Energy Gap in HTSC's as Determined From Photoemission Spectroscopy

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Photoemission spectroscopy has been utilized to determine the electronic structure of high- T_c materials. The observation of dispersive bands at E_F suggests a Fermi surface similar to that obtained from a band calculation. The results apparently are not inconsistent with the notion of a correlated Fermi liquid consisting of hybridized p-d bands.

However, it is becoming more and more difficult to distinguish between Fermi liquid behavior in the new high- T_c superconductors and behavior expected on the basis of the novel new non-Fermi liquid theories. The differences are now predicted to be on an energy scale smaller than our experimental resolution. We point out that, while deviations from simple band theory certainly do exist in the form of core and valence band satellites, band narrowing, and rapid photoemission peak broadening away from E_F , there are sufficient agreements with the overall DOS (at least for the 123 structure) that it should be considered a good starting point for the electronic structure. For example, the calculated Fermi surface for both the 123 and 2212 structures is reasonably well reproduced experimentally and the bands at E_F consist of p-d hybridized orbitals just as predicted by local density functional theory.

Our spectra clearly show that a BCS-like DOS is obtained at the Fermi energy as a gap opens up below T_c . This is just one more indication that the old conventional models should be considered more seriously.

Normal State Electronic Structure and the Superconducting Energy Gap in HTSC's as Determined From Photoemission Spectroscopy

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Photoemission spectroscopy is utilized to determine the electronic structure of high- T_c materials. The observation of dispersive bands at E_F suggests a Fermi surface similar to that obtained from a band calculation. These results, together with a BCS-like energy gap, are not inconsistent with the notion of a Fermi liquid consisting of hybridized p-d bands.

Key words: Photoemission, Band Structure, Fermi Surface, BCS Gap, Correlated Fermi Liquid

I. INTRODUCTION

In spite of intense efforts by a large body of researchers, the nature of the electronic structure of high- T_c materials remains elusive. Early measurements on poorly characterized sintered specimens yielded a variety of confusing and contradictory results which spawned a number of theories¹, almost none of which have been conclusively proven or disproven to this day. The early photoemission measurements² on sintered 123- and 214-type samples were a major contributor to the conflicting results, since they yielded spectra that showed no intensity at the Fermi energy, thus fueling speculation that we are possibly dealing with insulators or, at best, semiconductors. At about the same time that the more stable 2212 compounds with their well-defined Fermi edge were discovered, we found that the failure to observe emission at E_F in the 214's and the 123's was due to a surface reconstruction caused either by a near surface oxygen loss, or the formation of an insulating oxide on the surface, or both (The surface reconstruction was actually first observed by Takahashi et al.³, in the 214's, although they failed to observe a well-defined Fermi edge). Thus at least it was clear that we are dealing with metals, although the verdict is still not in on whether these are conventional metals (i.e., spin 1/2 Fermions) or a more exotic version⁴ where spin and charge reside on different particles.

Existing theories can be roughly divided into three categories: 1) Conventional metals, whose properties can be understood from a band picture⁵; 2) Correlated Fermi liquids, whose properties can be derived from some variant of the Hubbard or Anderson model⁶; and 3) Unconventional metals, or non-Fermi liquids⁴. It is difficult to experimentally distinguish between the various approaches, since most theories are still in the evolutionary stages. Within each approach, for example, there is now the prediction of a reasonably sharp momentum cutoff at the Fermi energy and a well-defined Fermi surface, which is often cited, perhaps naively, as the mark of a Fermi liquid. We will try to show that the normal state electronic structure is at least not inconsistent with the notion of a correlated Fermi liquid, and that band calculations can serve as a good starting point in determining the density of states.

In the following sections we will review our most significant data and try to point out the features which can be utilized to distinguish between the various models. We will show that at least in the 123 materials the density of states (DOS) is well described by a band calculation, that in general there exists a Fermi surface resembling a calculated Fermi surface, that the states at E_F consist of hybridized p-d bands, that to within 30 meV the Fermi energy is indistinguishable from that of a Fermi liquid, and finally that a BCS-like superconducting energy gap with its associated pile-up of

the DOS at E_F , is clearly distinguishable at $T \ll T_C$. All of which would point to a correlated Fermi liquid ground state.

II. EXPERIMENTAL

The research reported here was performed at the Synchrotron Radiation Center in Stoughton, WI, on the Ames-Montana ERG-Seya beamline, as well as on the Minnesota-Argonne-Los Alamos ERG beamline. The angle integrated measurements were done using a CMA electron analyzer with a best overall resolution of 100 meV (usually 200 meV), while the angle-resolved work was done with a VSW ARIES system having an overall best resolution (including beamline) of 30 meV. The single crystals of HTSC materials were cleaved in-situ at $T = 20$ K to avoid surface reconstruction. A chamber base pressure of 3×10^{-11} Torr insured an adsorbate free surface for a period of at least 24 hrs. As we have shown previously⁷⁻⁹, it was necessary in the case of 123 materials to make all measurements at $T = 20$ K since the surface reconstruction rapidly sets in at elevated temperatures. The much more stable 2212's could be heated above T_C (≈ 85 K) without noticeable damage to the surface.

III. RESULTS AND DISCUSSION

A) The Densities Of States

An angle-integrated photoemission spectrum in essence measures the density of states (DOS) of a material, convoluted with an energy-dependent transition cross-section. A number of methods have been developed to calculate the cross-sections for a given material, and a calculation of a photoemission spectrum starting from a DOS is now common procedure¹⁰. In this section we will compare such theoretical spectra with our results. However, because photoemission is such a surface sensitive measurement, it is necessary first to consider the problem of surface reconstruction, and the need to insure that the measured spectra are representative of the bulk electronic structure.

In Fig. 1 we show angle-integrated spectra of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ for $x = 0.9$ (spectra A and B) and 0.25 (spectrum C) at $h\nu = 50$ eV. The energy resolution is 200 meV. Spectrum A was taken at 20 K in the a-b plane of a freshly cleaved single crystal. It shows a resolution limited Fermi edge, as well as substantial intensity in the -1.5 to -2 eV region. As the sample is warmed to 300 K (spectrum B) the intensity at E_F , as well as in the -1.5 to -2 eV region, is dramatically reduced. Associated with the loss of intensity at E_F is the growth of the satellite at -9.5 eV which is known to have O-2p character owing to its resonant enhancement at $h\nu = 22$ eV. These effects are easily understood if it is postulated that an insulating surface oxide, not related to the 123 structure, has formed on the surface from the 123 structure in the near surface region. In all probability this is some form of Ba-O, since there is reason to believe that the crystal cleaves in the Ba-plane (i.e., Ba core levels show a surface component). At low temperatures this reconstruction is inhibited. The fact that we are not dealing with a simple case of oxygen loss can be deduced from curve C where an oxygen-deficient crystal shows no sign of the -9.5 eV satellite, even while showing no intensity at E_F . The -9.5 eV satellite is thus not related to the 123 structure, and need not be considered as a mystery peak. It is an oxygen satellite from a more localized (insulating, as evidenced by charging effects) surface oxide. We find that the rate of surface reconstruction is a strong function of stoichiometry and surface perfection. Thus the observation of a Fermi edge at room temperature is not inconsistent with our data if the measurements are made on fully oxygenated samples with a non stepped surface.

In Fig. 2a we compare an angle-integrated spectrum for $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$, taken at 20 K at a photon energy of 70 eV, to a calculated spectrum from Ref. 10. In this figure, the secondary electron background has been subtracted out in the usual fashion in order to better compare with calculations. By shifting the calculated spectrum by 0.5 eV to higher binding energy we get almost exact agreement between band theory and experiment. All the features in the spectrum are

reproduced in the calculation, including the bottom of the band at -6 eV. The major disagreements are the 0.5 eV shift, and a sizable reduction in the intensity at E_F , both of which could be a consequence of the effects of electron correlation. Spectra at other photon energies show a similar agreement, thus indicating that the calculation obtains the correct mix of 2p and 3d orbitals. Thus it is strongly suggestive that at least for $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$, a band calculation correctly captures the essential features of the DOS.

Fig. 1 Valence band spectra for single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ at $h\nu = 50$ eV: a) $x = 0.9, T = 20$ K; b) $x = 0.9, T = 300$ K; c) $x = 0.25, T = 20$ K.

Fig. 2 Comparison of valence band spectra (with backgrounds subtracted) to calculated spectra from Ref. 10 and 11 at $h\nu = 50$ eV: a) $\text{YBa}_2\text{Cu}_3\text{O}_8$; b) $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

The same cannot be said for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$. In Fig. 2b we show a similar comparison of a spectrum to a band calculation¹¹ as in Fig. 2a. The disagreements are rather obvious. Takahashi has shown that not only the position, but also the curvature of the band at -5.5 eV is completely at odds with band structure results. We would find it unlikely that these disagreements are the result of increased correlation effects in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ vs. $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$. This particularly in view of the fact that the -12 eV Cu satellite, the major indicator of Coulomb correlation, is much smaller in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ relative to the valence band maximum than it is in $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$. It is more likely that the band calculations themselves should be re-examined. In particular, it will be necessary to include relativistic effects of the heavy Bi atom, as well as the effects of the superlattice, before any definitive comparisons can be made. The relativistic effects may be important at E_F as well, where band calculations find a Bi-O band crossing E_F near the M-point in the zone. Indications are that this crossing may not exist (see below).

B) Band Dispersion And The Fermi Edge

Angle-resolved data were first obtained by Takahashi et al. in the 2212 system. They showed a poor correspondence with band calculations. However, the very existence of dispersion and a crossing of the Fermi energy, indicated that a Fermi surface must exist for this system, and that the possibility of a Fermi liquid electronic structure must be considered for the normal state. The data of refs. 12 and 13 did not convince all skeptics of the existence of a Fermi surface owing to a low count rate at E_F , and a relatively low resolution. The low count rate necessitated work at 18 eV photon energy in order to enhance the intensity via a presumed Fano resonance. However, because of very strong matrix element effects, the jury is not yet in on whether this is a true Fano

resonance. In our 2212 samples the intensity at E_F was generally more than 5% of the valence band maximum so that it was possible to obtain data away from 18 eV where dispersion is much more pronounced. We preferred not to deal with the additional complication of an unknown resonance at 18 eV. With much higher resolution and intensity, there was no doubt about a band crossing. An analysis of the $h\nu$ -dependence indicated that the bands at E_F consist of about a 30-70 mix 3d and 2p orbitals.

In Fig. 3 we display angle resolved data taken along the [110] direction at a temperature of 20 K, showing only the data within the first 350 meV of E_F . The total dispersion shown in Fig. 3 is approximately half the predicted value (on the order of 400 meV), although the Fermi level crossing (at $\theta = 20^\circ$) is at approximately the k -value where the Bi-O band is expected to cross E_F . Along the [100]-type directions, the crossing of E_F occurs precisely at the predicted values, so that at least in these directions the calculated Fermi surface has the correct topology.

There are several interesting features of the data in Fig. 3. First it will be noticed that the dispersive feature is considerably broader below E_F than at E_F . Indeed, the broadening goes as $(E - E_F)^2$, although because of the large energy scale it is not certain that this can be used as a proof of Fermi liquid behavior. The rapid broadening may, however, be an indication of correlation effects, since the broadening should be related to the effective mass. This is also reflected in the narrower than predicted band width.

A second noticeable feature is the asymmetry of the photoemission peaks due to a relatively large background, which cannot be entirely accounted for on the basis of secondary electrons. Indeed, less than half of the background intensity can be associated with secondaries. It has been suggested that this background is possibly a manifestation of the diffuse background of electronic states in the non-Fermi liquid theories, while the well-defined peaks represent holon or spinon bands. We would point out that while we cannot rule out this possibility, there are other, much more mundane sources of background intensity, such as surface imperfections, which should be considered. This particularly in view of the fact that there is considerable sample dependence to the data. For example, in some crystals of 2212 material, no dispersive features are evident near E_F , in spite of a sharp superconducting transition. Only a featureless background is seen. Under normal circumstances we would interpret this as a bad surface due to a bad cleave. But, in addition, we also wish to point out that in Fig. 3 for $\theta > 22^\circ$ (i.e., for k -values beyond the Fermi level crossing), there is almost no background intensity. This is not inconsistent with the concept of limited surface damage with some residual k -dependence in the damaged regions. It would, on the other hand, necessitate one more modification to the non-Fermi liquid theories, in order to include k -dependence in the diffuse background of electronic states.

It had been assumed that one could distinguish between a Fermi liquid and a non-Fermi liquid from the sharpness of the Fermi edge, with the Fermi liquid following Fermi-Dirac statistics exactly. However, it is now predicted^{16,17} that the differences (if any) may be extremely subtle. In Fig. 4a we show overlayed on the same plot, the Fermi edges of Pt metal and that of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ taken at 90 K (i.e., above T_C). To within experimental resolution of 30 meV (i.e., smaller than the broadening due to the Fermi function) there is no difference between them. Any deviation from a Fermi liquid is on a scale smaller than 30 meV, or a temperature equivalent to T_C .

C) The Superconducting Energy Gap

One can see in Fig. 3 from data taken at 20 K, that as the band approaches the Fermi energy, the lineshape of the peak structure changes dramatically. A sharp, resolution limited peak develops at E_F at 20 K (i.e., below T_C), but not above T_C , as seen in Fig. 4a for $\theta = 18^\circ$. Associated with the peaked structure is a decrease in the Fermi energy, which can be seen in Fig. 4b where Fermi edge overlays for the same samples and the same angles as in Fig. 4a are again shown, but now at 20 K. The most straightforward explanation for the sharp peak and the change in the Fermi energy is

that we are observing a BCS-like energy gap opening at E_F , with its associated pile-up of states. Modeling studies show that the spectrum in Fig. 4b (for the 2212 material) can be obtained from the spectrum in Fig. 4a by convoluting the latter with a BCS density of states function, assuming an energy gap $2\Delta = 7 kT_C$. This gap value, which shows little anisotropy with k , is exactly twice the theoretical BCS value, and thus should be an interesting clue in solving the coupling mechanism.

Fig. 3 Energy dispersive curves for a single crystal of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ along the Γ -M direction in the Brillouin zone at $T = 20 \text{ K}$ and $h\nu = 22 \text{ eV}$. The Fermi energy crossing of the dispersive peak is near 200° . Note the lineshape change near E_F .

Fig. 4 Comparison of the Fermi edge of Bi-2212 (at $\theta=180^\circ$) to that of Pt: a) at 90 K; b) at 20 K.

IV) CONCLUSIONS

It is difficult to distinguish between Fermi liquid behavior in the new high- T_C superconductors and behavior expected from the novel new non-Fermi liquid theories. The differences are predicted to be on an energy scale smaller than our experimental resolution. However, while deviations from simple band theory certainly do exist in the form of core and valence band satellites, band narrowing, and rapid photoemission peak broadening away from E_F , there are sufficient agreements for the overall DOS (at least for the 123 structure) that it should be considered a good starting point for the electronic structure. The calculated Fermi surface for both the 123 and 2212 structures is reasonably well reproduced experimentally; the bands at E_F consist of p-d hybridized orbitals as predicted. A large BCS-like energy gap is observed below T_C .

This work was performed under the auspices of the U.S. Department of Energy. The Ames Lab. acknowledges DOE support under contract # W-7405-ENG82 while Argonne National Lab acknowledges DOE support under contract # W-31-109-ENG-38. The Synchrotron Radiation Center, whose staff we wish to gratefully acknowledge for their assistance, is supported by the NSF under contract #DMR8601349. KV is supported under contract #STC-880954.

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