Luttinger Fermi Surface of Metallic Gap Spectral Weight in Nd_{1.85}Ce_{0.15}CuO_{4-y}

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Angle-resolved photoemission measurements for bulk single crystals of $Nd_{2-x}Ce_xCuO_{4-y}$ with x = 0.15 show that the spectral weight which builds in the gap of the x = 0 insulator displays a Luttinger Fermi surface, the first such observation for a material which is electron doped and which must be driven metallic by alloying. This finding greatly strengthens our previous conclusion that the metal's chemical potential lies in gap-filling spectral weight, and sharpens the distinction between theories in which this does or does not occur.

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The normal metallic state of the superconducting cuprates results from doping a parent compound which is insulating because of the large Coulomb repulsion U_{dd} between 3d electrons at the Cu sites. In the simplest paradigm materials, i.e., hole doped $La_{2-x}Sr_{x}CuO_{4}$ and electron doped $Nd_{2-x}Ce_xCuO_{4-y}$, the x=0 insulating parent compound violates the Wilson rules and in turn the Luttinger theorem [1], the former in that the planar Cu-O antibonding band of $d_{x^2-v^2}$ symmetry is half filled so that the material should be a metal, and the latter in that the volume of the noninteracting system's Fermi surface is thus not preserved in the presence of U_{dd} , but is instead rendered zero. The scenario whereby doping then produces a metal, and the nature of the resulting metallic state, is a fundamental question in its own right and is even thought by many to hold the key to explaining the large values of the superconducting transition temperature T_c found in some cuprates.

Angle-resolved photoemission spectroscopy (ARPES) studies of the metallic state of the cuprates Bi2Sr2Ca- Cu_2O_8 and $YBa_2Cu_3O_7$ have made the remarkable finding of a Fermi surface which satisfies the Luttinger theorem [2,3], i.e., has the volume predicted in a band calculation. Both of these materials are (a) hole doped and (b) more complex electronically than the simpler paradigm materials cited above because they have additional bands crossing the Fermi energy (E_F) , enabling internal doping of the planar Cu-O $d_{x^2-v^2}$ band such that the materials are metals without alloying. Here we report the observation by ARPES of a Luttinger Fermi surface for $Nd_{1.85}Ce_{0.15}CuO_{4-\nu}$. This result is significant as the first instance of a Luttinger Fermi surface for a material which (a) is electron doped and (b) has the simpler one-band paradigmatic electronic structure which requires alloying to achieve a metal.

elucidating the route to the metallic state. Based on angle-integrated photoemission spectroscopy (PES) data for ceramic samples of $La_{2-x}Sr_xCuO_4$ and single crystal and ceramic samples of $Nd_{2-x}Ce_{x}CuO_{4-y}$, we have previously advanced a scenario of the doping-induced insulator-to-metal transition in which either hole or electron doping leads to a growth of spectral weight in the gap of the insulator, with the chemical potential μ of the metal lying in this new spectral weight [4]. Central to our scenario was the expectation that the metallic gap states would display a Luttinger Fermi surface. The present result verifies this expectation for $Nd_{2-x}Ce_{x}Cu$ - $O_{4-\nu}$ and also shows clearly that the gap weight is not analogous to an impurity band [5].

This finding of gap spectral weight and the metal μ position disagrees fundamentally with some current theories, but is of central importance to others, as summarized near the end of the paper. Further, data from oxygen 1s x-ray absorption spectroscopy (XAS) have been interpreted [6] as showing the alternate picture that μ lies either in the insulator's valence or conduction band for hole or electron doping, respectively. The fact that μ is observed directly in photoemission, but must be inferred after data interpretation in XAS, strongly favors the photoemission result on this question, but the greater surface sensitivity of photoemission relative to XAS has been cited as a reason to ignore the photoemission results. In this paper we also present new data on the buildup of the gap spectral weight, and we point out that our finding of the Luttinger Fermi surface adduces to this gap spectral weight the same credibility as is now given to the near- E_F ARPES data in Bi₂Sr₂CaCu₂O₈ and YBa₂Cu₃O₇. We also note that varying x or δ to produce an insulator in $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_8$ [7] and $YBa_2Cu_3O_{7-\delta}$ [8] produces a non-rigid-band loss of ARPES weight over a range of energy 0.4 to 0.6 eV below μ , although there is

This result is also highly significant in the context of

additionally a shift of μ by 0.1 to 0.2 eV over the range of x or δ needed to produce the insulator, probably involving the carrier reservoirs associated with their more complex electronic structure.

The samples of Nd_{2-x}Ce_xCuO_{4-y} studied in this work are single crystals with x = 0, 0.10, and 0.15 and ceramics with 0 < x < 0.21, prepared at the University of California at San Diego as described in Refs. [9-11]. Two sets of ceramics and eight single crystals yielded consistent results. The single crystal samples with x in the superconducting regime show sharp resistivity transitions, with $T_c \sim 18$ K. X-ray diffraction measurements show the socalled T' phase for all samples. Sharp x-ray Laue patterns were observed and used in orienting the single crystals.

The angle-integrated photoemission measurements were made both at the Synchrotron Radiation Center (SRC) and at the National Synchrotron Radiation Light Source (NSLS) using, at the SRC, monochromatized synchrotron radiation on the Ames-Montana and 062 beam lines, and at NSLS, the U4A beam line. The ARPES measurements were made on the Ames-Montana beam line. The ceramic and single crystals were cooled to, cleaved, and measured at ~ 100 K and ~ 20 K, respectively, *in situ*, in a vacuum better than 5×10^{-11} torr. Kinetic energies of the emitted photoelectrons were analyzed with a cylindrical mirror analyzer and with a 50 mm radius hemispherical analyzer having a 2° full angular acceptance angle for the angle-integrated and the angle-resolved studies, respectively. An overall resolution of 150 meV was found to be adequate to resolve all features in the ARPES spectra, and the resolution for the angle-integrated spectra ranges from 170 to 230 meV. For the metallic samples the energy of the chemical potential was determined from the Fermi edge of a clean Pt foil in thermal and electrical contact with the sample. The spectra from almost all samples were free of the socalled 9 eV peak, commonly considered an indication of poor sample quality. The only exceptions were the angle-integrated single crystal data, where weak 9 eV emission sometimes occurred. In the ARPES studies, selected test spectra were found to be reproducible over a 5 day duration, at which time 9 eV emission first appeared, along with a diminution of emission near E_F .

Figure 1 shows the x-dependent growth of spectral weight within the insulator gap for the angle-integrated photoemission spectra of ceramic samples. The inset shows the entire valence band spectrum for x=0 and 0.15. The emission between 9 and 15 eV binding energy is the well-known Cu $d^9 \rightarrow d^8$ "satellite" part of the spectrum, which is resonantly enhanced for the photon energy hv=74 eV. The x=0 insulator spectrum has been positioned on the energy axis relative to those for nonzero x by aligning the higher binding energy main band and satellite features, whose relative energies are essentially independent of x, as found previously [4], and



FIG. 1. Angle-integrated photoemission spectra of Nd_{2-x} -Ce_xCuO_{4-y} polycrystals. The position of E_F is about 0.5 eV above the top of the valence band which implies that the spectral weight induced by doping lies in the insulating gap.

as can be seen in the inset. The main part of the figure shows that E_F falls ~0.5 eV into the ~1.5 eV gap [12] of the insulator, i.e., a full 1 eV away from the insulator conduction band, and does not change noticeably with x for nonzero x. These data confirm previous synchrotron [4] and x-ray photoemission results [13] for polycrystalline samples. We mention that for the samples measured at the SRC, O 1s XAS data taken at the same time, using the electron yield mode, are identical to published XAS data taken in the absorption mode [14], showing the necessity of eventually finding a unified interpretation of the data from both spectroscopies.

Figure 2 shows angle-integrated spectra like those of Fig. 1, but for single crystals with x = 0, 0.1, and 0.15. Our previous [4] single crystal spectra were only for x = 0.15. Comparison by eye with Fig. 1 reveals small differences in the main band shapes, and greater sharpness in the single crystal d^8 peaks. For the buildup of spectral weight in the insulator gap, the single crystal and



FIG. 2. Angle-integrated photoemission spectra of Nd_{2-x} -Ce_xCuO_{4-y} single crystals are very similar to those of the polycrystals of Fig. 1.

ceramic data agree very well, except that the x=0 onset is somewhat sharper in the single crystal.

Figure 3 shows ARPES spectra for a Nd_{1.85}Ce_{0.15}Cu- $O_{4-\nu}$ single crystal taken for various values of k along the Γ -X direction of the Brillouin zone. An asymmetric peak emerges from a flat background at about 400 meV binding energy and crosses E_F for the spectrum with k = 0.50 Å⁻¹. A band calculation, done for the x = 0material [15] in the local density approximation (LDA), is shown as an inset together with bars to indicate the positions of the measured peaks. E_F lies in the single $d_{x^2-y^2}$ antibonding band and, following Ref. [15], has been positioned 0.2 eV higher in energy for x = 0.15 than for the x=0 calculation, assuming a rigid shift with added electrons. With this shift, the experimental dispersion and the value of k_F is very close to the band theory prediction. Two differences from findings for the hole doped materials are that the dispersion in the Γ -X direction is little renormalized from the band theoretical results, and that the peak does not sharpen greatly as k approaches the Fermi surface. The former is puzzling for a material with strong Coulomb interactions, while the latter may be caused by scattering associated with the alloying. Additional details of these spectra will be presented elsewhere, including studies which show that the polarization dependence of the excitation in Fig. 2 agrees perfectly with group theoretical ARPES selection rules [16] for $d_{x^2-\nu^2}$ and that along the Γ -M direction the excitation disperses



FIG. 3. ARPES spectra for the Γ -X direction in Nd_{1.85}-Ce_{0.15}CuO_{4-y}. An asymmetric peak emerges from 0.4 eV binding energy in the Γ -X direction and disperses through E_F . The inset shows an LDA band theory calculation for the undoped system [15], shifted 0.2 eV deeper in binding energy, together with points taken from the observed dispersion.

to within $\sim 250 \text{ meV}$ of E_F , but does not cross E_F . We also note a very strong hv dependence of near E_F intensity of the ARPES spectra, such that the information presented here is observable only for hv < 18 eV in the range 15 < hv < 30 eV. The near- E_F dispersion can also be observed for hv around 70 eV.

Figure 4 summarizes the ARPES results for the x=0.15 Fermi surface. Open circles mark the places in k space for which spectra were taken, and closed circles mark the observed E_F crossings. The hole pocket around X is roughly circular with a radius 0.6 Å⁻¹. The solid line is a theoretical Fermi surface for x=0.15, obtained by us from a tight-binding planar band structure including O-O interactions, which reproduces the x=0 LDA Fermi surface [15]. The LDA Fermi surface is shown as a dashed line. The excellent agreement between experiment and theory for x=0.15 shows that the Luttinger sum rule is indeed obeyed.

The location of the chemical potential is emerging as a fundamental issue for theories of doped cuprates. It is a tautology that the first hole or electron added to an insulator must result in μ at a band edge. Even for larger doping, Lanczos and Monte Carlo numerical studies of the commonly used one and three band Hubbard models suggest that μ crosses the gap when electron doping replaces hole doping [17]. Although some of these studies generate considerable spectral weight in the gap upon doping, and thus do not act like rigid band systems, nonetheless μ lies very near the insulator band edges.

However, a quite different situation is obtained if the system achieves a new ground state in the presence of many holes or electrons, i.e., if a phase transition occurs with doping. For models of this type μ in the gap is possible and even central. For instance, Ruckenstein and Varma [18] have formulated a microscopic theory of the marginal Fermi liquid in which μ is renormalized into the gap by a kind of excitonic instability. It seems possible



FIG. 4. The Fermi surface determined by ARPES for Nd_{1.85}Ce_{0.15}CuO_{4-y}. Open circles mark the places in **k** space for which spectra were taken, and closed circles mark the observed E_F crossings. The diameter of the circles corresponds to the 2° experimental resolution. The solid line is a tight-binding calculation for the x = 0.15 Fermi surface and the dashed line is the calculated x = 0 LDA Fermi surface.

that Anderson's Luttinger liquid picture [19] could ultimately lead to μ in the gap as well [20]. In connection with these theories, we take cognizance of current thinking [19,21] that a Luttinger Fermi surface does not inevitably imply a Fermi liquid, contrary to the tacit assumption made in our previous paper [4]. In another kind of phase transition scenario, Emery and Kivelson [22] interpret spectral weight and μ in the gap as being a consequence of a dynamic, microscopic phase separation between insulating and metallic regions in the material.

In summary, ARPES spectra for bulk single crystals of $Nd_{2-x}Ce_xCuO_{4-y}$ with x=0.15 show that the spectral weight which builds in the gap of the x=0 insulator displays a dispersing asymmetric peak with E_F crossings at k values consistent with band theory. This is the first observation of a Luttinger Fermi surface for a material which is electron doped and which must be driven metallic by alloying. This finding greatly strengthens our previous conclusion [4] concerning the chemical potential in doped cuprates, and highlights theories in which a new ground state is achieved upon doping.

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