Electronic structure of $Nd_{1.85}Ce_{0.15}CuO_4$: Evidence for a disparity between hole and electron doped cuprate superconductors

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We have performed angle resolved photoelectron spectroscopy (ARPES) on the electron doped cuprate superconductor $Nd_{1.85}Ce_{0.15}CuO_4$. We find a linear dependence of the peak width (scattering rate) on the binding energy along the zone diagonal, which is in contrary to one's naive Fermi liquid expectation. In addition, two components with very different momentum dependences are seen in the near E_f spectral function. This contrasts strongly with the hole doped materials.

1. Introduction

$Nd_{1.85}Ce_{0.15}CuO_4$ is one of a few electron doped cuprate superconductors.[1] The undoped material is an antiferromagnetic insulator. With the substitution of tetravalent Ce for trivalent Nd, the Neel temperature rapidly drops around x=0.13, with superconductivity occuring between $0.14 < x < 0.18$ for oxygen reduced samples. As compared to the hole doped materials, the antiferromagnetic phase persists to much higher doping levels with superconductivity occuring in a doping range that is narrower by a factor of almost 5.

$Nd_{2-x}Ce_xCuO_4$ and other electron doped cuprate superconductors show very different behavior as compared to the p-type materials; transport measurements seem to indicate a more conventional normal state that is reminiscent of Fermi liquid behavior (ρ is linear in T^2 vs. linear in T)[2]; there is evidence for both electron and hole charge carriers in the temperature dependence of the Hall coefficient[3]; the incommensurate neutron scattering peak that is seen in the hole doped cuprates appears to be absent in $Nd_{2-x}Ce_xCuO_4$.[4]

Previous ARPES experiments on $Nd_{2-x}Ce_xCuO_4$ have succeeded in revealing the presence of a Luttinger volumed Fermi surface and a $(\pi, 0)$ extended saddle point that is found 350 meV below E_f.[5] These features were shown to be well postdicted by the $t - t' - t'' - J$ model.[6]

2. Experimental

Single crystals of $Nd_{1.85}Ce_{0.15}CuO_4$ were grown by the traveling solvent floating zone method. The as-grown crystals are not superconducting and must be subsequently annealed in flowing argon gas for 100 hours at 1000 C. The annealing lowers the oxygen content by 0.5% and is believed to remove oxygen from apical sites that can exist as impurities in the T' phase. The resulting crystals show an onset of superconductivity at 25K and a superconducting volume (Meissner shielding) of almost 100% at 20K.

Recent advances in photoelectron spectroscopy are allowing for unprecedented energy and momentum resolution. The possibility of energy resolution on the order of 5 meV and angular resolution < 0.5 degrees makes $Nd_{1.85}Ce_{0.15}CuO_4$ an ideal system for reinvestigation. These ARPES measurements were performed at the newly commissioned beamline 5-4 of the Stanford Synchrotron Radiation Laboratory. This system is a Scienta SES 200 electron spectrometer coupled with a normal incidence monochromator (NIM)

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which is capable of 5 meV energy resolution and 0.2 degree angular resolution. Samples are positioned by a custom dual axis sample manipulator that can be cooled below 10 K.

Data reported here was collected with 10-15 meV energy resolution and an angular resolution of 0.5 degrees (1% of the Brillouin zone). The photons in all measurements had an energy of 16.5 eV and unless noted otherwise were polarized at 45 degrees to the Cu-O bonds. The chamber pressure was lower than 4×10^{-11} Torr. Samples were cleaved in situ at 10K which resulted in shiny flat surfaces. LEED shows clean, well-ordered surfaces with a symmetry commensurate with the bulk. All displayed spectra were taken in the superconducting state. Angle integrated photoemission reveal a large well formed valence band and a near E_f foot. All angle resolved measurements were performed on this foot. ARPES spectra did not change for duration of the experiment ($\simeq$ 24 hours) and were reproduced on a large number of samples.

3. Results

In Fig. 1 we show raw ARPES data from the three main symmetry direction in superconducting $Nd_{1.85}Ce_{0.15}CuO_4$. As can be seen in Fig. 1a, along Γ to (π,π) a large feature disperses out of the background, moves towards E_f, sharpens, and then disappears after passing above E_f. This behavior is relatively universal among the cuprate superconductors at optimal doping.[6][7] This is an interesting point. It can be determined that the feature's width is more or less linearly proportional to its binding energy. This is contrary to one's naive expectation of a E^2 dependence (Fermi liquid) as expected from the temperature dependence of the resistivity. In Fig. 2 the HWHM of the low binding energy side is plotted vs. the binding energy (defined as the feature's centroid) from EDCs from along the zone diagonal. One can see that it is linear over a large energy range. The finite constant offset is probably due to both finite temperature and finite instrument resolution.

The dispersion from Γ to $(\pi,0)$ differs from the p-type. In both cases the band has an extended
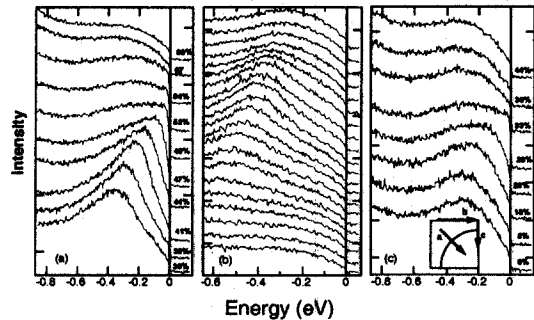

Figure 1. EDCs from high symmetry directions in the Brillouin zone taken at 10K (a). Γ - (π,π), (b). Γ - $(\pi,0)$ EDCs are from 18%-100% of π/a at even intervals, (c). $(\pi,0)$-(π,π), EDCs are as marked.

saddle region near $(\pi,0)$. However, the minimum is quite close to E_f in the hole-doped materials, whereas as seen in Fig. 1b it is 350 meV below.[6] This is significant in some models as the extended flat region creates a large density of states well below the Fermi energy, whereas in the hole doped materials a large density of states is created quite close to E_f.

Along $(\pi,0)$ to (π,π) an additional component appears in the EDCs. In Fig. 1c one can see that while the broad maximum shows a small dispersion ($< 100meV$), a smaller spectral weight and lower binding energy feature develops as a shoulder near the Fermi energy. Moving away from $(\pi,0)$ the large feature appears to disperse back to higher binding energy, while the low binding energy feature disperses towards E_f and then eventually disappears. The presence of the low energy component is seen more clearly in Fig. 3 where we plot EDCs along the ostensible Fermi surface. The two features are distinct in energy near $(\pi,0.2\pi)$ but as we approach $(\pi/2,\pi/2)$ the low energy feature loses spectral weight and disappears, while the high energy feature disperses towards E_f. This two component electronic structure had not been identified in earlier ARPES studies due to poorer resolution and is very dif-
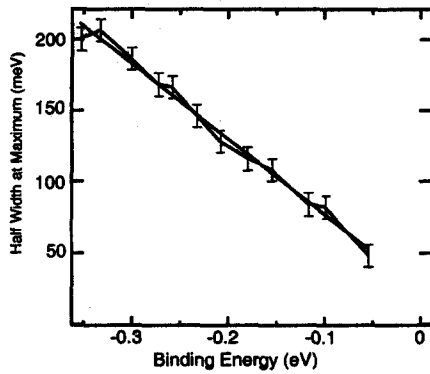
Figure 2. The half width at half maximum of the dispersive feature's width from along the zone diagonal plotted vs. the centroid energy of the same feature.

Figure 3. EDCs along the k_f contour of the large feature. Temperature was 10K. One can resolve the two features as seperate in energy. The arrows are a guide to the eye.

ferent from that of the p-type superconductors. In particular, the two near-E_f features in the spectral function of Bi2212 only become distinct below the superconducting transition temperature.[7] In the present case, the features are not associated with the onset of superconductivity as they are distinct in the normal state (not shown). The large higher binding energy feature shows a near E_f dispersion which is remarkably similar to that seen in the protoypical undoped insulator $Ca_2CuO_2Cl_2$.[8] This insulator's near E_f feature was seen to have a dispersion that was well approximated by the d-wave form. In the present case we also see a d-wave like dispersion along the ostensible Fermi surface i.e. the maximum of the large feature does not cross the Fermi energy near the $(\pi, 0.2\pi)$ position, which is near it's Fermi surface as determined from $\nabla n(k)$ (not shown).

4. Discussion

In a canonical Fermi liquid the low energy spectral feature should have a width proportional to the square of its binding energy by consideration of the phase space available for scattering. This E^2 scattering rate gives typical Fermi liquid be-
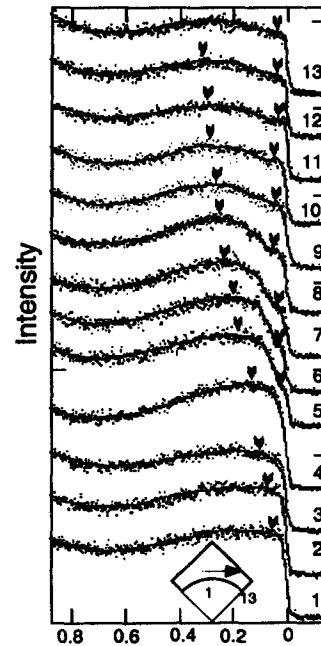
havior such as a T^2 dependence of the resistivity. In contrast, the hole doped cuprates are typified by many manifestations of a linear scattering rate which is a manifestation of their decidedly non-Fermi liquid behavior. This is observed in the linear temperature dependence of the in-plane resistivity,[9] the linear scattering rate in the optical conductivity,[10] as well as a linear energy dependence of the quasi-particle width in the hole doped cuprate Bi2212.[11]

In the present case of $Nd_{1.85}Ce_{0.15}CuO_4$, the material has a T^2 dependence of the resistivity. This has been thought to be an indication of perhaps more Fermi liquid-like behavior. The electrons that lie along the zone diagonal direction have by far the greatest contribution to this trans-

port as the Fermi velocity is maximal there. One might expect therefore that the zone diagonal features have a Fermi liquid like E^2 dependence of the scattering rate. As shown above, this expectation is not realized.

It is underemphasized that the linear T dependence of the in-plane resistivity in the cuprate superconductors only occurs for optimal doped samples. In the overdoped regime the resitivity regains its familiar T^2 dependence, while at least in Bi2212 the zone diagonal features in the ARPES spectra retain their linear dependence on binding energy of the scattering rate. This may mean that whatever operative mechanism is responsible for the linear temperature dependence of the resistivity in the optimal p-type cuprates is suppressed in the same way in $Nd_{1.85}Ce_{0.15}CuO_4$ as in the overdoped hole doped cuprates.

The existence of two features in the near-E_f spectral weight is interesting. In Hubbard models, one dopes electrons to a half-filled Cu-O plane by adding them to an upper Hubbard band, which consists of states which have a strong local character of double occupied Cu sites. In $Nd_{1.85}Ce_{0.15}CuO_4$ we should expect to observe a filled lower Hubbard band as well as a partially filled upper Hubbard band that are separated by an energy U that represents the large onsite Couloumb repulsion. In fact, we observe two features, but they are quasi-degenerate in certain regions of momentum space and there is no sign of the Hubbard U. It is of course possible that the second feature could be one not endemic to the CuO_2 planes, for instance a narrow impurity band driven by Ce doping. However, this is not likely due to the low Ce concentration and the feature's momentum dependence and finite dispersion.

5. Conclusions

In conclusion, we have performed angle resolved photoemission on the electron doped high temperature cuprate superconductor $Nd_{1.85}Ce_{0.15}CuO_4$. We find that the scattering rate of the near E_f peaks is linear with binding energy. This is quite similar to the non-Fermi liquid behavior seen in the p-type materials. In contrast to the p-type materials we see two components in the low energy spectral function. One shows a dispersion very similar to that observed in the undoped insulator, while the other is distinctly resolved quite close to the Fermi energy. The fact that two features are not predicted is a challenge to our understanding of the nature of doped electrons in the CuO_2 planes.

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