The question of how a metal evolves into an insulator is one of the most fundamental in solid state physics. For non-interacting electrons, the Fermi surface shrinks and eventually disappears as a band is filled. When electron-electron interactions dominate, the situation is less clear. By considering the strong Coulomb interaction, Mott qualitatively described how a material predicted by band theory to be a metal would in fact be an insulator. However, it remains unclear as to how the details of the electronic structure evolves from a half-filled metal to a Mott insulator. One of the most characteristic features of any metal is its Fermi surface which can be defined by the surface of steepest descent in the electron momentum distribution function, \( n(\vec{k}) \). Thus an equivalent question to the one above is how does the Fermi surface of a metal vanish as strong electron correlations drive the system into an insulator?

In the context of specific many-body models such as the Hubbard model, it has been shown that a structure in \( n(\vec{k}) \) survives even when the on site Coulomb U drives the system insulating, albeit the discontinuity in \( n(\vec{k}) \) which existed in the metal has been washed out. \( \boxed{2} \) This effect is linked to the fact that \( n(\vec{k}) \) reflects the underlying Fermi statistics of the electronic system. For the specific case of a two dimensional square lattice that resembles the CuO\(_2\) planes of the cuprates, there is a drop in \( n(\vec{k}) \) across a line that is close to the antiferromagnetic zone boundary. \( \boxed{3} \)

It is believed that the information under the sudden approximation of the momentum distribution function, \( n(\vec{k}) \), can be extracted from angle-resolved photoemission spectroscopy (ARPES) data via the relation \( n(\vec{k}) = \int A(\vec{k}, \omega) f(\omega) d\omega \). \( \boxed{3} \) Here \( A(\vec{k}, \omega) \) is the spectral function, and \( f(\omega) \) is the Fermi function. In a real experiment, the \( n(\vec{k}) \) structure would be further modified by the photoionization cross-section. In the metallic state of optimally doped Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\) (BSCCO) the steepest descent of \( n(\vec{k}) \) gives a Fermi surface consistent with traditional ARPES analysis methods, despite the complication of matrix elements. The intriguing new result is that the \( n(\vec{k}) \) pattern of the insulator, Ca\(_2\)CuO\(_2\)Cl\(_2\) (CCOC), is strikingly similar to the \( n(\vec{k}) \) pattern seen in BSCCO. \( \boxed{4} \) This realization, coupled with many-body theoretical calculations on various forms of the Hubbard model \( \boxed{3} \), suggests that the insulator pattern contains information that is related to a Fermi surface which has been destroyed by strong electron-electron interactions thus giving a qualitative concept of a remnant Fermi surface as the surface of steepest descent in \( n(\vec{k}) \). Although the detailed shape of the remnant Fermi surface is not crucial, it acts to emphasize a robust feature which we observe in the insulator. While this may not be a rigorous definition, as the Fermi surface is only defined for a metal, this idea allows a practical connection from the pseudogap seen in underdoped cuprates to the properties of the insulator. \( \boxed{3} \)

A photon energy dependence study of this issue is important to extract the intrinsic \( n(\vec{k}) \) structure from the raw data which can be affected by the matrix element. Because the single particle spectral function obtained from the half filled CCOC insulator will not suffer from possible complications arising due to intrinsic electronic inhomogeneity, bilayer splitting, or superstructure as in other cuprates, this study may be able to provide insight on some of the recent controversy regarding the Fermi surface seen in BSCCO.

In this report we show that despite the aberrations caused by the matrix element effect, the drop in \( n(\vec{k}) \) as one crosses the antiferromagnetic zone boundary is a robust feature of CCOC. This result differs from the conclusion of a recent study based on data from only a single cut across the magnetic zone boundary. \( \boxed{4} \) We also show that the dispersion remains independent of photon energy even when the matrix element does not. This is an expected result from an ideal two dimensional system.

The measurements are conducted at Beamline V-3 of...
the Stanford Synchrotron Radiation Laboratory (SSRL). The angular resolution is ±1°. The CCOC samples are oriented prior to the experiment by the Laue method and are cleaved in situ. The light is incident at 45° to the surface normal and has an in-plane component of the polarization along \( k_x \), which is also parallel to the Cu-O bonds. The chamber pressure is better than \( 5 \times 10^{-11} \) torr. Data is taken in the \( \vec{k} \)-space octant \((0,0) - (\pi,0) - (\pi,\pi) - (0,0)\) and at temperatures ranging from 100 to 185K, well below the Néel temperature of 250K. The insulating samples are checked for charging by varying the photon flux. All data are normalized by the photon flux which is measured by a Au mesh upstream from the sample.

We begin with a short discussion of matrix element effects in photoemission. Extracting the single particle spectral function from ARPES measurements is complicated by the fact that the measured photoemission intensity is a product of the spectral function and the matrix element. In interacting electron systems, it is impossible to calculate the matrix element exactly, thus further complicating the ARPES analysis. In general it is a function of the experimental geometry, photon energy, and the electronic wave function. However, symmetry arguments can be very powerful in understanding some of the general properties of the matrix element. Since its details are not well understood, the objective in a given photoemission study must be to focus on only those features of the data which are robust against variations in the matrix element.

For the \( n(\vec{k}) \) plots on CCOC, the spectra are integrated from -0.5 to 0.3 eV binding energy relative to the valence band maximum. Spectra are taken at the crosses, and the data is symmetrized about \( k_x = k_y \) to better illustrate the remnant Fermi surface. Figure 1 shows the original comparison between metallic BSCCO and insulating CCOC in panels (A) and (B). In both cases the geometry of the experimental setup is identical. Assuming a \( d_{x^2-y^2} \) orbital symmetry we would expect a vanishing cross section as one approaches the line \( k_x = 0 \). This is observed in both data sets. In BSCCO the only drop in intensity which is not naturally explained by matrix elements is where a Fermi surface crossing has occurred. As seen in the figure the intensity drop matches the traditional method for determining a Fermi surface crossing by following the dispersion by eye is indicated by the dots. A similar drop in intensity is observed in the insulator. Although the feature is less well defined here, the striking resemblance it bears to the metal suggests that the origin is similar, and hence it is qualitatively described as a remnant Fermi surface. This allows us to make a natural link between the dispersion in the insulator and the pseudogap in the underdoped regime of the high-T\(_c \) cuprates.

As mentioned above, we must still show the robustness of this feature to demonstrate the validity of this concept. Panels B through E of Figure 1 show \( n(\vec{k}) \) patterns of CCOC taken at photon energies of 25.2, 29, 32.3, and 41eV. At first glance they may appear different. One clearly sees the position of maximum intensity shift. Thus the position of this maximum along the antiferromagnetic zone boundary is a consistent feature. (F) shows the remnant Fermi surface determined from (B) through (E). The shaded region gives an estimate for the uncertainty in the remnant Fermi surface. The boundary of this shaded region is drawn with black lines on panels (B) through (E).

![FIG. 1. (color) Integrated spectral weight. The crosses indicate where spectra were taken. The data is symmetrized about the \( k_x = k_y \) line. A color scale is given on the right. (A) optimally doped BSCCO. The white hashed region indicates the approximate location of the Fermi surface determined from \( n(\vec{k}) \). The dots illustrate the position of the Fermi surface as determined by the traditional method for analyzing ARPES data. (B) CCOC shows a striking similarity of the insulator to the metal allows the identification of the white hashed region as a remnant Fermi surface. Comparison of (B) through (E) show CCOC taken at photon energies of 25.2, 29, 32.3, and 41eV. The intensity maxima varies between different panels, but the loss of intensity as one approximately crosses the antiferromagnetic zone boundary is a consistent feature. (F) shows the remnant Fermi surface determined from (B) through (E). The shaded region gives an estimate for the uncertainty in the remnant Fermi surface. The boundary of this shaded region is drawn with black lines on panels (B) through (E).](image)
exact shape of the remnant Fermi surface may change slightly, but at all photon energies used there is a loss of spectral weight as one crosses the approximate AF zone boundary from (0, π) to (π, 0). Panel F plots the remnant Fermi surface as determined by figures 1B through 1E. The shaded region in F gives a rough estimate for the shape and uncertainty of the remnant Fermi surface. Thin black lines in panels B through E form the border of the shaded region shown in F. It may appear that the remnant Fermi surface is more hole like or electron like depending on the photon energy chosen, but the broadness and the minor variability due to matrix elements prevent one from clearly identifying the remnant Fermi surface as either case, as illustrated by the hashed region. However, the point which is clear from the data is that globally, there is always an unexpected loss in intensity as one crosses the region in the Brillouin zone indicated by the shaded area in figure 1F, which represents the remnant Fermi surface. Indeed, the remnant Fermi surface is a robust feature of the insulator.

Although the $n(\vec{k})$ image plots can provide a wealth of information and are extremely good for summarizing data, it is important to look at the raw data to fully appreciate what information is being given by the image plots. This is done in Figure 2. Panels a through d (e through f) plot the EDC’s from (0,0) to (π, π) ((0,0) to (π/2, π/2)) taken at 25.2, 29, 32.3, and 41eV photon energy respectively. We find the spectra are qualitatively similar. This is true even at 41eV where the peak is poorly defined throughout the zone. To examine them more closely, Figure 3 plots both the dispersion of the peak position and $n(\vec{k})$ together for the (0,0) to (π, π) cut. Only slight differences exist between the 4 photon energies. It is apparent that the matrix element can be responsible for shifting the maximum in intensity relative to the maximum in dispersion. Such an effect is seen between the 29 and 41eV data, which may be rigorously compared. A similar shift of spectral weight has been reported by Haffner et al. in Sr$_2$CuO$_2$Cl$_2$. These authors conclude that there is no $n(\vec{k})$ structure or remnant Fermi surface. We believe the intensity shift is a small modulation due to the a matrix element compared with the robust underlying $n(\vec{k})$ structure seen throughout the zone at all photon energies. To illustrate this point we also plot the spectra, dispersion, and $n(\vec{k})$ along (π,0) to (0, π). From the raw spectra in figure 2 and the peak positions shown in figure 3 it is clear that the dispersion does not change. Meanwhile the intensity of the feature varies randomly along this cut. In the extreme case between 25.2eV and 29eV the intensity is increasing as one approaches (π/2, π/2) for the former, and decreasing for the latter. This completely contrasts the situation along (0,0) to (π, π), where there are only small shifts of the intensity profile, but the global structure remains the same.

The cuprates are generally believed to be two dimensional electronic systems. In such an ideal case, the electronic states probed by ARPES are independent of photon energy, and hence $A(\vec{k}, \omega)$ extracted from the spectra will remain the same. Thus it is natural to question this assumption when photon energy dependent shifts in the physics are reported. This may seem particularly true for the half filled cuprates which possess three dimensional
long range magnetic order. However, in figures 2 and 3 we have shown that the dispersion of the insulator is essentially independent of photon energy. Three dimensionality may still play a role in causing a small shift in dispersion with photon energy, but it is a safe assumption to treat the electronic structure of the half filled insulator as basically two dimensional.

Recently, the role of the matrix element effects is also under examination regarding the Fermi surface of BSCCO. Several groups have reported the existence of an electron-like Fermi surface centered at (0,0). These groups report a subtle change of dispersion near the flat band region around $(\pi, 0)$, causing a change in the shape of the Fermi surface. Largely due to the difficulty in understanding the effects caused by photoelectron matrix elements, there is no consensus in attempting to resolve this issue. The features in BSCCO are better defined than the relatively broad features in CCOC, and one might suspect that, as a result, they may be more susceptible to photon energy dependence. However, the idea that the dispersion should be independent of photon energy for an ideal two dimensional system is the same, regardless of the resolution. A change in the dispersion can not be a trivial matrix element effect. Possible explanations for a dispersion which changes with photon energy include: dispersion in $k_z$, bi-layer splitting, and phase separation. With the exception of Pb doped samples BSCCO also has the additional complication of superstructure in the Bi-O planes. In BSCCO, it would be surprising for the dispersion in $k_z$ to have a large effect considering the two dimensional nature of the cuprates. In reality though it is likely that all of these effects play roles which result in a potentially very complex picture.

Interestingly, models which have multiple component electronic structures where the wavefunctions and dispersions differ could have a simple explanation for the observed controversy. In such a case, variations in the matrix element could now lead to different interpretations of the observed ARPES spectra at different photon energies. This is possible because there may now be a different matrix element for each component, and each matrix element may vary differently with photon energy. Thus causing one component to be more dominant at one photon energy while hidden at another. This would naturally lead to different interpretations as a function of photon energy.

We have shown that the loss in intensity as one crosses the antiferromagnetic zone boundary is a robust feature of the insulator, $\text{Ca}_2\text{Cu}_2\text{O}_2\text{Cl}_2$, which can not be explained solely by matrix element effects. However, the photon energy dependence does underscore the qualitative rather than quantitative nature of the remnant Fermi surface concept. We argue that much physics can be learned in spite of the effect which matrix elements can have in ARPES, as long as care is taken to properly sort out the intrinsic versus the extrinsic physics. In particular, the resulting connection between the d-wave like dispersion and the pseudogap in the underdoped regime is robust.

We thank C. Dürr, M.S. Golden, and J. Fink for very open and stimulating discussions. This work was performed at Stanford Synchrotron Radiation Laboratory which is supported by the DOE Office of Basic Energy Science, Division of Chemical Sciences and Material Sciences. It is also supported by the ONR grant N00014-98-1-0195 and NSF grant DMR-9705210.

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