

Synchrotron Studies of Narrow Band and Low-Dimensional Materials

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I. Overview

Objective and technical approach of the project The objective of this work is to determine the single-particle electronic structure of selected narrow band and low dimensional materials and to analyze the data with a view to elucidating the relation between their electronic structures and novel low energy properties, such as mixed valence, heavy Fermions, the Kondo effect, insulator-metal transitions, non-Fermi liquid behavior or high temperature superconductivity.

We measure the single particle electronic structure of a material using photoemission spectroscopy (PES) and inverse photoemission spectroscopy (IPES). The DoE contract supports PES work at various synchrotrons. These data are combined with data from X-ray photoemission spectroscopy (XPS), ultraviolet photoemission spectroscopy (UPS), X-ray IPES (often called BIS), and Auger electron spectroscopy (AES), obtained in an in-house laboratory, supported by the NSF. It is very important to have both the synchrotron-excited and the in-house data for a given material. The synchrotron work is of three main types, (1) resonant angle-integrated PES at a variety of photon energies to identify and extract the spectrum of the narrow band electrons of the material, and (2) high resolution, low temperature angle-resolved photoemission spectroscopy (ARPES) to determine the crystal momentum (\mathbf{k}) dependence of the hole excitations of a single crystal. (3) the making of Fermi surface intensity maps. All these data are analyzed in the home laboratory by comparison to theoretical results from local density-functional (LDA) calculations, from solutions of model many-body Hamiltonians, from phenomenological single electron self-energies, and most recently to calculated many-body spectra from dynamic mean field theory (DMFT) combined with LDA, i.e. DMFT+LDA.

Plan and timing of the report This is the final report of the 12-year grant. The research descriptions are organized by topic.

Synchrotrons and collaborations The experiments were performed on various beamlines located primarily at the Stanford Synchrotron Radiation Laboratory (SSRL), the National Synchrotron Light Source (NSLS), the Advanced Light Source (ALS), the Synchrotron Radiation Center (SRC) of the University of Wisconsin, and the SPring8 Synchrotron in Japan. The work benefited greatly from close relationships at various times with personnel at these institutions: Z.-X. Shen (SSRL, ALS); L. H. Tjeng (NSLS); J.-H. Park (NSLS); C. G. Olson (SRC); H. Höchst (SRC); Jonathan Denlinger (ALS); S. Suga (SPring8). Tjeng was a postdoc in our group after leaving his NSLS postdoc. Park was a graduate student in the group before going to the NSLS, first as a postdoc and then as a beamline scientist. Denlinger came to the group as a postdoc after being a postdoc at the ALS, and then returned to the ALS as a beamline scientist.

Special PI Recognition During the term of this research grant the PI received the 2002 Frank Isakson Prize of the American Physical Society, "for outstanding contributions to the field of spectroscopy in strongly correlated electron systems leading to elucidation of many-body physics." The prize has been given every two years since 1980, "for outstanding contributions in the field of optical effects in solids," in the past only to an optical or infrared spectroscopist. The PI is the first recipient whose work has substantial photoemission, inverse photoemission, and x-ray synchrotron spectroscopy components.

II. Research Results

Publication numbers refer to the reprints/preprints list in sub-section III of this report.

A. 3d Transition Metal Oxides

1. *Test of systematics of the Zaanen-Sawatzky-Allen classification scheme*

In the early 90's we took advantage of the then newly developed capability at the NSLS Dragon Beamline to perform resonant photoemission spectroscopy (RESPES) of transition metal (TM) 3d states of a series of the most important transition metal oxides (TMO's) using photon energies at the TM 2p edge. Initially the Dragon beamline postdoc L. H. Tjeng had demonstrated such RESPES for CuO and had shown that it could reveal the 3d emission with much better contrast than the RESPES done up to then at the TM 3p edge. We formed a collaboration with Tjeng and we obtained from the Purdue Materials Preparation Center single crystal samples of CoO, MnO, Fe₃O₄, FeO, V₂O₃ and Ti₂O₃ with the particularly large size required at the beamline. Suitable samples of Cr₂O₃ were already in hand. One important thrust of the work was to make the first systematic test of the TMO 3d energy level structure postulated in the Zaanen-Sawatzky-Allen (ZSA) classification scheme, in which the early and late transition metal materials are proposed to be Mott-Hubbard (MH) and charge transfer (CT) materials, respectively. Especially for the early TMO's the 2p RESPES enabled the 3d spectrum to be clearly separated from the oxygen spectrum for the first time. The spectra revealed that for the early TMO's the ZSA scheme required modification to acknowledge that in the absence of metal-oxygen hybridization the materials lie slightly into the CT regime rather than being in the MH regime. But the effect of the large hybridization for the early TMO's is to produce an energy level structure whose lowest energy part can be regarded as effectively of the MH type. [Pub # 25].

2. *Verwey Transition in Fe₃O₄*

The crystal structure of Fe₃O₄ has tetrahedral A sites with Fe³⁺ ions and octahedral B sites with both Fe²⁺ and Fe³⁺ ions. In the famous Verwey transition at $T_V \approx 120\text{K}$ there is with increasing temperature a large jump increase in the conductivity which is generally understood to signal a insulator-to-metal transition caused by an order-to-disorder transition of Fe valence on the B sites, such that in the high temperature state the B sites fluctuate freely between the two valence states. In the simplest picture the material is then a metal above T_V , but the fact that in this phase the conductivity increases with increasing T shows that a simple metal picture is not correct. We used 2p RESPES to identify the emission near the Fermi energy (E_F) as involving Fe²⁺ 3d states. We studied this emission on heating through the transition and found that the low temperature phase band gap is not collapsed but is merely reduced by 50 meV, showing that a true insulator-to-metal transition does not occur. The change in the gap is quantitatively consistent with the two orders of magnitude conductivity jump at T_V . Thus even above T_V short-range charge ordering rather than full site equivalency dominates the single-particle excitations and the electronic properties. This result is consistent with the fact that the entropy of the high

temperature phase is much smaller than that which would obtain for free valence fluctuations, as emphasized long ago by P.W. Anderson. [Pub # 34]

3. *New understanding of metal-insulator transition in $(V_xA_{1-x})_2O_3$ ($A=Cr, Ti$)*

$(V_{1-x}A_x)_2O_3$ ($A=Cr, Ti$) is the most well known and well studied example of a Mott-Hubbard metal-insulator transition. The transition is traditionally interpreted within the framework of the Brinkman-Rice theory of the one band Hubbard model. Over the entire period of this report our work has had great impact in bringing the understanding of this famous system to an entirely new level of sophistication. The Dragon beamline work at the NSLS in the early 90's set the stage for the new developments. That work determined the valence band photoemission spectrum of all phases of the system for photon energies from 90 eV to 500 eV, the lower energy range giving spectra which are still the equal of any taken more recently and the higher energy range giving generally the first spectra at such high photon energies and particularly the first RESPEC results at the 2p edge. That work also included x-ray absorption spectroscopy (XAS) at the oxygen and vanadium edges, initially unpolarized and then extended to include the photon polarization dependence after UM graduate student J.-H. Park, who studied the system for his thesis work, became the NSLS Dragon beamline postdoc in 1994.

Theoretical analysis of our polarized x-ray absorption spectra, carried out as part of a collaboration with G.A.Sawatzky's group, led to the first major new development, which was the finding that a more realistic many-band description is not just a detail, but is essential to the physics. Specifically, it was found that the V^{3+} spin state is $S=1$, impossible in a one-band model, and that the changing occupations of the V 3d states through the transition involved all of the three lowest d-states split off by the material's crystal field. These results gave the essential experimental support for a similar conclusion reached from LDA+U calculations done at the time in Sawatzky's group. There was then a clear need for a many-body theory applied to a multi-band Hubbard model realistic for V_2O_3 .

The next major development coming from our work is more recent and involves the valence band photoemission spectrum. The first multi-band many body theory has been given by Vollhardt's group using a scheme for combining the dynamic mean field theory (DMFT) with the local density approximation (LDA) of band theory. DMFT is a new development of many-body theory of the early 90's, Vollhardt being one of its inventors, and in application to the Hubbard model it is significant as the best description that can be made using a k -independent self energy. It may be formulated as a mapping of the lattice problem onto an effective Anderson impurity model coupled self consistently to an effective conduction band bath. In the metallic phase, although the large Hubbard U value acts to separate much of the bands' spectral weight away from E_F into the so called upper Hubbard band (UHB) and lower Hubbard band (LHB) there remains at E_F a distinctive quasi-particle (QP) peak, not accidentally reminiscent of the Kondo/Suhl-Abrikosov resonance of the Anderson impurity model (that was first observed by the PI and his collaborators in work predating this grant, and that has been further studied under this grant). The LDA + DMFT calculations yield detailed predictions for the lower Hubbard band and QP peak, which should be observable in the valence band photoemission spectrum. These predictions in turn demand equally detailed and accurate experimental results to which the

theory can be compared. In our most recent work we have obtained such spectra and made such comparisons, in collaboration with Vollhardt's group.

Our early work on the NSLS Dragon beamline at the higher photon energies, for which the photoemission probe depth is larger than for lower photon energies, had given strong indications that spectra obtained at lower photon energies contain a large component originating from a more strongly correlated surface region. But the resolution available at the higher photon energies was insufficient for detailed studies of the metal-insulator transition. In the late '90's we formed a collaboration with S. Suga to use his new high resolution high photon energy beamline at SPring8 to continue our seminal studies of the V_2O_3 system. In addition, because our exploratory experiments at the ALS spectromicroscopy beamline 7.0 showed the importance of using a small photon spot (100 μm diameter) to minimize the effects of surface steps and roughness, such a small spot was also used at SPring8. We have thereby obtained the first well resolved high photon energy (and therefore very bulk sensitive) photoemission spectra for all phases of $(V_{1-x}Cr_x)_2O_3$. These spectra have yielded the first observation of a prominent QP peak in the metallic phase of V_2O_3 , qualitatively consistent with the QP peak of new LDA + DMFT calculations performed for a temperature comparable to that of the experiments. But detailed comparison shows that the experimental peak width and weight are significantly larger in the experimental spectrum than in the theory. These results take the famous V_2O_3 problem to an entirely new level of sophistication for both theory and experiment and set the stage for future studies of the QP peak and its changes through the phase transition. [Pub #'s 25, 45, 66, 67]

B. Basic issues in superconducting cuprates

1. *Non-rigid band behavior and anomalous chemical potential position with doping in $Nd_{2-x}Ce_xCuO_4$ and $La_{2-x}Sr_xCuO_4$*

The two cuprate superconductor systems $Nd_{2-x}Ce_xCuO_4$ and $La_{2-x}Sr_xCuO_4$ are electron and hole doped, respectively. Our work on these two systems in the early 90's led us to pose a fundamental question that continues to this very day, the issue being the evolution with x of the chemical potential. One has an insulator for $x=0$ and the least energy to add a hole or an electron is then, by definition of the meaning of the single particle spectrum, just the states that define the insulating gap. In the simplest rigid band picture one then expects that, with deliberate doping, added holes or electrons will cause the chemical potential (E_F for $T=0$, but more generally to be denoted as μ) to jump from its position for $x=0$ someplace in the insulator gap to some point in the valence or conduction spectral weight, below and above the gap, respectively. In passing from hole doping to electron doping one expects μ to jump the gap. We obtained spectra, initially from PES and later from angle resolved photo emission spectroscopy (ARPES), that caused us to propose that the chemical potential did make such a jump, but instead that it departed from rigid band behavior and remained in the insulator gap in new spectral weight that appeared in the gap as x increased from 0 and evolved with x in a non-trivial way. The ARPES work was for $Nd_{2-x}Ce_xCuO_4$, showing that the spectral weight at E_F dispersed with k to define a Fermi surface in good basic agreement with predictions from the LDA, important because the growing spectral weight therefore could not be attributed to a simple impurity band in the gap.

Our proposal was very controversial at the time, widely disbelieved, and even dismissed by many workers. Subsequently various theories have offered scenarios in which such an anomalous dependence of μ with x could occur and in the late 90's the issue has been taken up again by two major cuprate photoemission groups, that of Z.-X. Shen and A. Fujimori, and also by many theorists and other workers. That there is not rigid band behavior in any doped cuprate system is now undisputed and it has been reported by Fujimori, working with Shen, that for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ μ indeed remains in the gap in new spectral weight that grows with x . For $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ Shen has reported data showing that μ jumps to the conduction band but that the system's spectra evolve with x in a highly nontrivial way. It seems likely that this issue is not yet settled and further, it seems fair to say that the work done in the early period of this grant can be credited with having raised a continuing issue now acknowledged by all to be of fundamental importance and entirely non-trivial. [Pub #'s 5, 15, 25].

2. $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_4$

Pr in this system acts initially to quench superconductivity. It is exceptional in this respect because other rare earths doped into this system, except for Ce and Tb, which have the drastic effect of inducing a new crystal structure, have no effect on the superconductivity. For large enough x the material becomes an insulator. In the late '80's we performed PES and RESPES and other spectroscopic studies of this system using ceramic samples. We found that it is very difficult to describe its Pr 4f spectra within the framework of the Anderson impurity model, surprising to us because of the general success of that model for the 4f electrons of Ce materials and also for other Pr materials. In continuing studies we found it difficult to obtain reproducible data from ceramic samples. We discontinued further work on this system and experimental work on cuprates in general, in favor of taking up a different approach to the general problem of the cuprates, as described next.

3. *Alternate general approach to the cuprate problem*

In the early 90's several groups worldwide developed vigorous programs of using PES and ARPES to measure cuprates. We looked for an alternate research direction that would nonetheless address fundamental issues raised by these materials. Three early ideas that remain vital after many years of cuprate research are (1) that the quasi-two-dimensionality is of fundamental importance, (2) that power law tails of ARPES lineshapes above the superconducting transition temperature T_c signal non-Fermi liquid (NFL) behavior that is also fundamental to the system, and (3) that items (1) and (2) are closely linked. Specifically, one of the most interesting ideas came from P.W. Anderson, that the NFL behavior involves electron fractionalization and that such fractionalization is actually of broad general importance in condensed matter systems. Initially Anderson proposed fractionalization for the cuprates within the framework of his "resonant valence bond" (RVB) picture and subsequently within the framework of the Luttinger liquid (LL) behavior of a one-dimensional interacting electron gas, but extended to two dimensions. The latter idea has proved to be theoretically very contentious and in recent years Anderson has returned to the RVB picture, but all the while maintaining the basic idea of fractionalization. To us the general importance of the failure of the Fermi liquid theory, of the idea of electron fractionalization as a mechanism for this failure, and of the

fascinating properties of the LL, appeared to be of even more interest and possibly more fundamental character than the superconductivity of the cuprates.

So we embarked upon a program of analyzing cuprate ARPES lineshapes and of studying selected non-cuprate low dimensional systems' ARPES lineshapes, with a focus on NFL behavior from the fractionalization viewpoint. Our early analysis of the NFL power law tails of cuprate lineshapes showed that these tails almost certainly could not be attributed to some extrinsic mechanism like inelastic electron scattering, and therefore subtracted out, but that essentially all the ARPES spectral weight must be regarded as an intrinsic part of the lineshape. Our ARPES work on non-cuprate systems is described in the next section. In our opinion it remains very possible that this line of research will turn out to be as relevant to the ultimate understanding of the cuprates as are the many beautiful results that have been obtained by various workers doing ARPES studies of the cuprates themselves. The theoretical work of Kivelson and Emery and their collaborators has also been very influential on us in developing and sustaining this line of research, especially in recent years. We bring out the relevance to the cuprates of our work on low dimensional materials in sub-section C.5 below. [Pub # 1, 68].

C. Distinguishing Fermi liquids and non-Fermi Liquids in Low-dimensional materials

1. *Establishment of TiTe_2 as an ARPES Fermi liquid paradigm*

In our cuprate ARPES lineshape fitting efforts the need to calibrate ARPES for a Fermi liquid material became obvious. In the early '90's, with strong input from Ralph Claessen, then a postdoc in the group, we proposed TiTe_2 as an ARPES FL paradigm. Over the years, the material has been accepted by the ARPES community as playing exactly that role. It has been studied in detail by many groups other than ourselves and continues to be studied as the ARPES capabilities for energy and \mathbf{k} resolution continue to improve. The material is special for having a non-degenerate Ti 3d band that has an E_F crossing at a place in \mathbf{k} -space where it is isolated from all other bands and so can be studied un-obscured by emission from other states. The ARPES lineshape is immediately significant relative to cuprate spectra because it lacks the NFL power law tails that occur for cuprate spectra. Although it is now known that the material is more three-dimensional than originally thought, nonetheless it is sufficiently two-dimensional that with careful selection of the photon energy one can minimize the photoelectron lifetime contribution to the ARPES linewidth that, for three-dimensional materials, gives one of the contributions whereby the ARPES lineshape differs from the single particle spectral function of many-body theory. ARPES work on this material has shown its basic Fermi liquid character but has also stimulated more sophisticated modeling of its spectral function and single particle self energy. Most recently we have focused on the self energy's \mathbf{k} -dependence by analyzing a new set of ARPES data taken at the ALS. [Pub #'s 8, 23, 24, 25, 26, 29, 52, 66].

2. *SmTe_3 as an ARPES paradigm of high temperature CDW formation*

An aspect of low dimensional materials is their strong tendency to have the special Fermi surface geometry known as nesting, which leads to gap formation that stabilizes a spin or charge density wave (SDW or CDW). SDW or CDW formation is competitive with LL formation in quasi-low-dimensional materials and fluctuations toward a CDW or SDW can produce spectral

effects that mimic in some ways the characteristic features of the LL spectrum. Thus it is important for our program to have and study a CDW paradigm. Nesting and gap formation are crucial parts of CDW theory which have gone unstudied for lack of a material with sufficiently large gaps and sufficiently stable surfaces for detailed ARPES studies of the kind which have been done for superconducting gaps in high T_C cuprates. Working in collaboration with UM colleagues Meigan Aronson and Stephen Lee, we overcame these problems in quasi-2-d SmTe_3 , a very novel material with an incommensurate CDW that appears to be stable up to the crystal's melting temperature. The crystal structure features square planar Te sheets, the electronic structure of which, particularly the FS, appears to be remarkably well described by a simple tight binding band theory and is in excellent agreement with the experimental FS features. Although the shape of the observed pattern has the four-fold symmetry of the calculation, its intensity distribution has two-fold symmetry. Parts of the pattern are dim because the CDW induces E_F gaps in one direction. The FS displays imperfect nesting, with vectors distributed around the incommensurate CDW wavevector Q known from electron diffraction. The CDW gap has been tracked over the entire FS. It is variable, but, in quite beautiful agreement with basic theory ideas of nesting, non-zero only for the nested portions. The mean field condition determining Q for imperfect nesting is found to be obeyed within 4%. A PRL referee described the work as "almost a textbook case deserving rapid dissemination." [Pub # 37].

3. *1-d CDW material $(\text{TaSe}_4)_2\text{I}$*

$(\text{TaSe}_4)_2\text{I}$ is a quasi-one-dimensional material with a low temperature CDW. Other workers (Terrasi et al) reported that for the electric vector polarized perpendicular to the one-d axis a new dispersing excitation is seen which is anticyclic in k relative to that observed for parallel polarization. This remarkable finding, if true would require a complete re-working of the usual CDW theory for this material. In collaboration with Stefan H fner and Ralph Claessen (Saarbr cken) experiments done both at the synchrotron and in the Saarbr cken laboratory were made to check this remarkable finding. It was found always that the spectra in both polarizations are the same. [Pub # 35].

4. *Dimensional crossover in 1-d CDW material $\text{K}_{0.3}\text{MoO}_3$*

We made ARPES studies of the quasi-1d blue bronze material $\text{K}_{0.3}\text{MoO}_3$, in collaboration with Ralph Claessen, in the time period after his postdoc with us had ended and he had joined the group of Stefan H fner in Saarbr cken. $\text{K}_{0.3}\text{MoO}_3$ is a CDW material and the tight-binding band structure calculations of Whangbo show it to have two bands dispersing to cross E_F along the quasi-one-dimensional b-axis. A previous ARPES study by other workers had found a single dispersing peak in the metallic phase above T_{CDW} , the CDW transition temperature. In our initial work in the mid '90's we found two dispersing peaks along the quasi 1-d axis in the high temperature phase. Their dispersions are about two times greater than in the tight binding band calculations, a difference we have found also for other bronzes and which seems to be generic to the tight banding calculations of Whangbo for these materials. We also showed experimentally that these peaks have essentially no dispersion in the two directions perpendicular to the b-axis. In the early part of his thesis work, Gey-Hong Gweon then made an ARPES Fermi surface map showing that the two pieces of Fermi surface, one from each band, are separated, i.e. nested, by

exactly the same wave vector measured for the low temperature CDW by x-ray scattering, as expected in the nesting model of CDW formation.

In his thesis work, Gey-Hong Gweon also addressed the possibility of LL behavior in the metallic phase. At the time we began our work on non-cuprate low-d materials, it had just been reported by Y. Baer's group that the angle integrated photoemission spectra of several quasi-1d metals shows, instead of a Fermi edge, a power law onset. $K_{0.3}MoO_3$ is one of the metals they studied. Such behavior is a theoretical prediction for the LL. However a strong suppression of E_F weight, i.e. a pseudogap, can also occur due to fluctuations to CDW order and CDW gap formation above T_{CDW} and so the PES result is rendered ambiguous as to LL formation. We first reproduced the power law vanishing E_F weight in the angle integrated photoemission spectrum. We then analyzed the ARPES lineshapes and found that they are described neither by fluctuating CDW models or by LL models. It seems almost certain that a model including both the Luttinger and the CDW interactions is necessary, e.g. the Luther Emery (LE) model.

There is recently a theory by Kivelson and his co-workers for the LE model, treating the 1d to 3d crossover behavior expected across a phase transition in a quasi-1d system. A significant result of the theory is the finding the spectral peaks do not show mean field shifts but rather that spectral weight re-distribution occurs, involving the appearance of a quasi-particle peak in the ordered phase. It was argued that this is an explanation of the sharp peak appearing in cuprate spectra below the superconducting transition. Although the theory was done specifically for a transition to superconductivity, according to Kivelson the results should be similar for a CDW transition. We have also made ARPES and angle-integrated measurements across the blue bronze CDW transition. We reproduce Baer's previous findings of a nominally BCS-like gap opening in the angle-integrated spectrum, which might lead one to expect mean field like peak shifts. However in the ARPES spectra we find no shifts of peaks, but rather spectral weight shifts at spectral edges, somewhat like in the theory of Kivelson. But we do not see a sharp quasi-particle peak. It remains to see if the theory can be used to estimate the spectral weight of the peak, to determine if perhaps a small weight would preclude its observation. [Pub #'s 27, 32, 52].

4. *Establishment of quasi-1d $Li_{0.9}Mo_6O_{17}$ as an ARPES paradigm of electron fractionalization in the Luttinger liquid*

Starting in the mid '90's and continuing to the present we have made a series of ARPES studies that establish $Li_{0.9}Mo_6O_{17}$, also known as the Li-purple bronze, as an ARPES LL paradigm. The major contributors have been Gey-Hong Gweon during the time of his thesis work and later as a postdoc in the group, and Jonathan Denlinger during his postdoc period with the group. $Li_{0.9}Mo_6O_{17}$ is a quasi-1-d metal that displays a phase transition of unknown character at $T_X \approx 24$ K and superconductivity below 1.9 K. Below T_X its optical and magnetic properties show no CDW or SDW gap opening and repeated x-ray diffraction studies show no CDW or SDW. The PES spectrum shows a NFL power law onset above T_X . As T is decreased through T_X the NFL behavior persists, albeit with some sharpening, and no gap is observed. Band calculations by Whangbo show four bands, two disperse to become degenerate just before crossing E_F and two that do not cross E_F . By ARPES we have found all four bands essentially as predicted

theoretically but with bandwidth roughly twice as large and we have mapped the FS in three dimensions and shown it to be highly one-dimensional.

The dispersing ARPES lineshapes that define the Fermi surface consist of an edge and a peak that disperse with different velocities. This behavior is generally well described by theoretical lineshapes for the LL, which have two dispersing features in contrast to the single dispersing peak of the Fermi liquid. The origin of the two features is the fractionalization of the electron in the LL. There are no quasi-particle excitations but only collective density waves of spin and charge known respectively as spinons and holons and having different velocities. Removing an electron results in the excitation of holons and spinons with momenta and energies as needed to make up the momentum and energy of the electron removed and gives the two featured lineshape. The lowest energy feature is usually a spinon and has an edge or a peak character depending on whether the so-called "anomalous dimension" α is greater or less than 0.5. α can be measured from the PES E_F onset power law and also controls other aspects of the k -dependence of the ARPES lineshape. All determinations find α for the Li purple bronze to be greater than 0.5, consistent with dispersing edges. Over time we have obtained data with better and better angle and energy resolution, and found that the improved data showed worsened agreement with the $T=0$ theory that was available at the time of our initial work. However, in this same time period Orgad has used the ideas of quantum criticality to calculate LL lineshapes for non-zero temperature and we find that the new T -dependent theory gives a good description of our best ARPES data with a holon to spinon velocity ratio of 2 and $\alpha = 0.9$. As a result of our work there is much developing interest in this material and other workers, using different experimental methods such as scanning tunneling microscopy (STM), muon spin rotation and relaxation, and nuclear magnetic resonance are seeking to verify our LL interpretation. The STM work has been stimulated by Kivelson and his collaborators, who have calculated the tunneling spectra for the LL model using the parameters we have determined with ARPES.

Most recently we have measured spectra for samples prepared by a different method than was used for the samples studied in all our previous work. The new samples come from the Mandrus group at the Oak Ridge National Lab and are prepared by a temperature gradient flux technique, whereas our previous samples came from Marcus and Schlenker in Grenoble and were prepared by an electrolyte reduction technique. We find that spectra from the two kinds of samples are the same, which dispels any possibility that large differences between ARPES spectra reported another group for temperature gradient flux samples of this material and our ARPES spectra for electrolyte reduction samples stems from the different sample preparation methods. These differences included the claim of Fermi liquid behavior and a large low temperature gap, findings that are inconsistent with the results of the transport measurements cited above, some of which are now re-confirmed by measurements in the Mandrus group. At the time of writing this report a short paper reporting the new data from the Mandrus samples is in preparation. [Pub #'s 26, 32, 43, 49, 52, 56, 66, 68].

5. *Non-Fermi liquid fractionalization behavior of the 2-d bronzes $\text{NaMo}_6\text{O}_{17}$ and $\text{KMo}_6\text{O}_{17}$*

The molybdenum bronzes $\text{AMo}_6\text{O}_{17}$ ($A=\text{Na}, \text{K}$, the “K- or Na-purple bronzes”) are quasi-2d CDW materials. They are especially important for our work because they display “hidden one-dimensionality” arising from the presence in 2d layers of three weakly coupled 1d chains oriented mutually at 120° to each other. They are thus bridging materials between 1d and 2d. ARPES Fermi surface intensity maps obtained by Gey-Hong Gweon above their CDW transition temperatures show for both purple bronzes the “star of David” topology predicted theoretically for the three weakly coupled 1-d bands, in disagreement with the much different topology reported in PRL in a previous ARPES study of $A=\text{Na}$ by other workers. Most recently, the “star of David” shape has even been imaged across multiple Brillouin zones in our work at the ALS. The nesting vector inferred from the maps agrees perfectly with the CDW q -vector found in diffraction measurements.

In spite of the good agreement with band theory and the basic CDW nesting theory, the ARPES lineshapes above T_{CDW} have a definite NFL character that can be explained within a fractionalization picture. First, the PES E_F onset is much better described by the finite temperature LL theory of Orgad, with $\alpha = 0.3$, than by a Fermi edge. This is the first quasi-2d material to show non-zero α , $\alpha=0$ being a Fermi edge. Second the ARPES spectra show both the dispersing peak that defines the Fermi surface and a k -independent part that is manifestly decoupled from the dispersing part. This divided character of the spectra cannot be understood as extrinsic, e.g. due to inelastically scattered photoelectrons in the experiment, because the k -independent part is perfectly confined to the electronic bandwidth, which is well isolated in energy from the oxygen states. Further one cannot simply invoke k -smearing due to the known charge disorder of the material because one has also dispersing peaks that give a well defined Fermi surface. We advance a hypothesis of electron fractionalization, with the k -smeared part due to holons badly scattered by the charge disorder and the dispersing part due to spinons unscattered by the charge disorder. The fact that one has in this material dispersing peaks instead of the edges seen in the Li purple bronze is nicely consistent with the α value found from the angle integrated spectrum being less than 0.5 in this material. This explanation is admittedly speculative but has great intuitive appeal within a fractionalization picture. We gratefully acknowledge that the idea was given to us by Phil Anderson, in the context of an explanation for the NFL power law “background” of cuprate ARPES spectra. Our most recent paper on this topic develops the amazing similarity of the NFL aspects of cuprate spectra and quasi-2d purple bronze spectra and argues that the latter provides a way to recognize in cuprate spectra the effects of charge disorder on fractionalization. [Pub #'s 32, 33, 44, 66, 68]

D. 4f materials displaying Kondo, heavy Fermion, and mixed valence properties described by the Anderson impurity and Anderson lattice models

1. *Kondo resonance and surface effects in Yb compounds*

Kondo resonance in YbAl_3 Yb is the one hole analog of Ce and so, from the viewpoint of the impurity Anderson model, the Yb electron-removal spectrum should be like the Ce electron addition spectrum. For Ce the addition spectrum contains the main part of the Kondo resonance and cannot be measured with high resolution because the resolution in BIS is rather poor. Thus Yb offers the possibility to exploit the much higher resolution of photoemission to study the lineshape and temperature dependence of main part of the Kondo resonance. We measured the

4f lineshape at 20K in YbAl₃ at the SRC Ames-Montana beamline, and, in collaboration with L. H. Tjeng and S.-J. Oh, measured the temperature dependence of the lineshape on the NSLS Dragon beamline. We found results in agreement with predictions of the impurity Anderson model. These results still stand today as the best example of the Kondo resonance in an Yb material. Studies of other Yb materials have been plagued by large surface effects such that there is a large difference in Yb valence and Kondo temperature between the bulk and the surface material probed in various electron spectroscopies, as described next. [Pub #'s 6, 14, 17, 19, 20]

Large valence change in a thick near-surface buffer layer of YbInCu₄ Yb offers the possibility to exploit the much higher resolution of photoemission to study the main part of the Kondo resonance. The drawback is that Yb materials always display prominent surface shifted peaks from which the bulk spectrum of interest must be disentangled. Often, with the prominent exception of our work on polycrystalline YbAl₃, the width and weight of the "bulk" spectrum so inferred has not shown very good agreement, in the framework of the impurity Anderson model, with bulk thermodynamic properties. Curiously, for YbAl₃, a problem was found to occur for cleaved single crystal surfaces, where rather broad lines were observed, but not for polycrystalline surfaces. It is clear that there exists what is now known in the correlated electron community as "the Yb surface problem." In work with collaborators we have made progress on this problem.

In experiments begun in collaboration with us at SSRL and then continued at BESSY, our collaborators F. Reinert, R. Claessen and S. Hüfner (Saarbrücken) have measured photoemission on single crystals of YbInCu₄, a material which exhibits at ≈ 60 K a first-order valence transition with some resemblance to the alpha-gamma transition in Ce. The "bulk" Yb peaks display a time dependent decay after cleaving, with time constant that depends on the temperature and suggests an Arrhenius model for relaxation, with a characteristic energy of a few meV. The valence deduced just after the cleave is initially far from the bulk value inferred from transport measurements and relaxes toward the bulk value, but never achieves the bulk value even at long times. Perhaps most significant is that the valence shows no abrupt change at the 60K-transition temperature, in contrast with X-ray absorption spectroscopy (XAS), which does show an abrupt change. It seems nearly certain that there must be a near-surface region which "buffers" the mismatch between the surface valence and the bulk valence, which makes a substantial contribution to the "bulk" peaks, and which experiences some atomic scale relaxation with a small activation energy. This buffer layer would have a much larger Kondo temperature and would not display the bulk phase transition. This general picture was put forth in the early 90's by J. Lawrence (UC Irvine) to explain his findings that a number of Yb materials for which photoemission spectra showed disagreement with bulk properties within the framework of the impurity Anderson model, nonetheless showed XAS spectra in fair agreement with the model expectations. [Pub #'s 36, 53]

2. XMCD in ferromagnetic Ce compounds

CeFe₂ and CeRh₃B₂ are unusual cerium materials in being ferromagnetics. Also the observed magnetic moments are much reduced from the free ion Ce³⁺ value. The ferromagnetism can be studied by measuring x-ray magnetic circular dichroism (XMCD) at the Ce 3d edge. During the period that L.H. Tjeng was a postdoc in the group, we made such measurements at the NSLS on the Dragon beamline. In CeFe₂ we found evidence for a magnetic moment on the Ce site. In contrast the 4f photoemission and BIS spectra obtained in our group some time ago implies the presence of the Kondo effect with a very large value of the Kondo temperature T_K , much larger than the Curie temperature. The tension between the Kondo effect and the tendency toward magnetic ordering is the essence of the Kondo lattice problem. In CeFe₂ one could imagine that the Kondo effect is responsible for the small Ce moment, but that it is unable to overcome the tendency to ferromagnetism due to the Fe atoms. A likely scenario is that the ferromagnetism forms in a Kondo Fermi liquid established at the much higher Kondo temperature. The quasi-particles of this Fermi liquid would involve mixing of the Fe 3d and Ce 4f, both of which would then be polarized in the ferromagnetic state. We also measured the ferromagnets CeCo₂ and CeCo₅.

3. *Heavy Fermion/Kondo Ce materials*

Detailed analysis of electron spectroscopy and the Kondo volume collapse model for the α - γ transition of Ce metal I. In collaboration with O. Gunnarsson, N.E. Christensen and O.K. Andersen, impurity Anderson Hamiltonian parameters were obtained for the α and γ phases of Ce metal by a detailed analysis and comparison with the published electron spectroscopy data, using the 1/N expansion theory with energy-dependent hybridization calculated in the local-spin-density-functional approximation. With the surface contribution included, the theory provides a remarkably good description of various spectroscopic data with the same set of Hamiltonian parameters for all spectroscopies. The calculated susceptibility for these parameters is in good agreement with the experimental value. A previous result of analyzing the valence-band spectrum gave a susceptibility value that is much too large compared with the experimental one. We showed that this discrepancy is caused by ignoring the surface emission in the spectral analysis. [Pub #'s 4, 11, 48, 50, 66].

Detailed analysis of electron spectroscopy and the Kondo volume collapse model for the α - γ transition of Ce metal II. The Kondo volume-collapse (KVC) model of the α - γ transition in Ce metal was examined quantitatively using Anderson impurity Hamiltonian parameters obtained from the electron spectroscopy analysis described in the preceding paragraph. After the hybridization from spectroscopy is scaled by 1.12 to reproduce exactly the experimental zero-temperature susceptibility, the calculation, with no further adjustable parameters, predicts a phase boundary in good agreement with experiment. It was found that the cohesive energy contribution from the hybridization of un-occupied f states and conduction states is quantitatively important in Ce, with a value much larger than the Kondo energy. This contribution is equally large and important for La and Pr, for which the hybridization is often ignored. It is however almost spin independent, so that it does not contribute directly to the Kondo energy. Thus the 4f cohesive energy contribution is large in both the α and γ phases, while only the Kondo spin fluctuation energy (and entropy) causes the α - γ transition. This distinguishes the KVC model from the Mott transition model. The Anderson-Hamiltonian-based

KVC model is also distinguished from the the Kondo-Hamiltonian based spin-only version of the KVC model in that the latter approach cannot make direct contact with spectroscopic data because charge degrees of freedom are ignored from the outset. This work provides quantitative confirmation that a unified understanding of the high-energy spectroscopic and low-energy thermodynamic properties of Ce metal has been achieved at the quantitative level. [Pub # 7, 11, 48, 50, 66].

Impurity Anderson Hamiltonian successful for angle integrated PES spectra of Ce 4f states of CeSi₂ CeSi₂ has served as a focus of debate about the applicability of the impurity Anderson model for describing Ce 4f spectra. In the early '90's, with major help from UM undergraduates, we made a detailed analysis of all the then available CeSi₂ spectra from Ce 4f photoemission and BIS, and Ce 3d photoemission, using the 1/N expansion theory for the impurity Anderson Hamiltonian and an energy dependent hybridization calculated from the LDA. An excellent unified description of all the spectra was obtained by allowing for the possibility of different surface and bulk model parameters, resulting in a prediction of a surprisingly large difference in the bulk and surface 4f photoemission spectra. We measured at the NSLS Ce 4f RESPES at the Ce 3d edge, giving much greater bulk sensitivity relative to RESPES at the 4d edge. Analysis of this data shows that the predicted large difference actually occurs. The result is a unified analysis of all the bulk spectra with parameters that also yield the Kondo temperature known from transport studies. [Pub # 48].

Direct experimental verification of applicability of single-site model for angle integrated PES spectra of small T_K concentrated cerium compounds—bulk sensitive resonant 3d-edge photoemission of (Ce_{1-x}La_x)Al₂ The single-impurity model Anderson model (SIAM) has been used with great success to describe the angle integrated 4f photoemission spectra of dense-Kondo Ce compounds, both the low energy Kondo resonance features and the higher energy ionization excitations, after proper subtraction of a surface spectrum. This success contrasts with the Nozieres' "exhaustion" idea, that in the simple Kondo lattice there are not enough conduction electrons to screen out the magnetic moments as in the SIAM, essentially because only electrons located within the Kondo temperature of E_F can take part in the Kondo screening. The exhaustion problem is thus must serious in small T_K systems and would imply a decreased T_K in the lattice system relative to the impurity system. In this work, in collaboration with M.B. Maple and S. Suga, we made a direct experimental comparison of the 4f spectra of concentrated and dilute small T_K Ce materials (Ce_{1-x}La_x)Al₂ by performing bulk-sensitive high-resolution resonant 4f photoemission with photon energies at the Ce 3d edge. The work was done on Suga's beamline at the SPring8 synchrotron in Japan. It is important for obtaining spectra in the extreme dilution sample that the resonant contrast at the 3d edge is extremely large. The Laves phase structure of CeAl₂ has only 4 near Ce neighbors of a Ce site, so the for x=0.04 85% of the Ce ions are completely isolated from other Ce ions if next-nearest Ce-Ce couplings can be neglected, so the spectrum is essentially that of a single ion. The spectra are nearly x-independent, giving a theory-independent proof of the applicability of an impurity model ansatz for angle integrated Ce systems. The known reduction of T_K upon dilution, due to volume expansion, is opposite to that expected in the "exhaustion" idea, but easily explains the small reduction of the overall near-E_F intensity within a SIAM description, including the stronger decrease of the nearest E_F weight relative to the spin-orbit sideband at 0.25 eV. We also obtained data for a high T_K system (La_{1-x}Ce_x)Ru₂. These data are complicated by a shift of E_F

with x due to the differing valences of Ce (3.3) and La (3.0), and the data are not yet fully analyzed. [Pub # 60]

f-electron Fermi surface exclusion above the Kondo temperature in CeRu₂Si₂ Going beyond the Anderson impurity model description of the angle-integrated 4f spectrum entails the interplay of Kondo physics with solid state Fermi surface properties. The Luttinger Fermi surface sum rule (counting theorem) of many-body field theory insists that, whatever the route to a FL, if their magnetic moments are quenched, then the f-electrons must be counted in the Fermi surface (FS) volume. An important theoretical conjecture concerning the Kondo lattice ansatz, made by Zwicknagl and Fulde, is that above T_K where the f-electron moment is no longer quenched (as revealed by a high temperature Curie-like T-dependent magnetic susceptibility, and also neutron scattering spectra), the f-electrons should now be excluded from the FS volume. This idea is plausible but there is no proof having the rigor of the Luttinger theorem and in any case it would not hold experimentally unless Kondo physics is indeed involved in heavy Fermion properties.

The literature paradigm for low temperature f-electron inclusion in the FS volume comes from beautiful FS studies in the early 90's of the isostructural compounds LaRu₂Si₂, CeRu₂Si₂, and CeRu₂Ge₂, using magneto-oscillatory (MO) techniques such as the de Haas-van Alphen (dHvA) effect. The LDA FS of LaRu₂Si₂, which has no f-electrons, has several pieces, including a large hole pocket around the X-point of the Brillouin zone and a multiply connected electron sheet centered around Γ . The LDA bandstructure for CeRu₂Si₂ includes the 4f electron of the Ce³⁺ ion in the FS. The electron is accommodated about 50% each in the large hole pocket, which then shrinks to become the so called "pillow" of CeRu₂Si₂, and the multiply connected electron sheet which expands. It is found that dHvA data shows all orbits expected with the renormalized LDA for CeRu₂Si₂, and shows the expected size differences for La and Ce systems. Further, CeRu₂Si₂ shows a metamagnetic transition under high magnetic fields and CeRu₂Ge₂ is a ferromagnet. The dHvA experiments find that in these two situations where the f-moment is restored, the measured FS is like that of LaRu₂Si₂, again showing that the Luttinger theorem is obeyed.

MO techniques do not provide a global view of the energy and k -dependence of the electronic structure, and so are limited for distinguishing among different many-body scenarios of heavy mass formation. MO techniques are also limited to low temperature and so have been silent about the crucial question of the FS volume above T_K . ARPES in principle provides this global view and is more easily done at high temperature than low temperature. The exciting finding of f-electron FS exclusion above T_K emerged from our CeRu₂Si₂ ARPES data. Our ARPES FS map of LaRu₂Si₂ shows the large X-point FS hole pocket in almost perfect agreement with the LDA prediction and with dHvA data. A careful comparison of detailed photoemission Fermi surface maps for the dHvA paradigm compound CeRu₂Si₂ above its Kondo temperature to those for LaRu₂Si₂ shows them to have near identical X-point hole pockets, and the so called "pillow" plus other predicted LDA features near the X-point are not observed. Since the number of conduction electrons for the La and Ce compounds are the same if the Ce 4f electrons do not contribute to the FS volume, we conclude that we have strong evidence of f-electron FS exclusion for T well above T_K , consistent with the Fulde/Zwicknagl conjecture. This work takes the study of k -dependence in heavy fermion systems to a new level. [Pub #'s 51, 59, 66].

4. *Mixed Valence in SmB₆*

For mixed valent SmB₆ we extended our early (1980) room-temperature photoemission studies made at SSRL with moderate resolution on polycrystalline samples to much higher resolution RESPES and ARPES studies of single crystals at ~20K. By RESPES, variable take-off angle and variable photon energy studies, we found for the Sm²⁺ 4f states near E_F (a) a surface shift of ~0.5 eV, also observed in earlier work, and (b) three pieces of fine structure in the unshifted peak, only two of which can be assigned as previously unresolved multiplet splittings of the 4f⁵ final state. The near-E_F 3-peak structure is shown in Fig. 5. The low photon energy data from the SRC has better resolution, but shows the highest energy peak hardly at all, while the high energy RESPES data from the ALS gives the crucial extra information that there are indeed three peaks. The ~180 meV separation of the two lowest energy peaks cannot be ascribed to crystal field splittings. We also observe in ARPES a broad peak that disperses across E_F and enables a determination of the "non-4f FS" in fair agreement with expectations from band theory, as described further in the section F below. [Pub # 47]

E. Electronic structure of 5f electron materials

1. *Lack of surface effects and 4d edge RESPES in UAl₂*

For Ce systems, it has been documented in the past five years or so that surface shifts of the 4f binding energy have important quantitative effects on the 4f spectra, especially in cases of strong hybridization. One might expect similar effects for the 5f spectra of uranium systems. The most powerful spectroscopy for studying these effects in Ce systems has been the comparison of RESPES spectra taken at the Ce 4d edge with those taken at the Ce 3d edge, because the larger kinetic energy of the latter photoelectrons leads to much greater bulk sensitivity in the spectra. Therefore we have tried to make analogous RESPES studies at the U 4d edge to compare to those at the 5d edge. To our surprise no resonant enhancement of the 5f emission occurs at the 4d edge. We rationalize this result by supposing that Auger channels involving the 5d core level dominate. UAl₂ was chosen for these studies because the weakness of the Al emission makes it possible to observe the U 5f states easily without the resonant effect. We measured the 5f emission for several photon energies between 40 eV and 1400 eV and find that the only change observed in the spectra can be fully ascribed to instrumental broadening, which increases as the photon energy increases. If this system is typical it appears that surface effects will not play a major role in obtaining a general understanding of U 5f spectra. [Pub. # 30]

2. *Anderson impurity model analysis of Pu 4f core level spectra*

Pu displays a complex phase diagram with α , β , γ , and δ phases which involve changes in the 5f electronic structure. Contrary to expectations within a traditional band picture, even though the volume per Pu atom increases considerably in the sequence, the measured 5f valence band width also increases considerably in the sequence. L.E. Cox at Los Alamos has measured the XPS 4f core level spectra of all four phases and finds a systematic change in the sequence of phases that

is generally similar to that observed in the Ce α - γ transition. In a collaboration with Cox and J.M. Peek at Los Alamos, we analyzed the 4f spectra within the framework of an impurity Anderson model of the kind that is successful for Ce. Very good fits of the spectra can be achieved, and Anderson model parameters thus obtained. [Pub # 42]

3. *Seminal Discovery of non-Fermi liquid behavior in $Y_{1-x}(U_yTh_{1-y})_xPd_3$.*

In the early '90's our collaboration with M.B. Maple's group at the University of California – San Diego (UCSD) led to the discovery of non-Fermi liquid behavior in $Y_{1-x}U_xPd_3$ and thereby opened up an entire new research topic that continues to be pursued both experimentally and theoretically by many groups. Our initial spectroscopic work on $Y_{1-x}U_xPd_3$ found that the Fermi level can be tuned about 1 eV with x, explained by us as arising from replacement of U^{4+} by Y^{3+} . Spectroscopy also showed that the variation of E_F tunes the 5f ionization energy ϵ_f , which decreases as x decreases. In Kondo models, the Kondo temperature T_K should increase rapidly as ϵ_f decreases, which motivated transport and additional spectroscopic studies. Further, D.L. Cox of Ohio State University (OSU) showed that for U^{4+} , a Kondo quenching of quadrupoles is possible, leading to non-Fermi liquid behavior as proposed by Nozieres and Blandin. The UCSD transport measurements showed all these things - T_K increasing as x decreases, with the appearance of signatures of the quadrupolar Kondo effect and non-Fermi liquid behavior. Spectroscopically we verified E_F tuning and a Kondo resonance-like growth of 5f spectral weight with decreasing x near E_F in the 5f BIS spectrum. Spectroscopic and transport properties of alloys with A=Th gave additional proof. Th should take the 4+ valence state, so there is no Fermi level tuning and no development of Kondo properties, exactly as observed experimentally. Studies on A=Zr showed similar results but were complicated by effects apparently due to surface segregation. After the seminal work on $Y_{1-x}U_xPd_3$ the impurity origin of the non-Fermi liquid properties was questioned, in part because the U concentration in the alloys is quite large. This issue was addressed by using the spectroscopic knowledge of the behavior of Th. By counter-doping with Th, one can vary the U concentration while keeping E_F fixed in the system $Y_{1-x}(U_yTh_{1-y})_xPd_3$. Transport studies at UCSD show that the major variation in T_K occurs when the Fermi level is tuned and the uranium concentration is fixed, rather than when the U concentration varies while E_F is fixed.

In our early work, in the home lab, we also found in $Y_{1-x}U_xPd_3$ a strong growth with decreasing x of 5f BIS spectral weight just above E_F . Several years later we were able to observe the corresponding 5f spectral weight growth just below E_F , for x as small as 0.01, by performing at the ALS much higher resolution resonant photoemission (RESPES) studies than could be done in our initial work. As this occurs, we found that the U 4f core level lineshape for small x changes from the two-peak shape of UPd_3 to the 3-peak shape that is more typical of mixed valent U materials. [Pub #'s 2, 3, 5, 9, 13]

4. *Non-Fermi liquid behavior and the role of Pd in $U(Cu_{1-x}Pd_x)_5$*

Another system studied in this NFL work is $U(Cu_{1-x}Pd_x)_5$, shown by another group to have similar transport behavior as occurs in $Y_{1-x}U_xPd_3$. For x=0, this system is a normal metallic antiferromagnet with the $AuBe_5$ structure, and as x increases its Neel temperature falls to zero slightly before x=1. In the range $1.5 < x < 2$ the material displays NFL behavior before reaching

a spin glass regime for $x > 2$. As x increases beyond 2 the material passes through a mixed phase region which gives way by $x=4$ to the AuCu_3 structure of the $x=5$ compound. We made a series of RESPES, XPS, UPS and BIS measurements on this system, using samples prepared at UCSD with $x=0, 1, 1.5, 2$ and 4 . As x increases from 0, a sharp PES peak near the Fermi energy E_F decreases, and a shoulder near E_F in the BIS spectrum decreases. At the same time, the leading edge of the U 4f core level spectrum also decreases. The U 4f core level XPS spectra of $\text{UCu}_{5-x}\text{Pd}_x$ can be fit within the impurity Anderson model. The general fitting techniques are ones developed for Pu core level spectra (topic 2 above). The three rather distinct sub-peaks of each spin-orbit component are interpreted as arising from final 5f valence states, in order of highest to lowest energy, $5f^2$, $5f^3$ and $5f^4$, but considerably mixed by hybridization. Multiplet splittings of the higher binding energy peaks, not included in the model, have been simulated with good success by variable energy broadening. The intensity reduction of the " $5f^4$ " peak relative to the " $5f^3$ " peak as x varies from 0 to 2 signals decreasing hybridization. The spectra for $x=1.5$ and 2 are remarkable in that for only one other metallic uranium system is the least binding energy peak not the largest. One sees by this that the special role of Pd in these alloys is to give unusually weak hybridization, yielding spectra that are more readily interpreted than usual. This may also be the reason why many NFL uranium materials are Pd-containing. For $x=4$ the spectrum is abruptly like that of U^{4+} in UPd_3 , a spectrum with two features which we ascribe to strongly admixed $5f^2$ and $5f^3$ final states analogous to the case of UO_2 . A good fit for $x=4$ (and UPd_3) can be achieved by further decreasing the hybridization and also shifting the 5f binding energy to stabilize the $5f^2$ valence state. For the Pd rich alloys, the analysis gives the valence expected from previous work on UPd_3 , nearly integer valent f^2 , with f^3 the lowest excited state. This in turn gives confidence in the result for the Cu rich materials, nearly 50% mixed valent but with f^2 lying slightly lower than f^3 . The crystal structure change correlates with this valence jump.

5. *$\text{UPd}_{3-x}\text{Pt}_x$ alloys –correction of results by other workers*

An early paper (J. Appl. Phys. **63**, 3680 (1988)) by other workers reported a remarkably large change in the U 5f spectrum of the alloy series $\text{UPd}_{3-x}\text{Pt}_x$ for x as small as 0.025. This conclusion and the interpretation in the paper, and also in subsequent papers by the authors, rest entirely on the observation that the spectra for $x=0$ and $x=1.5$ are anomalously out of the sequence of the spectra for x between 0 and 1.5. In RESPES work done at the ALS with Jonathan Denlinger we found that this conclusion is wrong. Curiously, the spectra reported previously for the minutely doped alloy are correct and reproducible, but the spectrum of UPd_3 itself ($x=0$) is quite different from the UPd_3 spectrum obtained repeatedly by us and other workers over several years time. The big change reported in the early paper is thus an artifact of comparing to an incorrect end point spectrum. In fact there is no significant difference for x even as large as 0.1. Further, the anomalous $x=1.5$ spectrum also cannot be reproduced. A great similarity to UO_2 spectra (actually noted in the early paper) makes it nearly certain that the incorrect spectra reported for $x=0$ and $x=1.5$ are in fact due to oxygen contamination. [Pub # 46]

6. *Signatures of the Anderson lattice in the Fermi surface and electronic structure of URu₂Si₂*

URu₂Si₂ is a well-known heavy fermion compound. It has a phase transition at 17.5 K, thought in early work to be due to anti-ferromagnetic (AF) order, and then becomes a superconductor below 1.2 K. The γ value for the specific heat shows a big jump at the supposed AF transition, from 180 mJ/mol-K² to 65 mJ/mol-K², attributed in some models to a large FS change. Since the late '90's strong evidence has been adduced from various studies, especially neutron scattering by Amitsuka and co-workers, that the 17.5 K transition involves a hidden order of some kind, and possible phase separation between a phase with the hidden order and an AF phase.

The XRu₂Si₂ system is near-ideal for studying the interaction between the atomic-like f electrons and the free-electron-like conduction band electrons, because (1) by varying X, the number of f electrons nominally changes from 0 (Th), to 1 (Ce) and to 2 (U), (2) the crystal structure is identical for different X's, (3) large single crystals are available and cleave well, (4) CeRu₂Si₂ and URu₂Si₂ have been heavily studied and thus a lot of background information is known about them, and (5) detailed first-principle one electron band calculations are available. Items (1) and (2) are essential to experimentally characterize the band structures with and without f electrons. This is vitally important because it enables us to find common features, i.e. the "background" conduction bands, between the bands of ThRu₂Si₂ and Ce/URu₂Si₂, and then to identify the f bands and the change of the band structure and FS due to the f bands. In collaboration with Jonathan Denlinger, we have used the combination of RESPES and ARPES to give a global overview of the f and non-f contributions to the near E_F states and the Fermi surfaces of the XRu₂Si₂ system. This is the most complete and systematic ARPES study of the **k**-dependent electronic structure of f-electron materials ever made. The work for X=Ce and La is described in section D above.

Thus far URu₂Si₂ has been measured above the 17.5 K transition. Although the Ru/Si spd bands are generally well given by the LDA, there are major differences between the observed E_F band crossings and those calculated in the LDA for URu₂Si₂. The LDA accommodates the extra f-electrons in f-bands that cross the Fermi surface and change its topology from what would obtain in their absence. We found that the observed E_F crossings can be understood much better in terms of flat renormalized f-bands hybridizing to the non-f electronic band structure, as in Anderson lattice models. In particular the clear signature of the Anderson lattice model is the finding that essentially non-dispersive f-weight is confined near E_F to the interior of d-band hole pockets, the latter being essentially those that would occur in the LDA in the absence of occupied 5f bands. [Pub # 47, 51, 66]

7. *T-dependent f-spectra in URu₂Si₂ ARPES*

A fundamental property of Kondo physics is the existence of a temperature scale below which local magnetic moments are quenched as the system settles into a single ground state. Such temperature dependence has been observed for 4f photoemission spectra in Ce and Yb compounds. No significant temperature variation in photoemission has yet been reported for any U compound. In the course of our ARPES work on cleaved surfaces of URu₂Si₂ we found a

dramatic temperature dependence of the U 5f spectral weight that is confined to the interior of a d-band hole pocket. Very Important for this finding is the relative absence of d-states just below E_F at this special point in k-space, which thus provides a nearly ideal location at which f-states alone can be probed with minimal interference from other spectral weight. Previously no significant T-dependence of angle-integrated valence spectra from scraped single crystals of URu₂Si₂ was observed. It is significant that angle-integrated spectra from our cleaved surface also do not reveal a discernable T-dependence, due to the small fraction of T-dependent weight. Hence the use of angle resolution and k-point selection has been essential in obtaining this first-ever observation of T-dependent U 5f spectral weight. The temperature range of the variation is consistent with the bulk observation by point-contact spectroscopy of a resonance at E_F which appears just below 60K and which is generally associated with Kondo physics. [Pub #'s 51, 57]

F. Electronic Structure of Divalent Hexaborides

1. *Bulk X-point gap in divalent XB₆ (X=Eu, Ca, Sr, Ba)*

Great interest in the divalent hexaborides has been generated recently by the discovery of ferromagnetism (FM) in La doped CaB₆ and by exotic theoretical models to explain the unusual magnetism, e.g. that it represents the ground state of a dilute electron gas or of a doped excitonic insulator. Central to most thinking about the divalent hexaborides is the presumed existence of a band overlap between the top of the boron valence states and the bottom of the cation d-conduction band at the X-point. In the absence of such overlap stoichiometric divalent hexaborides would be insulators. Band overlap is predicted by band structure calculations and magneto-oscillatory studies have been interpreted as showing the implied hole and electron pieces of FS. By showing an isolated X-point electron pocket separated from the X-point boron valence band maximum by a gap > 1 eV, our ARPES studies of EuB₆ and SrB₆ provided the first spectroscopic warning that the semi-metal picture could be wrong. Independently a band calculation that includes a GW self-energy correction predicted CaB₆ to have an X-point band gap of 0.8 eV, similar to what we have observed, but a dispute has arisen in the GW community on the matter because other GW workers have found that the self-energy actually enhances the band overlap. Thus the issue of the calculation of bandgaps in semiconductors, thought to have been resolved by GW calculations in the late '80's, has arisen again, in the context of a new material for which the experimental answer was not known in advance. Most recently we have obtained new ARPES data and complementary data from bulk sensitive soft x-ray emission and absorption which, taken together, demonstrate conclusively the existence of a bulk X-point gap and a global band structure in agreement with the initial GW calculations. An initially confusing aspect of the ARPES data, now recognized although not understood, is that the position in the bandstructure of the chemical potential at the surface can be quite variable, from cleave to cleave, with anion and even as a function of time. In any case, our work shows that no model of the FM that requires band overlap can be correct, and also provides guidance to theorists currently studying the problem. [Pub #'s 63, 64, 65]

2. *Vanishing X-point gap for trivalent hexaborides*

In contrast to our finding of no X-point band gap in divalent hexaborides, described in the item just above, we have found that ARPES spectra show that the gap is reduced for $\text{Ca}_{0.8}\text{La}_{0.2}\text{B}_6$, is essentially zero for mixed valent SmB_6 (≈ 0.6 conduction electrons/Sm) and negative for trivalent LaB_6 . These results indicate a nontrivial change of the band structure with the occupation of the conduction band, essentially that such occupation causes the conduction band to be stabilized relative to the boron-based valence band. [Pub #'s 47, 58]

G. Miscellaneous Studies

Collaborative work on several other problems in the electronic structure of solids resulted in publications during the period of the grant. [Pub #'s 10, 12, 16, 18, 21, 28, 31, 38, 39, 40, 54, 55, 61, 62]

III. Publications and Talks

Reprints and Preprints of papers

1. L.-Z. Liu, R.O. Anderson and J.W. Allen, "Crucial Role of Inelastic Background Subtraction in Identifying Non-Fermi Liquid Behavior in Existing ARPES Lineshape Data," J. Phys. Chem. Sol. **52**, 1473 (1991).
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Invited Conference and Summer School Presentations

1. J.W. Allen, "Electron Spectroscopy of Strongly Correlated Electron Materials-Recent Issues and Results," Adriatico Research Conference on "Open Problems in Strongly Interacting Electron Systems," ICTP Trieste, July, 1991.
2. L.-Z. Liu, "Fermi Level Tuning and the Kondo Resonance in $Y_{1-x}U_xPd_3$," March Meeting of the American Physical Society, "Indianapolis, IN, March, 1992.
3. J. W. Allen, "Fermi Level Tuning and the Kondo Resonance in $Y_{1-x}U_xPd_3$," International Conference on Strongly Correlated Electron Systems," Sendai, Japan, Sept., 1992.
4. J.W. Allen, "Electron Spectroscopy of Heavy Fermion Actinide Materials," Workshop on Synchrotron Radiation in Transactinium Research, Lawrence Berkeley Laboratory, Oct. 1-2, 1992.

5. J.W. Allen, "Luttinger Fermi Surface of Metallic Gap States in $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$," Gordon Research Conference On Superconductivity, Oxnard, CA, Jan. 4-8, 1993.
6. J.W. Allen, "Electron Spectroscopy and Hubbard-Problems and Opportunities," NATO Advanced Research Workshop on the Physics and Mathematics of the Hubbard Model, San Sebastian, Spain, Oct., 1993.
7. J.W. Allen, "Materials Systematics and Insulator to Metal Transitions in 3d Transition Metal Oxides," Japan Science and Technology Agency Workshop on Transition Metal Oxides," Hakone, Japan, Dec., 1993.
8. J.W. Allen, "Probing Correlated Electron Systems by Photoemission Techniques," International Conference on "Strongly Correlated Electron Systems," Amsterdam, The Netherlands, Aug. 15-18, 1994.
9. J.W. Allen, "Fermi Liquids and non-Fermi Liquids-the View From Photoemission," Euroconference on "Cross-Over Phenomena in Solid State Physics: From Weak to Strong Couplings in Electronic Systems," Turin, Italy, Oct. 3-7, 1994.
10. J.W. Allen, "Fermi Liquids and non-Fermi Liquids-the View From Photoemission," Stanford Conference on Spectroscopies in Novel Superconductors," Stanford, CA, March 15-18, 1995.
11. J.W. Allen, "Correlated Electrons in Solids: Changing Paradigms through Synchrotron Studies," Symposium on Synchrotron Radiation Research, Synchrotron Radiation Research Center, Hsinchu, Taiwan, June 22-23, 1995.
12. J.W. Allen, "Photoemission and Inverse Photoemission Studies On Actinide Materials-Does Any Model Work?" International Conference on VUV Radiation XI, Tokyo, Japan, August, 1995.
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14. J.W. Allen, "Photoemission Studies on Low Dimensional Materials," Princeton Workshop on Correlated Electron Materials, Princeton, NJ, November, 1995.
15. J.W. Allen, "Fermi liquids and non-Fermi liquids--the view from photoemission," Institute for Theoretical Physics Workshop on Non-Fermi-Liquid Behavior in Solids, Santa Barbara, CA, May 7, 1996.
16. J.W. Allen, "Fermi liquids and non-Fermi liquids--the View from Photoemission," Institute for Theoretical Physics Conference on Non-Fermi-Liquid Physics in Metals, Santa Barbara, CA, June, 1996.
17. J.W. Allen, "Correlated Electrons: something old, something new, something borrowed, something blue," Workshop des BMBF--Verbundprojetes 05--Synchrotron Spectroscopy of New Correlated Materials, Feb. 20-21, 1997, Würzburg, Germany.
18. J.W. Allen, "Photoemission Lineshapes in f-electron and Low Dimensional Systems," Institute for Theoretical Physics Workshop on Photoemission Lineshapes: the Sudden Approximation and Beyond," Santa Barbara, CA, April, 1997.
19. J.W. Allen, "Electronic Structure in Solids Studied by Photoemission Spectroscopy," DoE panel on the Future of U.S. Synchrotrons, M.I.T., Cambridge, MA, May, 1997.

20. J.W. Allen, two lectures on "Electron Spectroscopy of Solids," NATO Summer School on Correlated Electron Physics, Cerveira, Portugal, July, 1997.
21. J.W. Allen, "Applications of Angle-Resolved Photoemission to Correlated Electron Systems," U. of Wisconsin Synchrotron Radiation Center Workshop on "Correlated Electron Systems," held in Stoughton, WI, Oct. 24-25, 1997.
22. J.W. Allen, "Fermi Surfaces and Near E_F ARPES Lineshapes in Correlated Electron Materials---Mostly Low Dimensional and Mostly Oxides," Argonne National Laboratory Workshop on "Poorly Conducting Metallic Oxides," held in Argonne, Illinois, June 10-12, 1998.
23. G.-H. Gweon, "Gap Anisotropy and Imperfect Nesting of SmTe_3 : Detailed CDW Mechanism Revealed By ARPES," Aladdin Lamp Award Talk, U. of Wisconsin Synchrotron Radiation Center User's Group Meeting, Stoughton, WI, Oct. 23, 1998.
24. J. D. Denlinger, "Advances in Photoemission Spectroscopy of f-electron Materials," International Conference on Strongly Correlated Electron Materials, Nagano, Japan, August, 1999.
25. J.W. Allen, "ARPES lineshapes—a decade later," Topical Workshop on Low-Dimensional Systems, SRC User Meeting, Stoughton, WI., October, 1999.
26. J.W. Allen, "Kondo resonance behavior of heavy Fermion systems," 44th Annual Conference on Magnetism and Magnetic Materials, San Jose, CA., November, 1999.
27. J.W. Allen, "ARPES lineshapes and Fermi surfaces of non-cuprate quasi-low dimensional systems," Aseva Summer School Workshop WS-5 on "Electron Spectroscopies and Strongly Correlated Electron Systems," Avila, Spain, July 10-12, 2000.
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30. J.W. Allen, "Comparative Study of the Electronic Structure of XRu_2Si_2 ($X=\text{La, Ce, Th, U}$): Probing the Anderson Lattice," Stanford Synchrotron Radiation Laboratory Workshop on "New Scientific Opportunities in Ultra High Resolution Spectroscopy," Oct. 21, 2000, Stanford, CA.
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32. G.-H. Gweon, "Non-Fermi Liquid Photoemission Line Shapes in Quasi-One-Dimensional Mo Bronzes," 2001 March Meeting of the American Physical Society, Seattle, WA, USA, March 12-16, 2001.

33. J. D. Denlinger, "Comparative Study of the Electronic Structure of XRu_2Si_2 ($\text{X}=\text{La}, \text{Ce}, \text{Th}, \text{U}$)," 5th Hiroshima International Symposium on Synchrotron Radiation, Hiroshima, Japan, March 15, 2001.
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35. J.W. Allen, "Advances in Photoelectron Spectroscopy of Novel Materials," Fiskfest Workshop: The Future of Materials Physics," Aug. 13-15, 2001, Los Alamos, NM, USA.
36. J.W. Allen, "Absence of X-Point Band Overlap in Divalent Hexaborides: What Controls the Chemical Potential," International Conference on Strongly Correlated Electrons with Orbital Degrees of Freedom," Sept. 11-14, 2001, Sendai, Japan.
37. J.W. Allen, "Electron and Optical Spectroscopy of $(\text{V}_{1-x}\text{A}_x)_2\text{O}_3$ ($\text{A}=\text{Cr}, \text{Al}$): New Views and Echoes From the Past On a Paradigm Mott-Hubbard System," Workshop on Ordering Phenomena in Transition Metal Oxides," Oct. 7-10, 2001, Irsee, Germany.
38. J.W. Allen, "Electronic Structure Probes for Issues in Solid State Chemistry," NSF Workshop on Solid State Chemistry, held at the Univ. of California, Davis, Oct. 12-14, 2001, Davis, CA.
39. J.W. Allen *et al*, "High Resolution Bulk Sensitive Photoemission Study of the Metal-Insulator Transition in $(\text{V}_{1-x}\text{Cr}_x)_2\text{O}_3$: Comparison to LDA + DMFT Theory," International Workshop on High Resolution Photoemission Spectroscopy of Correlated Electron Systems, Jan. 15-17, 2002, Osaka, Japan.
40. J.W. Allen, "Isakson Prize Talk: Quasi-particles and their absence in photoemission spectroscopy," 2002 March Meeting of the American Physical Society, Indianapolis, IN, USA, March 20, 2002.
41. J.W. Allen, "Ubiquitous generalized signatures of electron fractionalization in quasi-low dimensional metals," International Conference on Frontiers in Condensed Matter Physics: Electronic Structure and Properties," June 10-14, 2002, Groningen, The Netherlands.
42. J.W. Allen, "Prominent Metal Phase Quasi-particle Peak and High Temperature Correlation Gap Filling in Photoemission Spectra of $(\text{V}_{1-x}\text{Cr}_x)_2\text{O}_3$: Comparison to LDA + DMFT Theory," morning seminar for Kavli Institute for Theoretical Physics Workshop on "Realistic Theories of Correlated Electron Materials," Sept. 16, 2002, Santa Barbara, CA.
43. J.W. Allen, "Angle Resolved Photoemission Spectra of Low Dimensional and Heavy Fermion Correlated Electron Materials," afternoon seminar for Kavli Institute for Theoretical Physics Workshop on "Realistic Theories of Correlated Electron Materials," Sept. 16, 2002, Santa Barbara, CA.
44. J. W. Allen, "Ubiquitous generalized ARPES signatures of electron fractionalization in quasi-low-d metals," for 2002 International Conference on the Low Energy Electrodynamics of Solids, Oct. 14-18, 2002, Montauk, NY.

45. J.W. Allen, "Prominent Metal Phase Quasi-particle Peak and High Temperature Correlation Gap Filling in Photoemission Spectra of $(V_{1-x}Cr_x)_2O_3$: Comparison to LDA + DMFT Theory," Kavli Institute for Theoretical Physics Conference on "Realistic Theories of Correlated Electron Materials," Nov. 18-22, 2002, Santa Barbara, CA.

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2. R. Claessen, R.O. Anderson, J.W. Allen, C.G. Olson, and W.P. Ellis, "Electron Self Energies Measured by High-Resolution Angle-Resolved Photoemission in 1T-TiTe₂," (abstract) Bull. Am. Phys. Soc. Ser II, **37**, 122 (1992).
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4. R. Claessen, M.G. Smith, J.B. Goodenough, and J.W. Allen, "Photoemission Spectroscopy Study of Ba(Sn, Sb)O₃," (abstract) Bull. Am. Phys. Soc. Ser II, **37**, 122 (1992).
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8. L.J. Tjeng, J.-H. Park, J.W. Allen, C.L. Seaman, M.B. Maple, C.T. Chen, "Ce 4f and Fe 3d Magnetic Moments in CeFe₂ Probed By Soft X-ray Magnetic Circular Dichroism," (abstract) Bull. Am. Phys. Soc. Ser II, **38**, 642 (1993).
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44. G.-H. Gweon, J.W. Allen, J.D. Denlinger, C.G. Olson, H. Höchst, J. Marcus, C. Schlenker, and L.F. Schneemeyer, "Ubiquitous generalized spectral signatures of electron fractionalization in quasi-low dimensional metals," (abstract) Bull Am. Phys. Soc. Ser. II, **47**, 801 (2002).
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IV. Personnel

The following personnel have been partially or entirely supported by this contract.

J.W. Allen, the PI.

Undergraduates:

Douglas Armstead obtained UM B.S degree and went to graduate school in physics at the Univ. of Maryland.

Henry Schek obtained UM B.S. degree and went to graduate school in medical biology at the Univ. of Michigan

Leah Shackman obtained UM B.S. degree and went to graduate school in physics at the Univ. of Texas.

Graduate Students:

Robert Anderson graduated from UM with a Ph.D.degree in 1993; now has a permanent position with the Eaton Corporation, Kalamazoo, Michigan.

Jae-Hoon Park graduated from UM with a Ph.D. degree in 1994; after graduate was first a postdoc and then a permanent Beamline Scientist at the NSLS; now Professor of Physics, POSTECH, Pohang, South Korea.

Yuxin Zhang obtained a UM M.S. degree in 1996 and left graduate school to pursue a career in the semiconductor industry.

Gey-Hong Gweon graduated from UM with a Ph.D. degree in 1999; continued in the group as a postdoc while his wife completed her UM Ph.D. degree work; now a postdoc at the ALS.

Jhules Clack graduated from UM with a Ph.D. degree in 1999; now residing in Cincinnati, OH.

Sung-Kwan Mo is a current student expect to graduate with Ph.D. degree in 2005

Feng Wang is a current student expected to graduate with Ph.D. degree in 2006

Postdocs:

R. Claessen was a postdoc in the group from September 1991 through September 1992; now Professor of Physics, Universität Augsburg, Germany.

L. H. Tjeng was a postdoc in the group from September 1992 through September 1993. now Professor of Physics, Universität zu Köln, Germany

J. D. Denlinger, posdoc in the group from Sept. 1996 to Nov. 1999; now a permanent Beamline Scientist at the Advanced Light Source, Lawrence Berkeley National Lab, Berkeley, CA, USA.

G.-H. Gweon, postdoc in the group from July 1999 to Sept. 2002; now a postdoctoral associate at the Advanced Light Source, Lawrence Berkeley National Lab, Berkeley, CA, USA.

H.-D. Kim, postdoc in the group from May, 2000 to Feb. 2002; now a permanent Beamline Scientist at the Pohang Light Source, Pohang, Korea.

V. Statement of residual funds

There are no residual funds remaining.