INVITED PAPER

ELECTRON SPECTRA OF HEAVY-FERMION SYSTEMS

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An overview is presented of current understanding and recent approaches and issues in studies of the single-particle electron spectra of heavy-fermion materials, with emphasis on uranium compounds.

1. Introduction

The various ground state and low-energy phenomena associated with heavy-fermion materials are generally taken to be electronic in origin, specifically due to the f-electrons of these materials, 4f for the rare earths, and 5f for the actinides. It is thus of fundamental interest to characterize the electronic structure of the f-electrons. For non-interacting electrons it is obvious that if one measures the spectrum of ionization and affinity energies for the electrons, that is, the spectrum to remove and add electrons, one will have learned all there is to know about the electronic structure. However, it is a near-certainty that the f-electrons of interest constitute a strongly interacting Fermi system, and that the interactions are responsible for the properties that make these systems interesting. What then should one measure?

For the interacting system, the spectrum to add and remove electrons continues to be of fundamental importance, as the only single-particle spectroscopic information allowed by the principles of quantum mechanics. In general the added hole or electron interacts with the remaining electrons, and the spectrum thus carries information about the interactions, which are, of course, operative to produce the ground state properties of interest. Two-particle spectra, e.g. those of a hole–electron pair, or two holes, in general carry new information, but can be more complicated to analyze because the two extra particles interact, not only with the remaining electrons, as occurs also with a single extra hole or electron, but also with each other. Examples of two-particle spectra are optical, neutron scattering and Raman scattering spectra, which involve electron–hole pairs with the same or opposite spins, and Auger spectra, which involve two holes.

This paper focuses on the single-particle electron spectrum. Theoretically the spectrum is given by the spectral weight of the single-particle Green's function G, and the closest experimental realization is provided by photoemission and inverse photoemission, or bremsstrahlung isochromat spectroscopy, (PES and BIS, respectively). In making the connection of PES and BIS to G, it is assumed that for the photon interaction the sudden approximation holds, i.e., that, respectively, the outgoing or incoming electron in PES or BIS has large enough kinetic energy that its interactions with the system after or before the photon event can be neglected. Then the photon event can be characterized by a cross section. It is important to remember that G was not invented to describe the results of PES/BIS experiments, but is a fundamental theoretical construct for characterizing an interacting many-body system.

The remainder of the paper describes current understanding, and recent approaches and issues, with emphasis on uranium systems because they are the least well understood at the moment.

2. Rare earth systems and the Anderson impurity Hamiltonian

Strongly interacting Fermi systems have multiple energy scales, most of which are displayed in the single-particle spectrum. Electron spectroscopy has had a major impact on heavy-fermion physics by revealing, and forcing cognizance of these energy scales for heavy-fermion rare earth systems containing, for example, cerium and ytterbium. For the rare earth systems, the spectra can be described in considerable detail using the degenerate impurity Anderson Hamiltonian. [1]

The degenerate impurity Anderson Hamiltonian is a model for an N_f-fold degenerate local

orbital hybridized to a band. It contains explicitly the charge fluctuation energy scales, U_{ff}, ϵ_f, and $\rho N_f V^2$, which are, respectively, the f Coulomb repulsion, the f-binding energy relative to E_F, and the hybridization width involving hybridization V of the f-state to the conduction electrons having density of states ρ. There is then a derived low energy scale, the Kondo temperature T_K, which depends exponentially on $(-1/J)$ where J is the Kondo coupling constant formed from the Hamiltonian parameters, roughly as $J = (\rho N_f V^2)/\epsilon_f$ when U_{ff} is very large. All these energy scales are both observable and observed in the spectra. [1,2] The largest energy scale, essential for understanding heavy-fermion behavior, is U_{ff}, which separates a PES ionization peak at ϵ_f and a BIS affinity peak above E_F. The low energy scale T_K is observed as a peak, the Kondo resonance, at E_F, and controls thermodynamic properties such as the T-linear specific heat coefficient, which is proportional to $1/T_K$. The volume dependence of T_K gives rise to the $\alpha-\gamma$ phase transition in cerium metal [3,4]. Another energy scale, the 4f spin–orbit energy Δ_{so}, is observed [2] as fine structure on the Kondo resonance, and in principle, the resonance should also show structure associated with the lattice effects neglected in the impurity approach. This renormalized bandstructure [5] of the quasiparticles has yet to be observed, or identified in electron spectra.

A screening description of cerium spectra also exists. Although the single-particle density of states obtained from the eigenvalues of the local density approximation (LDA) to the density functional method does not give a good description of the measured PES/BIS spectra of rare earth materials, it has been possible to use supercell LDA calculations [6] to obtain the site 4f excitation energies, i.e., the energies of the PES and BIS peaks that lie away from E_F. In the calculation the 4f occupancy of a central site is constrained to be, in the case of cerium, 0 or 2 for PES and BIS respectively. The physical interpretation given to this approach is to regard the peaks away from E_F as poorly screened, or valence-electron screened final states, and the peak cut by E_F as a well screened or f-screened final state, because it corresponds to the ground state charge density in which there is nominally 1 f-electron.

This screening approach has both a great simi-

larity to and a great difference from that of the impurity Anderson Hamiltonian. The similarity [7] is in the screening description, because in the Anderson Hamiltonian spectrum, the final states of the peaks below and above E_F have roughly 0 and 2 electrons, respectively, while those of the Kondo resonance have a number of f-electrons equal to that of the ground state. As has been pointed out in a pedagogical discussion [7], the spin–orbit fine structure can be interpreted as screening, or relaxation, in which the ground state number of f-electrons is obtained, but with the higher energy internal arrangement of spin and orbital angular momentum. The great difference is that the Kondo resonance has associated with it the small energy scale T_K, which governs its width and weight, and controls ground state properties. The LDA approach does not obtain the low energy scale, and so for heavy-fermion systems, the LDA eigenvalues have physical meaning only at the Fermi level, where they appear to give a reasonable description of the Fermi surface geometry in some systems. A related aspect is that LDA calculations do not now give the relative weights of the screened and unscreened peaks, which are also related to T_K.

One current disagreement for rare earth materials involves $YbAl_3$. There exist two sets [8,9] of PES/BIS spectra and two sets of Anderson Hamiltonian parameters in the literature for $YbAl_3$. For one set [8] the spectra are remarkably similar to those of $YbAl_2$ [10], and the valence and thermodynamic properties deduced therefrom are consistent with those deduced for $YbAl_2$ from other measurements. For the other set [9], the spectra are quite different from $YbAl_2$, and the valence and thermodynamic properties deduced spectroscopically are consistent with those of $YbAl_3$. Detailed comments on this discrepancy will be published by the two groups in due course. Here I offer the brief, and purely personal opinion, derived from experience with a number of $YbAl_3$ and $YbAl_2$ samples, that this difference does not reflect an overt lapse of proper experimental techniques by either group, but rather that there are subtle materials science issues involved, possibly concerning the grain boundaries of different samples. The general consistency [1] between thermodynamic and spectroscopic data found when the theory is applied to other systems is a strong

motivation to think that the consistent set of data [9] is, in fact, the one that is characteristic of $YbAl_3$. However, additional work will be required to clarify this important issue.

3. Concentrated uranium systems and alloys

3.1. Introduction

For metallic actinide systems, building up a picture as unified as exists for the rare earth materials is a focus of active research. This discussion concentrates on uranium, for which the largest amount of work has been done. In general, the f-spectra [11] of metallic uranium materials do not resemble those of the rare earth materials. For the latter the 4f spectra never agree with band calculations, and for the former, the 5f spectra nearly always have a bandlike appearance and sometimes agree rather well with band calculations. Thus uranium spectra do not display well-separated peaks showing U_{ff}, and for the heavy-fermion materials there is much more weight in the vicinity of E_F than would be expected from the small value of T_K implied by the very large T-linear specific heat coefficient γ, or by looking at cerium spectra for materials with very large γ. The famous exception is UPd_3, for which the PES/BIS spectrum [12] shows a gap around E_F. The standard understanding [12–14] of the UPd_3 gap is that the 5f electrons are localized and that the gap measures U_{ff}. UPd_3 is discussed further below in the section on dilution studies.

3.2. Use of resonant photoemission

Resonant photoemission (RESPES) for uranium [13] has been used to extract the 5f spectrum of uranium materials in which there is strong competing emission from other elements, such as transition metals with nearly filled d-bands, or systems where the uranium is very dilute. The most commonly used resonance occurs for the photon energy near the uranium 5d absorption edge. Viewed atomically, photons excite $5d^{10}5f^n \rightarrow 5d^95f^{n+1}$ transitions, and the resulting intermediate state decays via a super Coster–Kronig (sCK) Auger process, $5d^95f^{n+1} \rightarrow 5d^{10}5f^{n-1}$ + electron, to produce the same final state as in diirect 5f photoemission. It is thus a resonance in the 5f photo-

emission cross-section and interference between the resonant and direct processes produces a Fano-type lineshape.

Recently it has been claimed [15] that in many uranium materials, the width of the 5f spectrum determined by RESPES is greater than that determined at other photon energies. This conclusion was reached by studying materials such as USi_3, in which RESPES is not necessary to extract the 5f emission because it dominates spectra at other photon energies, e.g., that from X-ray photoemission spectroscopy (XPS). This extra width was attributed to a decay mechanism additional to the sCK one. In this mechanism, most easily described in a band picture for the 5f electrons, the intermediate state is a 5d core hole and a 5f electron above E_F. This state decays by a conventional Auger transition involving two electrons below E_F, accompanied by a transition of the 5f electron above E_F back to E_F. The result is a final state with a valence band hole and a hole–electron pair excitation with the excited electron being at E_F. Because of the second hole, the width appears to be potentially twice that of the "legitimate" one-hole spectrum.

Actually, the width associated with this mechanism is probably a legitimate part of the 5f one-hole spectrum. Viewed in more general terms the final state proposed has only one 5f hole, and the remaining 5f electrons are in an excited state of the $(n-1)$ electron system. A familiar example of this situation is rare earth 4f photoemission in which $4f^n \rightarrow 4f^{n-1}$ transitions produce a spectrum in which excited multiplets of $4f^{n-1}$ appear in addition to the lowest energy one. In this case it is understood by all workers that the 4f Coulomb interactions are responsible for a spectrum with total width much larger than that due to the one-electron bandwidth. The proposed uranium final state is essentially equivalent to that for the Ni valence band satellite, in which the Ni 3d Coulomb interaction produces in the one-hole spectrum extra structure due to final states with two holes and one excited electron. Although this structure in Ni is enhanced by RESPES, it can be observed away from resonance.

In fact, the claim of an extra width found only in RESPES has been challenged [16], convincingly in my opinion, on experimental grounds, based on measurements at various photon energies for

several of the compounds, including UGe_3, UBe_{13} and USi_3, where an extra width was reported. It was found that the RESPES spectrum completely matches the spectrum measured at a photon energy well below the resonance, and that differences from the XPS spectrum are entirely due to the different resolution in the XPS case. It is asserted that there is not extra width in a RESPES spectrum if differing resolutions are taken into account properly, and if the off-resonance and on-resonance spectra are properly normalized before extracting the 5f spectrum.

3.3. Comparison to LDA

One approach to understanding uranium spectra is to search for evidence of the Coulomb interaction by direct comparison of a full RES-PES/BIS spectrum with the LDA density of states. For heavy-fermion uranium materials the first such comparison [17] was made for UPt_3 and UAl_2. It was pointed out that both the PES and BIS 5f spectra are broader than the LDA density of states, and that this additional width must be attributed generically to uranium 5f Coulomb interactions. It was also suggested that the large weight around E_F in the spectra might be due to a sequence of sidebands on a Kondo-type resonance, analogous to the spin–orbit fine structure observed in cerium, but involving the multiplet structure of the relevant valence states. In assessing the roles of various Coulomb interactions, it is important to remember that even though the repulsion U_{ff} may be screened nearly to zero, the same screening mechanisms do not apply for multiplet splittings, which are less well screened [18].

Recently there have been quantitative theoretical predictions [19,20] of the XPS valence band spectrum based on the LDA for two heavy-fermion materials, UBe_{13} and UPt_3, and two other compounds UAu_3 and UIr_3. The theory gives a fully relativistic description of the photoemission process, and associated surface and matrix element effects, in an independent-electron approximation appropriate for LDA theories. After broadening appropriate to the typical resolution of an XPS experiment, and if the Fermi level is adjusted somewhat, it is found that the predicted spectra are in good agreement with measured ones. It was concluded [19] for UPt_3 that the excellent

agreement suggests that the band-structure or itinerant model for the uranium f-electrons is correct, at least at room temperature where many-body renormalizations are presumably weak.

The two conclusions of the work described in the two preceding paragraphs are clearly inconsistent. The situation is that the higher resolution of the RESPES spectra, relative to that of XPS spectra, shows that the experimental width is larger than the calculated one, even when the resolution is less than the calculated width. The authors of ref. [19] point out that the theory used to calculate the XPS spectrum is not applicable for the spectra taken at the lower photon energies, because at these energies the photoelectron is much more sensitive to multiple-scattering and surface effects, which were neglected. The implication is that surface effects may invalidate the lower photon energy data. However, the finding of ref. [16] that XPS and lower photon energy spectra agree apart from resolution differences is strong evidence to the contrary. Also, for UIr_3, angle resolved photoemission spectra [21] taken at lower photon energy agree quite well with band theory. Thus it seems that in the case of UPt_3 (and also UBe_{13}) the favorable theoretical comparison with XPS spectra is fortuitously good because of the low XPS resolution, and that there are significant differences of widths of experimental spectra from those of LDA predictions. The BIS/PES spectra of other heavy-fermion systems, UBe_{13} [22,23], U_2Zn_{17} [24], and URu_2Si_2 [25] show similar widths.

3.4. Satellite analyses

The observation [17] of excess width in the experimental 5f PES spectra of heavy-fermion materials, relative to the LDA densities of state, has been pursued vigorously by Arko and co-workers, [14,26] who have developed the idea that the extra width should be regarded as a satellite. In this work, the PES spectrum is analyzed into two components, which are thought of as a poorly screened state lying at higher binding energies and a well screened state lying at small binding energy. The well screened state would be expected to have larger intensity for larger hybridization and UPd_3 is then taken to be an example in which only the poorly screened peak occurs. If the spectra of cerium are also interpreted in terms of two screen-

ing channels, some commonality for the systematics of uranium and cerium are obtained. In this connection it is interesting that in URu_2Si_2 a similarity to cerium has been found in RESPES, that for the resonance between 102 and 108 eV, the 5f spectral weight closest to E_F resonates at slightly lower energy than the portion further from E_F. [25] For cerium this behavior [1] can be described [27] by the Anderson model, and it will be interesting to see if it occurs in other uranium materials. For BIS spectra, the extra width has also been analyzed from a satellite viewpoint by two groups. Sarma and co-workers [28] have employed a Hubbard model to describe the effect of U_{ff}, adapting a perturbation calculation [29] used to describe the Ni PES satellite. The essential idea underlying this approach is that the ligand–U5f hybridization is sufficiently large that the single-particle 5f spectrum is dominated by uranium–uranium coupling through the ligands, permitting a Hubbard description based on hybrid-Wannier orbitals. Sarma et al assert [28] that such a satellite should appear only in the BIS spectrum, and have argued that extra width in the PES spectrum either does not occur or has other than a satellite interpretation. [15] In contrast, Laubschat and co-workers [30] have recently extended Arko's PES analysis to include the BIS spectrum of UBe_{13} and some U–Pt compounds. Both the PES and BIS spectra are decomposed into two components, producing a picture crudely like that of cerium, with peaks lying above and below E_F, and a peak cut by the Fermi level. As part of the screening picture, the latter feature is taken to be the LDA density of states.

4. Uranium materials–dilution approach

Another approach to obtaining an understanding of uranium spectra is to study the effects of diluting the uranium with a chemically similar element lacking 5f electrons, such as yttrium. The motivation stems from the fact that the near neighbor uranium–uranium separation in many heavy-fermion and other uranium compounds precludes direct hybridization of 5f wavefunctions with one another so that the Anderson lattice model should apply at least as well for these materials as it does for rare earth compounds. The success [1] in understanding cerium spectra is en-

tirely because the impurity approximation, for which high quality theoretical solutions exist, provides a good description of the spectra. Similarly, the LDA treatment [6] of poorly screened peaks also relies on the dominance of single site effects in modeling the final state. It is thus of considerable interest to distinguish single site from lattice effects in at least the large energy scale features of the 5f spectra.

Two published studies are for the 5f PES and BIS spectra of $U_xY_{1-x}B_4$ with $x = 0$, 0.1, 0.3, 0.6 and 1.0, [31] and for the PES spectra of $U_xY_{1-x}Al_2$ with $x = 0$, 0.02, 0.1 and 1.0 [32]. Both of these systems have the same crystal structure for all x. For the PES spectra, especially with x as small as 0.02, RESPES can be very useful for enhancing the 5f emission. For BIS, it is necessary to determine the 5f spectrum by subtracting the $x = 0$ spectrum, a method that can also be used for PES. The latter method has the difficulty of properly relating spectra taken on different samples. In the $U_xY_{1-x}Al_2$ system, assuming random Y and U site distributions, it is straightforward to compute that the probability of at least one uranium nearest neighbor decreases from 0.35 to 0.08 as x decreases from 0.1 to 0.02. It is thus difficult to achieve truly isolated uranium atoms, but for x as small as 0.02 it is possible to be far from the band limit. In both the systems the PES spectra were found to be essentially independent of x, indicating that the large energy scale features are dominated by hybridization to near neighbors, as in the Anderson impurity model. Fig. 1 shows the spectra for the $U_xY_{1-x}Al_2$ system. For the $U_xY_{1-x}B_4$ system, as x decreased, a small increase in the portion of the BIS spectrum furthest from E_F was reported, but no qualitative change occurred. Thus it appears likely that an impurity approach could work for the major features of the two concentrated compounds.

The 5f PES spectrum has also been measured [33] for $Y_{1-x}U_xPd_3$, with a quite surprising result. As mentioned above UPd_3 plays a special role as an archtype of localized 5f electron behavior, for which the absence of 5f weight at E_F is taken to be a signature [12–14]. Within the standard picture of localized 5f electrons one would expect that dilution of the uranium would, if anything, enhance this localization, so that the 5f spectral weight would remain peaked below E_F, and per-
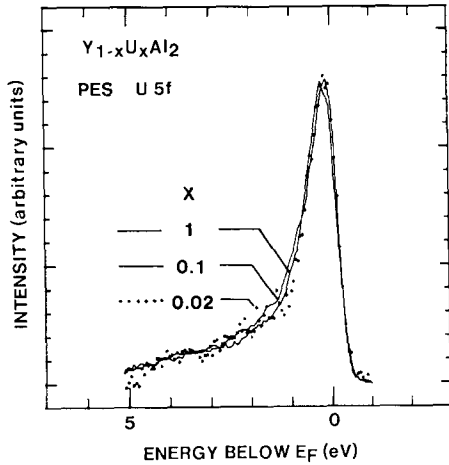
Fig. 1. U 5f spectral weight for $x = 0.02$, 0.1 and 1.0 in the $Y_{1-x}U_xAl_2$ system. The experimental resolution is 0.5 eV. The three spectra are essentially identical.

haps decrease its width. One finds instead the completely counter-intuitive result shown in fig. 2, that although the 5f spectrum does sharpen, its peak moves to E_F. A large fraction of the shift occurs for x between 0.1 and 0.5, and so the shift does not seem to be especially correlated with a crystal structure change that takes place between $x = 0.5$ and 0.9. Taking the standard picture at face value, one would infer that uranium dilution
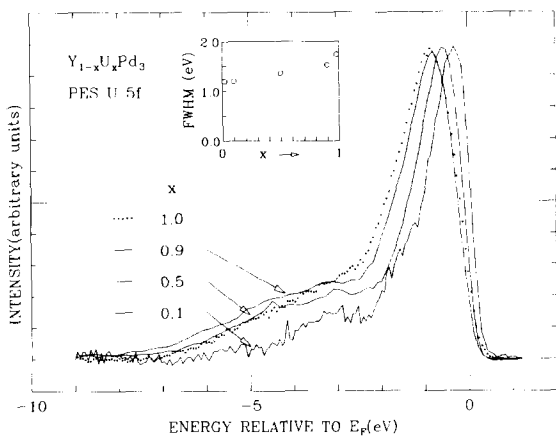

Fig. 2. U 5f spectral weight for $x = 0.1$, 0.5, 0.9, and 1.0 in the $Y_{1-x}U_xPd_3$ system. The experimental resolution is 0.5 eV. Note that the 5f peak moves to the Fermi energy as x decreases. A figure in ref. [33] shows that the spectrum for $x = 0.02$ is essentially identical to that for $x = 0.1$ within experimental uncertainty. Inset shows that the U 5f full-width-at-half maximum (fwhm) decreases as x decreases.

has caused the 5f electrons to delocalize. This inference seems very implausible, and a more probable conclusion is that the suppression of 5f weight at E_F in UPd_3 is somehow a lattice effect, requiring many uranium atoms. It is found that there is a considerable shift in the Pd 4d peaks towards E_F, and the possible connection of this with the 5f behavior is discussed in ref. [33]. Also, the presence of 5f weight at E_F in the dilute samples implies that this is not a good signature of 5f delocalization in the sense of 5f band formation.

5. Uranium materials-relation of thermodynamics and spectroscopy

For all of the various approaches described above it is not obvious how to understand the large mass enhancements of the heavy-fermion materials. The screening pictures intrinsically do not address this question, and for the parameters that have been deduced, [28] the mass enhancement available from the Hubbard model is too small. By far the most effective mass enhancement mechanism is that of the Kondo properties of the impurity Anderson Hamiltonian, where it is proportional to $1/T_K$. However, if the finding from LDA calculations of n_f near 2.5 is correct, as might be expected for this average ground state property, then even for the impurity Anderson Hamiltonian it may be difficult to account for the heavy masses, extrapolating from current results for cerium that n_f near 0.5 corresponds to the mixed valent regime with very large values of T_K and therefore very small mass enhancement. However, the impurity Anderson model seems to be rife with theoretical surprises. In the only theoretical work to date on the interplay of spin and charge fluctuations, addressed specifically at uranium, Rasul and coworkers have found [34–36] numerically in an inverse degeneracy treatment that the impurity Anderson Hamiltonian with fluctuating valence between f^2 and f^3 can have a small energy scale even for n_f near 2.5. This treatment did not include multiplet splittings. In a theory including multiplet and crystal field splittings for a uranium ion, but in the integer valence limit, Cox has described a novel quadrupolar Kondo effect [37]. These theories offer hope that the impurity Anderson Hamiltonian can provide

insight into heavy-fermion uranium compounds. Since the screening pictures being developed for uranium spectra can be regarded as an aspect of the Anderson model behavior, the plea [17,32] for a theoretical treatment of both the ground state and spectroscopic properties of an impurity Anderson model which is realistic for uranium is made here again.

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