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Photoemission and the Kondo model

Serious discrepancies between theory and experiment in heavy fermion systems

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Resonant photoelectron spectroscopy has been used to examine the electronic structure of Ce- and Yb-based heavy fermions. The experimental results are compared with Gunnarsson–Schönhammer calculations which include crystal field and finite U_{ff} effects. Comparison of the model and experimental results reveals a large disparity between predictions and actual results. In particular, the Kondo model predictions of width, energy position and spectral weight for features near the Fermi level are not realized. Additionally, there is disparity between calculation and photoemission data in the Fermi-level crossing positions for spectral features very near the Fermi level. These discrepancies are present for both Ce and Yb heavy fermion systems. We see no evidence of spectral features in the photoemission data which can be uniquely identified with the Kondo resonance while our calculations show the resolution is adequate to clearly distinguish such features.

1. Introduction

There has over the years, been a great deal of interest in the unique and fascinating electronic properties of heavy fermions. A great deal of the theoretical [1–7] and experimental [8–10] work has focused on Ce-based and more recently Yb-based heavy fermions [11–13]. For better or worse, photoelectron spectroscopy (PES) has often played a pivotal role in dispelling or encouraging various models and theories associated with the electronic structure of heavy fermions, ostensibly, because PES provides the most direct experimental probe into the density of electronic states (DOS) of the solid. Supported by PES experiments [8–10,14–16], the Kondo model (KM), based on the impurity Anderson Hamiltonian (IAH) [17] has risen to prominence among the contending models. In particular, predictions of a Kondo resonance (KR), its associated crystal field (CF) and spin-orbit (SO) sidebands, as well as the nature of the main '−2 eV' peak for Ce heavy fermions, have been put on a quantitative footing by the Gunnarsson–Schönhammer (GS) [1,3] and the noncrossing approximation (NCA) [2] computational methods. As the resolution of synchrotron PES has steadily improved over the last decade, there have been questions regarding the success of the KM in describing PES results [18–23]. From an experimental standpoint, the KM has recently been challenged from two opposite ends, high-T_K materials [19] and low-T_K materials [11,21–23], where T_K is the Kondo or characteristic temperature which sets the low-energy scale for spin fluctuations of the heavy fermion.

We examine several predictions of the KM in this paper. In particular, model predictions regarding (i) the width of the Kondo resonance (or its sidebands), (ii) the energy position relative to the Fermi level (E_F) and (iii) the spectral weight of the resonance features, are compared with PES results. Determination of the T_K value to use in the GS calculations comes from quasi-elastic neutron scattering line widths, specific heat, or magnetic susceptibility. Crystal field levels used in the GS calculations are obtained from inelastic neutron scattering. Our GS calculations are the only ones to include full CF levels and double f occupancy (finite U_{ff}). The particle-hole symmetry between Ce and Yb allows the KM to be tested with both Ce and Yb-based heavy fermions [2]. We have chosen relatively low-T_K materials (30–400 K) to test the predictions of the KM so that KR features might be easily distinguished from standard PES features (either nar-

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row bands or core-levels) and also to facilitate measurements above and below the T_K values with our range of temperatures (20–300 °K). Our results show a marked disagreement between model predictions and PES data. The data clearly show that the KM predictions for linewidth, energy position and spectral weight are in error. Some of these discrepancies have been noted previously [24,25] and have been attributed to finite-U_{ff} effects or to CF states – but without calculational proof. We have performed the necessary calculations and find the above explanations fail to explain the discrepancies. The cause of this disparity is not clear at this time and it should be noted that the discrepancies discussed herein are limited to PES results. Within this limited scope however, the discrepancies are large and warrant serious discussion as to the applicability of the IAH within the GS framework to PES.

2. Experimental

Experiments were carried out at the Los Alamos U3C beamline at the NSLS. The samples were flux-grown single crystals where possible ($YbCu_2Si_2$, $YbAl_3$, $CeBe_{13}$) or alternately, are-melted polycrystals for $CeSi_2$ or slow cooling of stoichiometric melt, sealed in Ta, for $YbAgCu_4$. The samples were cleaved in ultra high vacuum at measurement temperature and a chamber base pressure of 7×10^{-11} Torr. Temperature was controlled by placing the sample in direct contact with a cryostat cooled either by liquid helium (sample $T \approx 20$ K) or by liquid nitrogen (sample $T \approx 80$ K). The experimental resolution was determined to be 60 meV at $h\nu = 60$ eV and 95 meV at $h\nu = 120$ eV for high resolution work and 150 meV for full valence band scans at both energies. Although the characteristic Kondo and crystal field linewidths are expected to be smaller than this resolution, we show that the natural linewidth is ~100 meV, and thus experimental resolution is adequate. This point is established in detail elsewhere [21]. The heavy fermions herein are $YbCu_2Si_2$ ($T_k \approx 40$ K), $YbAl_3$ ($T_K \approx 400$ K), $YbAgCu_4$ ($T_K \approx 100$ K), $CeSi_2$ ($T_K \approx 40$ K) and $CeBe_{13}$ ($T_K \approx 300$ K).

3. Results and discussion

Resonant PES is used to enhance the 4f character of Ce heavy fermions. Separation of the 4f signal is achieved by subtraction from the on-resonance spectrum of either an off-resonance spectrum of the same material normalized to account for the 5d resonance, or a resonance spectrum of a La isoelectronic counterpart. The added signal of 13 f-electrons for Yb materials makes it unnecessary to work at resonance so an appropriate balance between cross-section and resolution is chosen in order to enhance the Yb 4f emission. In fig. 1 we present PES data for $CeSi_2$, $CeBe_{13}$ and their La counterparts ($LaSi_2$ and $LaBe_{13}$). The top frame of fig. 1 shows the normalized $CeSi_2$ and $LaSi_2$ spectra. The next frame shows the difference spectrum (i.e. the d-spectral weight subtracted – dots with a dashed line) representing the Ce 4f signal along with a GS calculation with parameters appropriate for $CeSi_2$. Both GS and NCA make approximations in solving the correlated electron problem which leads to limitations for these computational methods. The NCA method is inherently an infinite U_{ff} model but is capable of providing finite temperature information and consequently, temperature dependencies. The GS calculation is strictly valid only at $T = 0$ K but allows for the possibility of double 4f occupancy. We have chosen the GS method so that

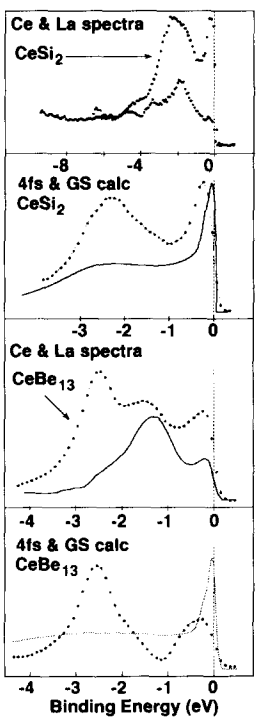

Fig. 1. Resonance photoemission spectra taken at $T = 20$ K. The top frame shows normalized $CeSi_2$ and $LaSi_2$ data. The second frame shows the 4f portion of the $CeSi_2$ emission (dots with dashed lines) and GS calculation for the $CeSi_2$ system (solid line) with parameters as stated in the text. The lower two frames are the equivalent results for $CeBe_{13}$ and $LaBe_{13}$.

double 4f occupancy can be included in the calculations, and restrict comparisons to cases where the measurement temperature is well below T_K. The GS calculations presented are the only calculations which presently take into account double 4f occupancy and full CF sidebands. The parameters used in fig. 1 are $\varepsilon_f = -2.0$, $\Delta_{SO} = 0.28$, $\Delta_{CF1} = 0.025$, $\Delta_{CF2} = 0.048$, BW = 5.0, $U_{ff} = 7.0$, $\Gamma = 0.0877$ (all units in eV) and $T_K = 37$ K, where ε_f is the bare f-level position below E_F, Δ_{SO} is the position of the SO sideband from the KR peak, Δ_{CF1} and Δ_{CF2} are the energy positions [26] of the CF levels, BW is the width of the conduction band below E_F, U_{ff} is the coulomb correlation energy between f-levels, and Γ is the hybridization strength $\sim V^2\rho(E)$. Analysis of Ce 4f data and GS calculations are generally divided into two regions which are the main or '−2 eV' peak representing $4f^1 \rightarrow 4f^0$ emission and the KR and sidebands region (or near Fermi-level region (NFL)). It should be noted that the calculation does not reproduce the data in either region. The width of the GS calculated peak is narrower than the PES data in the NFL region and the E_F crossing position on the GS calculated spectrum is high on the leading edge peak of the spectrum, while the E_F crossing for the data is at the center of the leading edge peak for $CeSi_2$ as it is for every Ce or Yb heavy fermion we have measured. The position of the E_F crossing on the leading edge of the peak nearest E_F is a strong indicator of the underlying DOS. Specifically, if a sharp, narrow DOS resides near E_F, as predicted by the KM, then the E_F crossing position will not fall at the midpoint of the leading edge peak which is the standard position with a broad DOS near E_F. Note that the E_F crossing position for all data presented in this paper is at the midpoint of the leading edge feature, while the GS calculations for Ce systems as well as composite lineshapes generated for Yb heavy fermions all show E_F crossings near the top of the leading edge spectral feature. This strongly suggests broad features in the NFL region of the experimental data.

The lower half of fig. 1 shows the $CeBe_{13}$ and $LaBe_{13}$ resonance PES in the third frame with the difference curve for the 4f emission and the GS calculation in the bottom frame. Once again, the calculation does not adequately reproduce the PES 4f features in either region. In particular, the region of the main peak is nearly flat in the GS calculation, in total disagreement with PES results. The GS parameters for the $CeBe_{13}$ calculations were $\varepsilon_f = -3.2$, $\Delta_{SO} = 0.28$, BW = 10.0, $U_{ff} = \infty$, $\Gamma = 0.1556$ (all units in eV) and $T_K \approx 300$ K. The large flat character of the main peak in the GS calculation arises from the large bandwidth (10 eV) and the large Γ needed to generate $T_K \approx$ 300 K. The constraints used in our GS calculations are as follows: We iteratively chose an ε_f value so that the resulting GS main peak matches the energy position of the '−2 eV' PES peak, we then adjust the Γ value so as to end up with the proper T_K value. Bandwidths are estimated from PES studies [27]. For the case of $CeBe_{13}$ ($CeSi_2$), the Ce atom is surrounded by Be (Si) atoms making the conduction band, with which the Ce f-level hybridizes, the sp band of Be (Si). The BW parameter must reflect this large sp band width.

High resolution PES data are shown in fig. 2 for the $CeSi_2$ NFL region, along with a Voigt function fit to the data and resolution-broadened GS calculations with finite and infinite U_{ff}. The PES in fig. 2 is that of Patthey et al. [14] and represents difference curves of He_{II} and He_I spectra. The superb resolution obtained in this manner (18 meV) has previous been compared to our moderate resolution resonance PES (95 meV) and the two data sets are entirely consistent when resolution differences are taken into account [21]. The Voigt function fitting to heavy fermion data has been shown to provide an adequate analysis of the temperature dependence of the $CeSi_2$ system [21] and results in a natural linewidth of the peak near E_F of ~140 meV. From the two GS calculations shown in fig. 2 one can easily see that the width of the GS calculation is too narrow when compared to PES results, even with CF, finite-U_{ff} and instrumental broadening included. Also, the position and shape of the SO sideband in the GS calculations is not at all consistent with the measured PES spectrum. The finite U_{ff} GS parameters are those used for $CeSi_2$ in fig. 1 while the values which changed for the $U_{ff} = \infty$ calculation to maintain a constant T_K and peak position for the '−2 eV' peak are $\varepsilon_f = -2.4$ and $\Gamma = 0.1296$. It is clear from fig. 2 that the inclusion of double f occupancy has little effect on the agreement (or lack thereof) between model calculation and PES results.

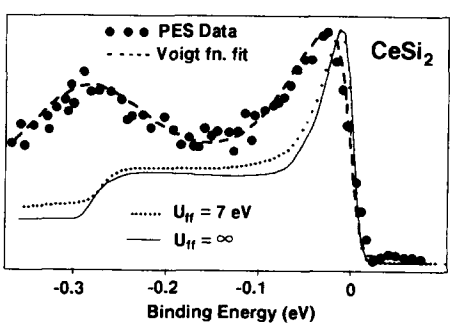

Fig. 2. PES results for $CeSi_2$ from ref. [14] compared to finite and infinite U_{ff} GS calculations which include crystal field effects.

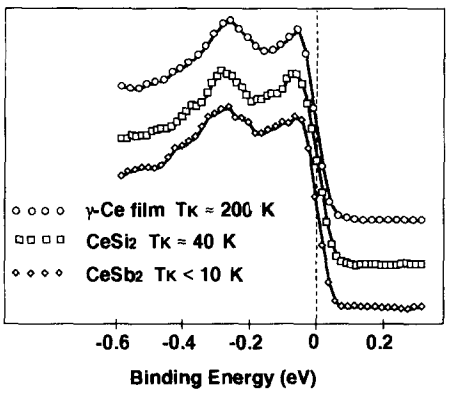

Fig. 3. PES results in the NFL region for the three Ce heavy fermions $CeSb_2$, $CeSi_2$ and γ-Ce. The γ-Ce film results are from ref. [20]. The T_k of these heavy fermions varies by a factor of 20 while the 4f PES emission remains invariant.

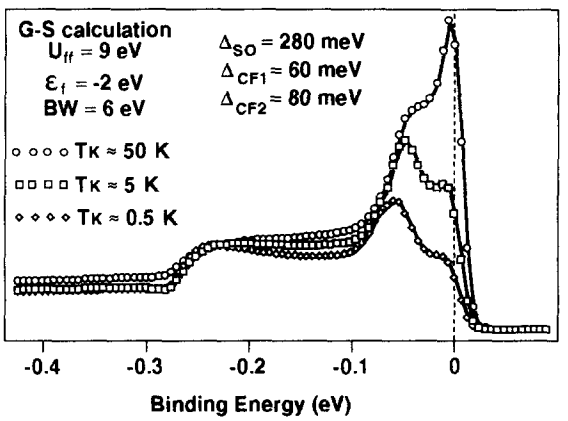

Fig. 4. GS calculations including CF and finite U_{ff} effects. Only the hybridization parameter was changed to vary the T_K values. The calculations clearly show a T_K dependence in-spite of invariant CF and SO sidebands as well as finite U_{ff} effects.

Systematics for Ce heavy fermions are presented in fig. 3. The material $CeSb_2$ has a magnetic transition at ~10 K which sets an upper limit on the T_k if the material does in fact have a T_K, while T_K for $CeSi_2$ is ~40 K and γ-Ce is ~200 K. The γ-Ce PES data is from ref. [20]. With T_K's ranging by a factor of 20, there is very little difference in the NFL region PES spectra of these materials. This T_K invariance would seem contrary to a model which predicts intensity variations with T_K such as the Kondo model. Also, the similarity of the spectra would seem to indicate a universal or invariant 4f character for Ce systems in the NFL region. Proponents of the KM claim that the invariant 4f features in the NFL region are a result of CF and finite-U_{ff} effects. In order to test these claims, we have performed full GS calculations with test parameters intended to elucidate the roles of finite-U_{ff} and CF effects. The parameters used were $\varepsilon_f = -2.0$, $\Delta_{SO} = 0.28$, $\Delta_{CF1} = 0.06$, $\Delta_{CF2} = 0.08$, BW = 6.0, $U_{ff} = 9.0$, $\Gamma = 0.0818$, 0.0887 and 0.0968 (all in eV) to arrive at $T_K = 0.5$, 5.0 and 50 K. The results of these GS calculations are shown in fig. 4. It is obvious that even with the SO and CF sidebands fixed, and full account taken of finite U_{ff} values, the calculated spectra show a clear and direct dependence on T_K. This is in stark contrast to the experimental results of fig. 3, and plainly displays the discrepancy between the KM and PES results. The comparison of GS calculations to PES data in figs. 1–4 shows a definite problem when trying to account for Ce-based heavy fermion PES data by use of the Kondo model. The problem occurs in both directions; PES cannot be used as verification of the validity of the KM, and likewise, the KM cannot be used as a model description of PES results for Ce-based heavy fermions. The remainder of the paper will demonstrate that the KM is also not an appropriate description for Yb-based heavy fermions.

Photoemission data for the heavy fermion $YbCu_2Si_2$ is shown in fig. 5. A complete description of PES results for $YbCu_2Si_2$ including temperature depen-

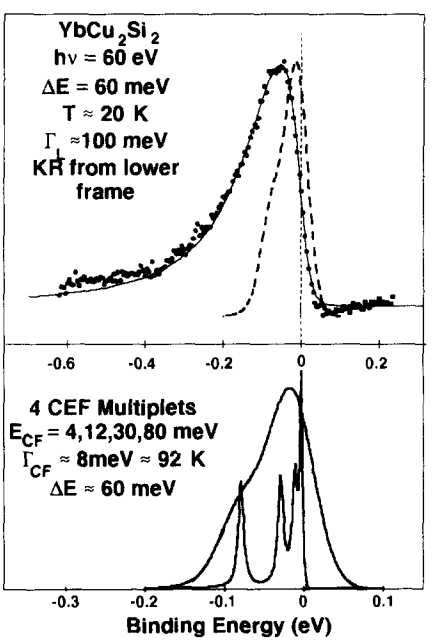

Fig. 5. Photoemission results and KM predictions for $YbCu_2Si_2$ with $T_K \approx 40$ K. The dashed line in the top frame is the resolution broadened composite of the CF and KR peaks in the lower frame. Dots represent the PES spectrum in the the top frame.

dence may be found in ref. [11]. Our present GS implementation for KM predictions does not extend into the empty electron states above E_F, and thus limits our calculations for Yb-based systems. We therefore make use of information from neutron scattering and thermodynamics to construct reasonable composite lineshapes for the Yb systems. In ref. [11], we demonstrated that these composite lineshapes error in favor of the KM in the sense that, the composite lineshapes were in better agreement with the PES data than the GS calculations (see fig. 5, ref. [11]). As mentioned in the introduction, particle-hole symmetry for the 4f levels of Ce and Yb allows a KM interpretation of Yb-based heavy fermions where the KR and all of the sidebands occur on the occupied side of E_F, and thus are directly observable in PES. The upper frame in fig. 5 shows the PES data of the bulk Yb $4f_{7/2}$ peak (dots) for $YbCu_2Si_2$. A Voigt function fit is shown as a solid line, and the dashed line represents a composite spectrum accounting for the KR and associated CF levels. The SO splitting for Yb is 1.29 eV and thus does not enter into the figure. Note that the KM composite spectrum is narrower and closer in energy to E_F than the PES data. Once again, the E_F crossing of the PES data is at the midpoint of the leading edge of the peak, while the crossing point for the calculated composite KM spectrum falls very high on the leading edge of this composite lineshape. The E_F crossing position is not as dependent on high resolution as other comparisons between the KM and PES data. It therefore is applicable even to older PES with lower resolution, and further serves as a consistency check between different data sets. The Voigt function fit to the Yb $4f_{7/2}$ peak indicates a natural linewidth of ~100 meV once instrumental resolution is removed, far greater than the entire width of the CF multiplet spectrum for $YbCu_2Si_2$. In the lower frame of fig. 5, we show the KR and CF multiplets for $YbCu_2Si_2$ with energy positions and widths determined by neutron scattering experiments [28]. Along with the KR and CF sidebands, the broad smooth curve represents the composite of these levels broadened by the 60 meV experimental resolution used in the PES experiment.

In fig. 6 we present PES data from the heavy fermion $YbAgCu_4$ taken at a temperature of 20 K, well below T_K for this material. The upper left frame of fig. 6 shows the full valence band of $YbAgCu_4$ including the Ag and Cu d-levels as well as the Yb 4f-levels. The upper right frame of the figure shows a high-resolution (60 meV) blowup of the NFL region which contains the Yb bulk $4f_{7/2}$ peak. Once again, lineshape analysis shows the natural linewidth of this peak to be greater than the experimental resolution

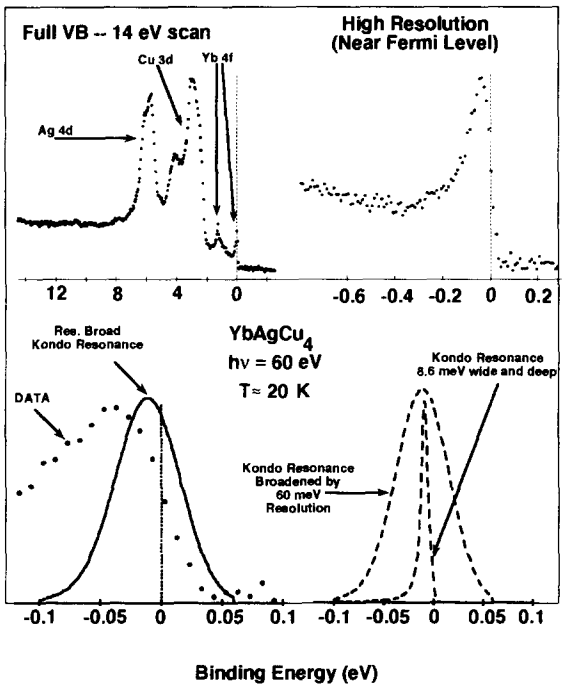

Fig. 6. $YbAgCu_4$ spectra are shown in the top with the full valence band on the left and high resolution data of the NFL region on the right. The lower right frame shows the sharp KR and the effect of 60 meV broadening. The lower left frame combines the KR broadened peak with the PES experimental results.

(of order 100 meV). This is particularly interesting since $YbAgCu_4$ has a $T_K \approx 100$ K (or 9 meV) and neutron scattering experiments do not show any crystal field splittings [29]. The resulting composite spectrum predicted by the KM is simply a peak $\sim K_B T_K$ or 9 meV wide (FWHM) and 9 meV below E_F, broadened by the experimental resolution. The expected KR peak and resulting resolution-broadened spectrum are shown in the lower right corner of fig. 6. This resolution-broadened KR lineshape is overlayed on the PES data for the NFL region in the lower left frame of fig. 6. Once again, the comparison reveals distinct differences between the KM and PES results, both in the energy positions and the widths of the KM and PES peaks. The case of $YbAgCu_4$ should be the cleanest or simplest case for KM studies since CF complications are removed and the KR predicted linewidth would be narrower than standard PES band or core-level features. Nevertheless, the KM fares poorly in this comparison. We have shown that the KM does not accurately describe PES results for Ce or Yb heavy fermions. Also, fig. 3 shows a remarkable *similarity* in 4f emission between *different* Ce-based heavy

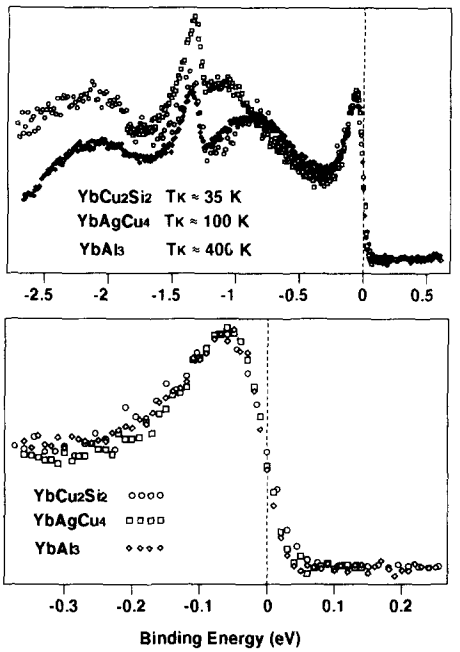

Fig. 7. Yb 4f emission for three Yb heavy fermions. The PES spectra are normalized in height for the peak nearest E_F (the Yb bulk $4f_{7/2}$ peak). The top frame shows full 4f emission with bulk and surface as well as $4f_{7/2}$ and $4f_{5/2}$ components. The lower frame is an enlargement of the NFL region showing the bulk $4f_{7/2}$ peak.

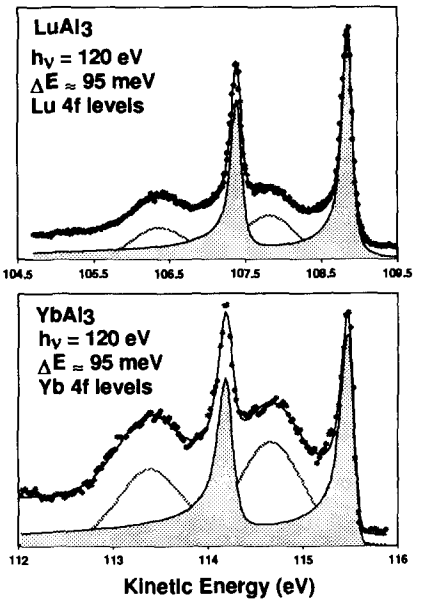

Fig. 8. $LuAl_3$ and $YbAl_3$ PES spectra of the 4f features with nonlinear least-squares fitting of the bulk and surface related features. The bulk 4f features are shaded gray to highlight the similarity between the Yb and Lu 4f levels.

fermions (as indicated by very different T_K's), indicative of a generalized Ce 4f feature. In fig. 7 we present similar results for the Yb-based heavy fermions $YbCu_2Si_2$, $YbAgCu_4$ and $YbAl_3$. The top frame of fig. 7 shows the Yb bulk and surface, $4f_{5/2}$ and $4f_{7/2}$ components for these three materials. These spectra represent the raw PES data which have only been normalized in intensity at the bulk $4f_{7/2}$ peak. The data were all taken at a photon energy of 60 eV and a temperature of 20 K. The lower frame of fig. 7 shows an enlargement of the NFL region and reveals the remarkable similarity of the Yb 4f character in these materials with different T_K's and CF levels. As was the case for Ce compounds in fig. 3, the Yb compounds in fig. 7 show distinct invariance in the 4f spectral features near E_F. Not only do these Yb-based heavy fermions show similarity in 4f features independent of KM parameters, the Yb 4f levels show a striking resemblance to the filled shell, core-like, 4f levels of Lu compounds. In fig. 8 we present PES results for the heavy fermion $YbAl_3$ in the lower frame, and the Lu counterpart $LuAl_3$ in the upper frame. The binding energy of the bulk Lu $4f_{7/2}$ peak is ~6 eV. A non-linear least-squares analysis of the $LuAl_3$ and $YbAl_3$ spectra reveals that the major difference is in the ratio of the bulk-to-surface peaks for the two materials. The bulk-related 4f features are shaded gray to facilitate comparison of the features. The fitting parameters for the two spectra include a 95 meV Gaussian broadening for the instrumental resolution at $h\nu = 120$ eV. The natural linewidths are for $LuAl_3$: $4f_{7/2} = 83$ meV, $4f_{5/2} = 92$ meV, branching ratio = 0.73; $YbAl_3$: $4f_{7/2} = 73$ meV, $4f_{5/2} = 89$ meV, branching ratio = 0.81. The strong similarity between Yb 4f PES spectra of fig. 7 and Yb and Lu 4f PES spectra in fig. 8, would seem to be inconsistent with a KM interpretation where a strong TK dependence is expected. As in the Ce heavy fermions, Yb heavy fermions show 4f PES spectra indicative of a universal 4f character for the host f-level atom rather than a correlation with the KM predictions based on the T_K of the materials.

4. Conclusions

Photoemission results for a number of Ce-based and Yb-based heavy fermions have been compared with predictions of the Kondo model. For the Ce-based heavy fermions, PES results are directly compared with the only GS calculations to include double f occupancy and full crystal field effects. The comparison yields disappointing results. Kondo model predictions based on GS calculations fail to account for the PES results in several crucial areas. In the near Fermi level region, the PES peak positions and widths for $CeSi_2$ and $CeBe_{13}$ are plainly at odds with the GS

calculated spectra. Also, the main or '-2 eV' features are grossly different in width and shape between the GS calculations and PES data. Additionally, the invariant nature of the Ce 4f emission in the NFL region would seem to be in direct conflict with KM predictions. Results for Yb-based heavy fermions show no better agreement than their Ce counterparts when evaluated within a Kondo model framework. The PES spectra can not be easily reconciled with crystal field levels and Kondo temperatures obtained by alternate experimental methods. Once again the PES peaks near the Fermi level are broader and further removed from the Fermi level than allowed for by predictions of the Kondo model. Like the Ce 4f levels, the Yb 4f levels for PES show a distinct invariance with Kondo temperature, and a marked resemblance to the Lu 4f PES spectra. Finally, we see *no* evidence in our heavy fermion PES results which could be unambiguously related to a Kondo resonance or its sidebands. The nature of the discrepancy between model and PES experiments is at this time unclear. What is clear however, is that the Kondo model within the GS framework is not an accurate description for PES spectra, and likewise, PES results do not presently serve to validate or verify the Kondo model description of heavy fermions.

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