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TITLE **Photoelectron Spectroscopy in Heavy Fermions: Inconsistencies with the Kondo Model**

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Photoelectron Spectroscopy in Heavy fermions: Inconsistencies With the Kondo Model*

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ABSTRACT

We have investigated a number of Ce and Yb heavy fermion compounds via photoelectron spectroscopy and compared the results to the predictions of the Imurity Anderson Hamiltonian within the Gunnarson-Schonhammer approach. For the low T_K materials investigated we find little or no correlation with T_K, the only parameter that can be determined independent of photoemission.

1. Introduction

Photoelectron spectra (PES) of Ce and Yb heavy fermions have been the subject of intense study[1-4] for a number of years. The valence band 4f excitations exhibit a complex structure[4] near the Fermi energy which cannot be reconciled with a band structure density of states[5,6] (DOS). Indeed, the heavy electron masses imply an extremely nasrrow structure at E_F which is not obtained from ordinary band theory. In UPt_3, for example, enhancements of the band mass (using the local density approximation, LDA) of order of 20 are needed[7] to compare with measurement. Clearly, the 4f electron interacts with a broad conduction band in a complex fashion which poses a challenge to the theorist.

While a number of models have been proposed[8] to explain the PES data, there appears to be a growing concensus that the Gunnarson-Schonhammer (GS) approach[9-11], as well as the similar non-crossing approximation[12] (NCA), give a comprehensive explanation of valence and conduction band PES for Ce and Yb systems. This very elegant and quantitative theory, based on the Anderson single impurity Hamiltonian[13], assumes that the 4f electrons in heavy electron systems interact very weakly with the conduction band so that a description in terms of this localized model is appropriate. The single impurity Hamiltonian has been extremely successful in describing various bulk properties of heavy electron compounds[8,14], despite the fact that a 4f "impurity" exists at every site, and thus the concept of a "Kondo lattice" has gained acceptance. The greatest impetus toward accepting the GS and NCA approaches was given by PES results[1-4] which for Ce compunds usually show a multi-peaked structure even though the 4f state is occupied by only one 4f electron. A semblance of systematics has been obtained in the past on Ce systems[2] but with instrument resolution far too large to look at fine details. Resolution has steadily improved until now we are ready to re-examine the systematics based on the GS model.

There are several key predictions[9-12] of the GS model. The first is the existence of the so-called Kondo Resonance (KR) which is predicted to be: (i) of width kT_K (where

T_K is the Kondo temperature), (ii) to be of integrated weight kT_K/Γ (where Γ is the hybridization width), and (iii) to lie a distance kT_K above (below) the Fermi energy E_F in Ce (Yb) compounds. For Ce compounds the KR is most appropriately viewed in inverse photoemission (BIS) since only the tail of the resonance lies below E_F ; for Yb compounds the KR can be directly observed in photoemission spectra (PES). An important feature of the KR is the prediction (iv) that the amplitude of the KR will dramatically decrease with temperature, while the total spectral weight decreases only in the amount that the f-occupancy (n_f) changes. This latter will be only a small effect for small T_K materials.

In the presence of spin orbit and crystal field splitting the predicted spectra are considerably more complicated[2]; in addition to the KR of the ground state multiplet, crystal field (CF) and spin orbit (SO) sidebands are predicted above and below E_F, with characteristic temperature scales for renormalization of the spectral weight which can be substantially different[9] than T_K. Within the NCA approach it is these sidebands which yield the bulk of the intensity in the near-E_F region, with the KR itself contributing only a small portion. CF states have been observed as quasielastic peaks in neutron diffraction data[15-17]. If we assume that to first order the neutron quasielastic linewidth is a measure of the temperature scale (T_{Kcf}) of the CF state[18], then these states would be expected to decay with a Kondo temperature of T_{Kcf}. The effective Kondo temperature of the SO sideband (T_{Kso}) can be substantially larger than T_K and renormalization is not likely to be observed[2]. The proponents of the GS model further claim that an additional complication in the near-E_F PES intensity is the effect of finite coulomb correlation[2,9-12] ($U_{ff} << \infty$) which allows a finite amount of double f-occupancy. This presumably adds substantial f-specteral weight in the KR region which will be temperature independent.

We thus see that the predictions of the theory are complex and difficult to separate out from simple mechanisms which also must be part of any metallic compound. Part of the difficulty can be overcome by working with Yb compounds where the KR is fully occupied so that one has the benefit of the full T-dependence[12]. We will show in this paper that despite the subtleties of the effects there are predictions which can be tested and which seem to be inconsistent with results. In the discussion we will look at each prediction in turn and see how it agrees (or disagrees) with experiment.

2. Experimental

Specimens of heavy fermions were either prepared in single crystal form (by flux growth[19]) or melted in an enclosed capsule as an on-stoichiometry melt. In each case they were characterized using X-ray diffraction as well as susceptibility to ensure that the compounds were single phase and displayed the proper T_K. Only two materials ($CeSi_2$ and $YbAgCu_4$) were poly-crystalline. Measurements were carried out at the Los Alamos U3C beamline at NSLS with specimens cleaved in-situ and at temperature to expose a clean surface. Typical base pressures in our UHV chamber were 1×10^{-10} Torr. The instrument resolution varied from ≈ 60 meV at 60 eV photon energy (primarily used for Yb compounds) to 90 mev at the Ce 4f resonance of ≈ 120 eV. Tempearture control was effected by having the specimens in contact with a cryostat cold finger with either liquid nitrogen or liquid helium as the cryogen. Typical spectra were collected in less than one hour.

3. Results and Discussion

In our recent publications[18,20,21] we call into question the systematics expected on the basis of the Kondo picture. In Fig. 1 we show low-resolution valence band PES spectra taken at the 4f resonance (hv = 120 eV) for a series of heavy fermion compounds having T_K's ranging from ≈ 3° K to ≈ 300° K. At this photon energy the 4f and 5d features are expected to dominate the spectrum. At first glance it would appear that the predictions of the GS theory are borne out since three 4f-related features are seen in the spectra corresponding to: i) the KR and the cluster of CF states nearest E_F, ii) the spin-orbit split sideband at -0.3 eV, and iii) the d-screened f^0 state at ≈ -2 eV sometimes called the "main peak" (actually, only the γ-Ce data of Ref. 22 have sufficient resolution in Fig. 1 to clearly resolve the two near-E_F features.) On closer inspection, however, problems develop. Let us look at each property in detail below.

3.1. *Spectral Weight and Systematics with T_K*

One is immediately struck in Fig. 1 by the relatively constant ratios between the spectral weights of the main peak and the two features near E_F. Indeed, the near-E_F peaks are smaller in $CeBe_{13}$ (T_K = 300K) than in materials with smaller T_K's (recall that the weight of the KR, and to a smaller degree the sidebands, should vary[12] as kT_K/T). High resolution data in Fig. 2 for the two near-E_F features show also that the weight ratio of the so-called KR to the SO-sideband seems to likewise remain mor or less constant with T_K (in contrast to Ref. 23 where systematics with T_K are presumably observed). This is especially striking for $CeSb_2$, a material with a ferromagnetic transition[24] at 10° K and possibly a vanishingly small T_K.

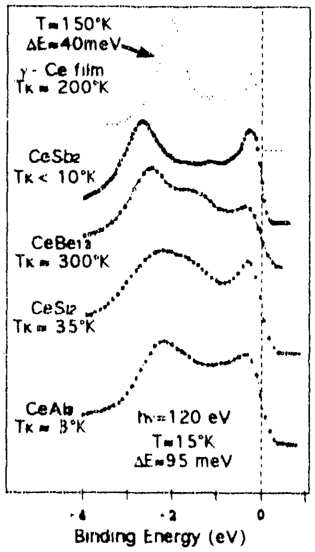

Fig. 1. (Left) Low resolution (ΔE ≈ 150 meV) valence band spectra for several Ce heavy fermions taken at ≈20 K. The high resolution spectrum for Γ-Ce is from Ref. 22. The T_K's vary by two orders of magnitude.

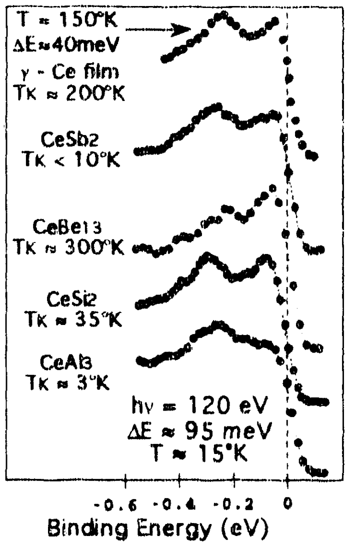

Fig. 2. (Right) High resolution spectra for the compounds in Fig. 1 within 600 meV of the Fermi energy. Note the similarity for all materials.

Two distinct mechanisms have been proposed to resolve the nearly constant spectral weight of the near-E_F features on the basis of the GS or NCA models. The first involves the introduction of the already mentioned CF and SO split states combined with lifetime broadening[23,25]. The difficulties with this approach are that i) the spectral weight (and width) of even these states is to a considerable degree a function of T_K, and 2) one would anticipate *a priory* that the lineshape and amplitude would be a strong function of crystal structure. This does not seem to be the case. While the lineshape will be considered in greater detail in the next sub-section, one can see from Fig. 3, where we show our GS calculation superimposed on the data of Ref. 25, that the mere introduction of CF and SO states results in a very poor fit to the data. Moreover, it would yield far too small a spectral weight for very small T_K materials (e.g., CeAl or $CeSb_2$) as discussed below.

A second proposed mechanism[2,9-11] stresses the importance of the effects of a finite U_{ff} such that doubly occupied 4f states are allowed. It is claimed that these f^2 states yield a dramatic amplitude increase at E_F which accounts for the nearly constant spectral weight in low-T_K materials. This effect is particularly important for very small-T_K materials where the model would otherwise yield only minimal spectreal weight at E_F (see inset of Fig. 4). We have tested this hypothesis with our own calculations for infinite and finite U_{ff} within the GS model. We find that if T_K is constrained to remain invariant for different values of U_{ff}, the net effect of double occupancy is to increase the 4f spectral weight throughout the valence band region without any significant change in relative intensities, and especially no change in widths of the near-E_F features (some narrowing occurs for the main peak since the hybridization must be decreased for small U_{ff} in order to maintain T_K). This is shown in Fig. 3 where we display the two calculated spectra (for $U_{ff} = \infty$ and $U_{ff} = 7$ eV) superimposed on the data of Ref. 25 which represent the highest resolution data to date (18 meV) on any heavy fermion system. Here T_K was constrained to 37°K ($\approx$ thermodynamic value[14] for $CeSi_2$) while the calculated spectra have been broadened by 18 meV to simulate instrument resolution[25] and have been normalized to each other at the main peak at -2 eV. In Fig.4 we also reproduce the spectra displayed in Ref. 2 using their published parameters. Curves A and B are identical to those in Ref. 2 and represent $U_{ff} = \infty$ and 6.5 eV respectively where T_K is allowed to vary from 0.004°K to 0.6°K. Curve C represents U_{ff}=6.5 eV but with T_K = 0.004°K. The unbroadened version of curve A is shown in the inset of Fig.4 superimposed on the broadened curve. Clearly a finite U_{ff} will not yield the

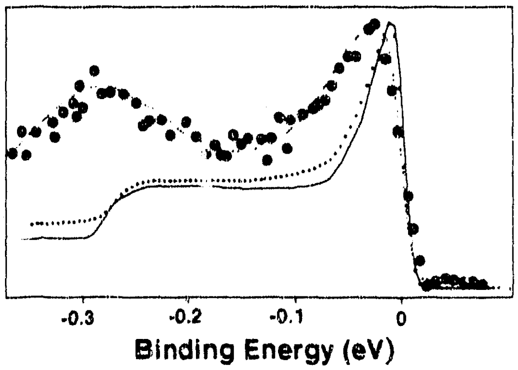

Fig. 3. Near E_F data from Ref. 25 for $CeSi_2$ taken at He I and He II energies with 18 meV resolution. The line through the data is our Voigt function fit. The solid and the dashed curves represent our GS calculations for infinite and finite U_{ff} (7 eV) respectively. T_K was held at the thermodynamic value of 37 K.

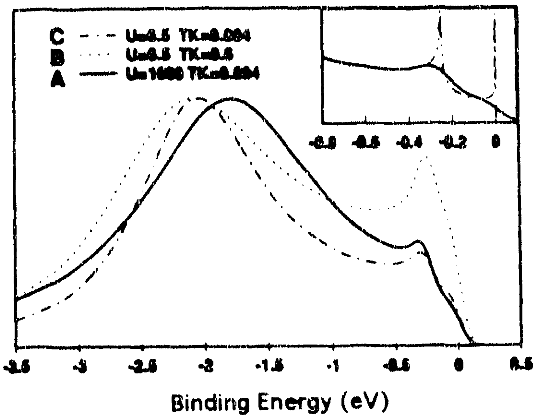

Fig. 4. Spectra obtained from GS calculations to show ineffectiveness of U_{ff} (see text). Parameters are from Ref. 2 for curves A and B. For curve C, U_{ff} = 6.5 eV but T_K is kept invariant with curve B yielding the same spectral weight at E_F. The inset shows the broadened and unbroadened versions of curve A to emphasise the spiked nature of the KR and SO sidebeand.

desired results and we must look for other mechanisms to explain the nearly constant spectral weight with T_K in the near-E_F region. Moreover, from the inset it is obvious that the weight and particularly the width of the SO sideband is strongly dependent on T_K.

3.2. *Spectral Width and Lineshape*

The width of the apparent KR and the SO sideband likewise is nearly constant with T_K in the data, and far broader than our resolution as well as expectations[12]. It is evident from Fig. 3 that the calculated spectrum, even after including CF (determined from neutron diffraction[15-17]) and SO states while at the same time working with U_{ff} = 7 eV, is still much narrower than the measured spectrum. The authors of Ref. 25, whose data we show, recognized that there was a problem with the width and assumed that lifetime broadening was responsible for the extra width, while in Ref. 2 the extra width is attributed to f^2 occupancy. We have already shown the inadequacy of the latter argument above. As for lifetime broadening, while it is included within the NCA model, this broadening cannot exceed kT_K, while the hole lifetime broadening should be insignificant at E_F. If we allow hole lifetime effects, such a large effect at E_F would translate into a 1eV wide feature[26] at 0.3 eV which is not the case. Thus the inordinately large width of the features at E_F relative to calculations remains unexplained. This is especially true for small-T_K materials where from the inset in Fig.4 one sees that the SO width should likewise be very small. Instrument resolution is not a limiting factor in seeing these effects.

The lineshape of these features does not agree with the calculated spectrum. This is particularly evident for the SO feature at -0.3 eV. We, on the other hand, have fit the data of Ref. 25 with Gaussian-broadened Lorentzians (Voigt functions) and obtained the excellent fit shown by the dashed lines through the data. Since Voigt functions are the conventional lineshapes used for fitting to core level spectra, the excellent fit obtained strongly suggests (but does not prove) a core level nature of these states. Indeed, we find that the lineshapes of all the Ce heavy fermions we investigated can be approximated by Voigt functions and vary between 100 meV and 200 meV in width, with the larger value generally applying to the SO peak at -0.3 eV.

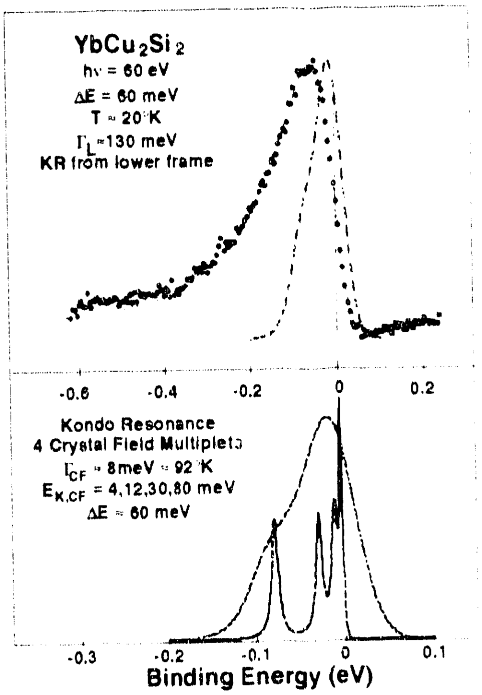

Fig. 5. The peak nearest to E_F in $YbCu_2Si_2$ with the remaining valence band spectrum subtracted off (upper frame). The dashed curve is a simulated Kondo resonance and its sidebands obtained in lower frame by broadening the measured (neutron diff.) crystal field states with a 60 meV Gaussian.

The nearly constant shape and width of the near-E_F features (≈ 100 meV to ≈ 200 meV) extends to Yb heavy fermions as well[27]. In Fig. 5 we show the peak nearest E_F (presumably the KR) for $YbCu_2Si_2$ where all the remaining parts of the spectrum have been subtracted. Here we approximate the crystal field states with equal amplitude doublets having a width determined from neutron diffraction. We have recently shown[18] that this actually overestimates the width of the near-E_F feature compared to a GS calculation. The bottom frame shows these states at the measured energies, with the envelope representing broadening by a 60 meV Gaussian (our resolution). This broadened line is then compared in the top frame to the data. One can see that the same problems are encountered in Yb systems that are encountered in Ce heavy fermions, namely, the width of the features is too large compared to the model and is entirely similar to the widths encountered in Ce systems.

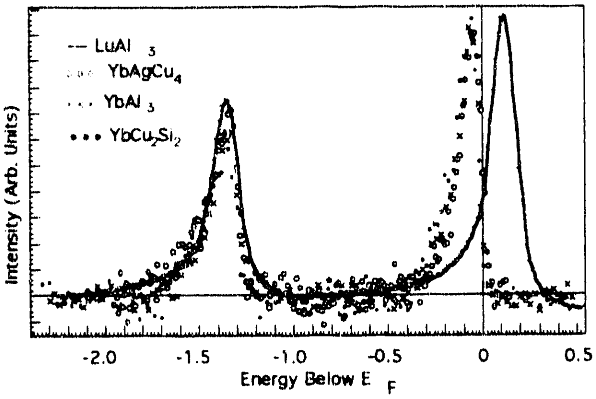

Fig. 6. The two near-E_F spin-orbit split features in three Yb heavy fermions obtained from fitting to raw data (taken at 20K) and stripping away the surface and underlying DOS features (see text). Note that they exactly fall on top of each other, including the branching ratio. The solid line is the same feature in $LuAl_3$ but shifted by 6 eV in order to fall on top of the Yb features. The spin-orbit splitting is somewhat larger, but the similarity to Yb 4f features is striking.

In Fig.6 we show the KR and the SO sideband for three Yb heavy fermions in which the underlying DOS, the ever-present surface features, as well as the secondaries have been subtracted away (more on these subtractions below). The data were taken at ≈ 20°K. The T_K's for these materials vary by more than an order of magnitude[14] (35°K for $YbCu_2Si_2$, 100°K for $YbAgCu_4$, and 400°K for $YbAl_3$) and yet it is possible to almost exactly overlay all the spectra after normalizing at peak amplitude. For effect we also show the 4f spectrum for $LuAl_3$ in which the 4f electrons are situated 6 eV below E_F and cannot possibly be interpreted as the KR and its sideband. We note that they have nearly the same width and branching ratio as the heavy fermion materials, though the spin-orbit splitting is somewhat larger, as expected. This nearly constant width becomes especially difficult to understand for YbAgCu4 where neutron diffraction and bulk property data are consistent[28] with a lack of CF states. If lifetime broadening were responsible for the large widths, then the lack of CF states should result in a much narrower line for this material. On the other hand, the near constancy of the intensity, width, and lineshape is much easier understood if we assume that we are dealing with screened 4f core levels[29-32] at E_F.

3.3. *Temperature Dependence*

Despite the fact that the Kondo resonance is expected to renormalize dramatically with temperature[12], the observation of this temperature dependence can be subtle and difficult owing to the existence of CF and SO states. Moreover, most of the temperature may only manifest itself as a broadening of the KR and its associated sidebands[25,34]. The situation is not entirely clear. Within NCA it appears[12] that the actual loss of spectral weight is predicted to vary with n_f which is a slow function of temperature. The exact Monte Carlo approach[33], on the other hand, suggests a rapid loss of spectral weight in the KR, much faster than the variation of n_f, with the main peak gaining the lost weight. Within NCA, the broad SO sideband may show no temperature dependence up to room temperature, while the CF sidebands may vary on a scale which is slower than T_K (we call it T_{Kcf}).

Bickers et al.[12] show the unoccupied states in Ce as having a different temperature dependence than the occupied states. Here both the KR and the SO sideband display equivalent variation with temperature unlike the SO state below E_F which is nearly temperature independent. This is very significant since a loss of intensity away from E_F is easier to distinguish from normal processes than a loss at E_F (recall that the Fermi function is temperature dependent). But now, because of particle-hole symmetry, the KR for Yb compounds occurs on the occupied side of E_F so that one obtains the full KR intensity rather than just a tail as in Ce materials, and the SO sideband should display a significant temperature dependence. Thus one anticipates a much larger temperature dependence in Yb compounds vs Ce compounds.The bottom line is that a search for temperature dependence over and above simple mechanisms is best done with high resolution PES in Yb compounds. Unfortunately, as we will see below, there are serious (but solvable) complications with surface-related features in Yb materials.

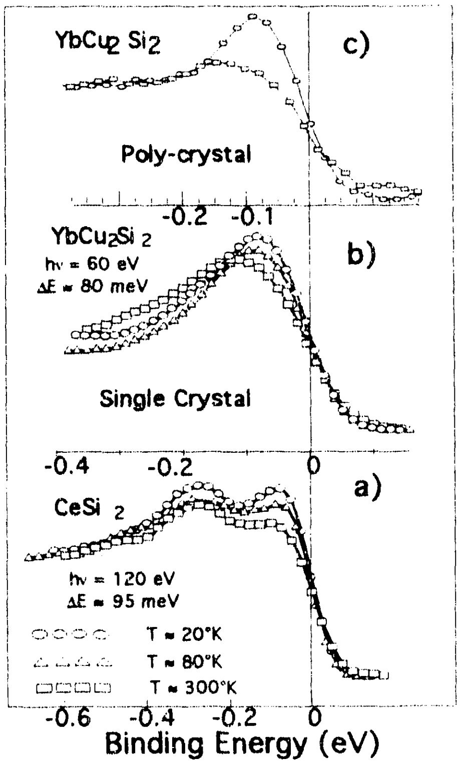

Fig. 7. Raw data at the indicated temperatures (see frame a) for the near-E_F peaks in $CeSi_2$ and $YbCu_2Si_2$. Very little temperature dependence is seen. Frame c is for poly-crystalline $YbCu_2Si_2$. Note that it has a large temperature dependence most likely due to changes in surface structure. For Yb compounds it appears that single crystals are mandatory.

In Fig.7 we show the raw spectra in the near-E_F region for $CeSi_2$ and $YbCu_2Si_2$ for three temperatures (20°K, 80°K, and 300°K). $T_K \approx 35$°K for both materials. The spectra were normalized using the Si-2p core levels for $CeSi_2$ (Fig.7a) and Cu-3d valence bands for $YbCu_2Si_2$ (Fig.7b). Very small, and nearly identical, temperature dependences are seen in both materials, suggesting that spectral wight losses (if any) are nearly the same in both materials. One would have anticipated far more weight loss (perhaps an order of magnitude) in the Yb compound owing to the full occupancy of the KR. We have previously shown[21] that any temperature dependence in $CeSi_2$ is entirely explainable from simple mechanisms (phonon broadening, surface effects, etc.). Thus it is likely that in Yb compounds simple mechanisms are the also the only source of the very small temperature dependence[18,27]. Fig.7c shows an apparent temperature effect seen in an arc-melted button of $YbCu_2Si_2$. We believe that the use of poly-crystalline materials vs single crystals with their stable surfaces can yield an apparent temperature effect (see below for discussion).

3.4. *Problems With Extracting a Temperature Dependence in Yb Materials*

Yb compounds tend to be far more complex in their surface structure than Ce compounds. Indeed, various surface features can be a strong function of temperature and/or surface adsorbates. In our example above we showed the temperature dependence (or lack thereof) for a very stable single crystal of a two dimensional material, $YbCu_2Si_2$, which has been cleaved in vacuum to expose the most stable plane. Even here we discovered that adsorbates (with changing temperature) had a strong effect on the surface related features, although happily the bulk features remained unaffected. Three dimensional materials, and especially poly-crystalline materials, have a less stable surface and are thus much more difficult to analyze.

In any attempt to extract a temperature dependence one must account for at least the following effects as one raises the temperature from 20°K to 300°K: i) changes in amplitudes of surface-related features since Yb is known to be divalent[1,2] at the surface; ii) appearance of additional components due perhaps to oxide formation; iii) changes in underlying DOS due to chemical changes beyond the first crystallographic layer; iv) increase in secondary electron emission due to adsorbates; v) increases in surface core-level shifts. If the bulk feature constitutes only a small percentage of the total spectral weight near E_F ($\approx$20% for $YbAl_3$), then even small changes in the surface features can yield an apparent temperature effect. Some, or all, of these effects have been observed by us in the study of single crystal $YbAl_3$ and poly-crystalline $YbAgCu_4$, as well as in poly-crystalline $YbCu_2Si_2$ (Fig.7c). Indeed, it was the apparent large temperature dependence of the near-E_F peak of the latter which launched our attempts to observe a KR in heavy fermions with our improved resolution. Use of the highly stable single crystals eliminated all interesting effects (Fig.7b).

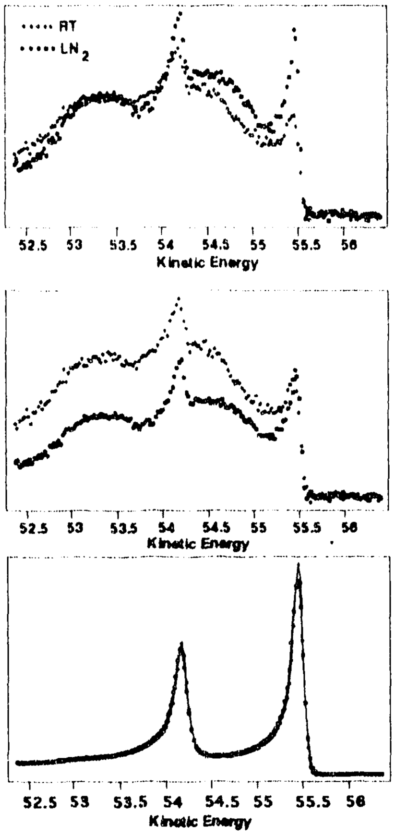

Fig. 8. Spectra at 20 K and 300 K for $YbAl_3$. In top frame the normalization is to surface states while in the middle frame the spectra are correctly normalized to Al-2p core levels. Fitting to the data to extract the bulk 4f's yields no temperature dependence (bottom frame).

In $YbAl_3$ even the use of single crystals fails to suppress the surface driven temperature effects which yield an apparent temperature dependence for the bulk peaks with improper analysis. We show these effects in Fig.8 where in frame A the 20°K and 300°K valence band spectra have been normalized to the surface state at -1 eV with a resulting large apparent temperature effect for the bulk features. In frame B we show the same spectra, but now more appropriately normalized to the Al-2p core levels. One immediately sees that the feature at E_F has not changed much with temperature, most of the changes being evident in the surface features. Thus normalization to surface features, no matter how reproducible the lineshape, can yield misleading results. One can only extract the 4f components from a fit to the data using the best estimates for the behavior of elements i) through v) above. We have performed such fits, which however, because of the complexity of effects are not entirely unique (the differences are mostly in the choice of the particular surface features). Nevertheless, nearly all fitting attempts resulted in a temperature-independence for the bulk 4f peaks. Frame C represents our most reasonable attempt to extract the bulk 4f states (at 20°K and 300°K) from the surface states and the underlying DOS, using a least squares minimization technique with the 4f's represented by Voigt functions. The fitting technique will be described in detail in a later publication[35]. There appears to be no temperature effect at all, which from our point of view is understandible since the first 4f peak occurs at ~60 meV where it would be only slightly affected by the Fermi function.

4. Conclusions

We have shown that many of the trends and systematics anticipated on the basis of the GS model are not realized in spectroscopic heavy fermion data. While the GS model predicts the correct number of 4f-related fetaures and their energy position, it fails to correctly predict the spectral weight, the width of both the KR (predicts much too narrow) and the main peak (predicts much too broad a feature, not discussed), temperature dependence (from NCA), as well as lineshape of the KR, the SO sideband and the main peak. The typical 4f-spectrum of a Ce (or Yb) heavy fermion seems to first order to be unrelated to T_K, crystal structure, or the s-p-d DOS of the conduction band. Indeed, the Yb KR and SO sideband very much resemble the 4f core levels of Lu and W. While we are not prepared to say that the GS model is to be abandoned, we point out that any successful model must be able to satisfactorily explain the near constancy of the data, as well as the magnitude of the spectral widths. Bulk property data are very successfully explained by the Anderson Hamiltonian so that we believe that it is only a matter of some invalid approximation which fails this otherwise elegant GS theory.

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