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ELECTRON SPECTROSCOPY IN NARROW BAND f-ELECTRON COMPOUNDS*

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Summary

We show by the combined means of high-resolution photoemission measurements at low temperatures on uranium and cerium compounds (including heavy fermions) and systematic resonant data on a series of uranium narrow-band compounds, that the "two-peaked" f-electron spectrum commonly found in cerium compounds is also found in uranium compounds. The 5f spectra consist of a superposition of features consistent with the band structure ground state plus a 5f satellite due to poorly-screened final state effects.

1. Introduction

Photoemission spectra in heavy electron systems appear not to display the systematics between cerium-based (4f) and uranium-based (5f) heavy fermion compounds which are otherwise observed in bulk property measurements such as specific heat C_p , magnetic susceptibility χ , resistivity ρ etc. A "two-peaked" structure is usually observed in cerium compounds [1] and identified with 4f emission (a peak near E_F and a second peak at about -2 eV) while in uranium compounds the intensity associated with 5f emission

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tends to display a generic single triangular shape [2, 3] having a very large amplitude at E_F and trailing off slowly to energies as large as -4 eV. Some structure is seen superimposed on the broad 5f spectra, which further complicates matters. Based on bulk property measurements [4] one would have expected extremely sharp features precisely at E_F at low temperatures. While some sharp structure is observed one is nonetheless confronted with 5f spectra having overall widths full width at half-maximum (FWHM) of the order of 1 eV which totally fail to explain the upturn in C_p below about 10 K.

In this paper we will show that the systematics that have thus far eluded researchers are actually present in photoemission spectra. The failure to recognize them previously is simply due to the subtlety of the effect. We will show that the apparent large width of 5f spectra is a result of the existence of a broad 5f satellite at about -1 eV due to final state screening. In the following sections we first give a background of the various models [5] used to explain the spectra, followed by the experiment and how it relates to these models.

2. Models for the excitation spectrum

While there is general agreement that at very low temperatures the f electrons can be treated as a (possibly highly correlated) Fermi liquid [5], there is still ample disagreement as to the wavefunction of the quasi-particles yielding this Fermi liquid; *i.e.* are we to treat this within the framework of a traditional band model [6 - 8], a Hubbard model [9 - 11] or perhaps the “periodic” Anderson model [12, 13] (*i.e.* the “Kondo Lattice”). All of these models require the development of a narrow, heavy electron band at low temperatures to explain experimental data. While photoemission measurements, particularly measurements taken at room temperature, will yield no information regarding the wavefunction of the heavy fermion state, they are nonetheless useful in sorting out the underlying electronic structure on which the heavy fermion state is superimposed. It thus becomes important to understand photoemission spectra and give them a proper interpretation. This is the main thrust of this paper.

Let us first examine what sort of photoemission spectra are anticipated on the basis of the various models. In the band approach the f electrons are hybridized (already at high temperatures) with the ligand p and d electrons and are thus fixed at E_F . Sharp features in the density of states at E_F (a sub-band with $T_F \approx 10$ K) and renormalizations within this feature result in the heavy fermion behavior. (A possible source of the heaviness is interaction with spin fluctuations.) In the band approach [6, 7], the creation of an f hole resulting from photoemission may dramatically perturb the ground state, possibly localizing the affected atomic site. Screening of the hole by an f electron constitutes the well-screened case which may reproduce the original band structure, while screening by extended d or p electrons results

in a poorly screened final state [14, 15]. A typical photoemission spectrum should consist of a superposition of spectra derived from the ground state density, plus an f electron satellite peak at lower binding energy caused by a partially screened localized excited state.

In the Hubbard model one assumes at the start nearly localized f electrons in a narrow band at E_F , only very slightly hybridized with the ligand electrons such that the hopping matrix $t < 0.1U$ where U is the Coulomb correlation energy. This model requires an integral number of f electrons per atom, and all the f density at E_F . A renormalization of the narrow band occurs in the Fermi liquid state. Details of the photoemission spectra are not worked out, but one would anticipate two lorentzians representing d and f screening.

The “periodic” Anderson model [13] is an extension of the dilute single magnetic impurity Anderson model [16] to the concentrated case where there is a magnetic impurity at each f site at ϵ_f below E_F . Gunnarson and Schönhammer [17] have applied these ideas to the calculation of ultraviolet photoelectron spectroscopy and bremsstrahlung isochromat spectroscopy (UPS and BIS) spectra and other properties of dilute cerium-based alloys. Subsequently it has been extrapolated to N_f degenerate and concentrated cases [18 - 24]. The calculated spectrum [21] yields (Fig. 1) a UPS peak at $-\epsilon_f$ (related to f^{n-1}), a structured peak at $\epsilon_f + U$ (U is the Coulomb correlation) related to double occupancy or f^{n+1} and a striking “Kondo resonance” near E_F (or kT_0 above E_F) having a width about $\pi kT_0/N_f$. T_0 is a characteristic temperature (closely related to the Kondo temperature T_k) below which the Kondo resonance develops. Split off above and below the Kondo resonance by the spin-orbit coupling are two sidebands which actually have more spectral weight and width (by perhaps an order of magnitude) than the Kondo resonance itself and are hence more stable in temperature. It is thought [1] that the double-peaked photoemission data in cerium and its compounds at high temperatures reflect the observation of

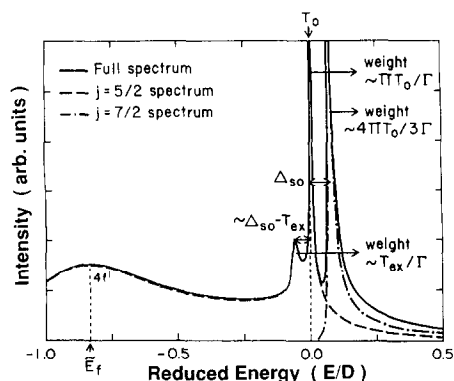


Fig. 1. Particle-hole excitation spectrum (after Cox *et al.*, [21]) expected from a Kondo model for Ce-based systems having only one f electron; the “main” peak is at \tilde{E}_f , both the $f^{5/2}$ and $f^{7/2}$ levels yield a Kondo resonance, as shown; energy scale is normalized to conduction bandwidth D .

the f^0 peak (at $-\epsilon_f$) and a Kondo sideband near E_F , while the Kondo resonance itself is usually not observed owing to insufficient resolution or insufficiently low temperatures. Both the band and the Kondo models can explain the cerium data.

Actinide-based heavy fermion photoemission spectra however appear to resemble neither the ‘‘Kondo resonance’’ predictions (no obvious two-peaked structure) nor the band structure predictions (much too broad). To some extent, there is better agreement with the latter since the features observed superimposed on the broad background correspond roughly in energy with the predictions of band calculations. Allen *et al.* [2] were first to point out the extra intensity relative to band calculation predictions in the -1 eV to -3 eV range. While they attributed the extra intensity to Coulomb correlation, they did not recognize it as a well-defined satellite having the signature of a localized 5f level like that found in UPd₃.

In the following sections we will present first the high-resolution data taken at low temperatures for cerium- and uranium-based heavy fermions to try to reconcile the various features with the above models. This will be followed by systematic room-temperature data on a series of uranium ternary systems to show the development of the above-mentioned satellite.

3. Experimental details

Most of the measurements reported here were done on the newly commissioned U2 beamline at NSLS in Brookhaven National Laboratory using an ERG monochromator. The overall instrument resolution at $h\nu \approx 100$ eV was about 250 meV. Samples were made by arc-melting the constituent materials several times and heat-treating the buttons at 1000 °C under argon atmosphere for about 3 days. X-ray powder patterns confirmed the single-phase nature of each sample. Electropolished parallelepipeds (approximately 3 mm \times 3 mm \times 8 mm) were fractured under vacuum (base pressure about 10^{-10} Torr) to expose a clean surface. All spectra were taken at photon energies of $h\nu = 92$ eV (antiresonance), 94 eV, 99 eV (first resonance) and 108 eV (second resonance). Each sample surface was checked for oxygen contamination by also taking a spectrum at $h\nu = 40$ eV where the oxygen 2p cross-section is substantially larger. No contamination was evident.

A spectrum representative of only 5f emission can be obtained by subtracting the spectrum obtained at the anti-resonance ($h\nu = 92$ eV) where 5f emission is nearly zero [25] from a spectrum at the 5d to 5f Fano resonance ($h\nu = 99$ eV or 108 eV) where two separate channels contribute to a strong 5f intensity. The assumption is made that the cross-sections for the remaining features are not changing dramatically in this range, so that the spectrum for $h\nu = 92$ eV is normalized to be equal to, or lower in intensity than the spectrum for $h\nu = 99$ eV at all electron energies. In this paper the 5f intensities in the figures were obtained from such a subtraction.

The high-resolution data reported here ($\Delta E \approx 0.1$ eV) were taken at low temperatures ($T \approx 20$ K) to reduce the Fermi functions smearing and to

increase the amplitude of any possible Kondo resonance. These data were taken at the Tantalus Synchrotron in Stoughton, Wisconsin. 40 eV photons were used because the resolution of the ERG monochromator is at a maximum while the f-electron cross-section is already substantial at 40 eV.

4. Results and discussion

4.1. High-resolution data

Photoemission spectra taken at about 20 K, with resolution of approximately 0.1 eV and $h\nu = 40$ eV are shown in Fig. 2 for CeAl_3 and CeCu_6 . Similar experimental conditions yielded the data in Fig. 3 for UPT_3 , UBe_{13} and U_2Zn_{17} . Considering first the 4f spectra of Fig. 2 it can be argued that they are basically in agreement with the Kondo model if one identifies the peak at about -2 eV as representing the f hole (or $f^1 \rightarrow f^0$) state created by photoemission while the peak near E_F (not precisely at E_F but rather at -0.3 eV) is identified as a Kondo sideband. The Kondo resonance itself is not observed, presumably because either 20 K is not yet a sufficiently low temperature or most of the already meager spectral weight is actually above E_F . The resolution of 0.1 eV is in any case still insufficient to observe a feature only a few millielectronvolts wide.

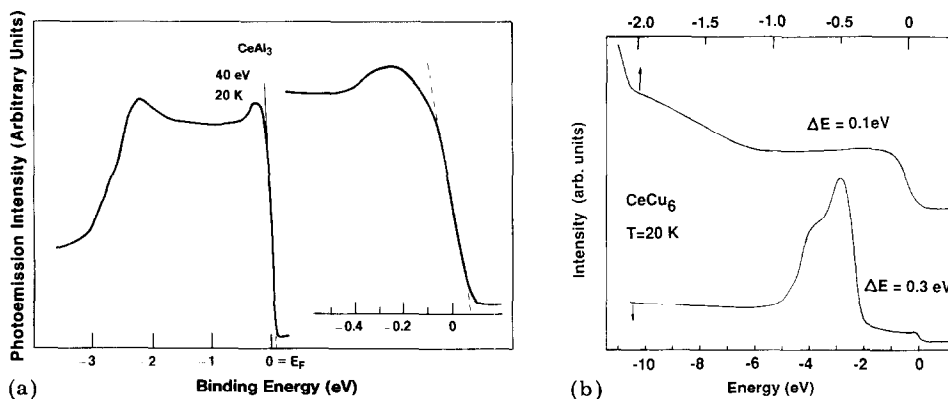


Fig. 2. High-resolution photoemission spectrum for (a) CeAl_3 at $h\nu = 40$ eV and $T = 20$ K; the expanded view near E_F clearly shows that the peak sometimes identified as the Kondo sideband is at -0.28 eV. (b) CeCu_6 , here the f intensity is very weak compared to the strong copper d band (at -3 eV). While difficult to see even in the expanded view (top curve), the “main peak” is at about -1.7 eV while the sideband is at about -0.3 eV. This is much more evident in data taken at resonance but not shown due to space limitations.

Uranium-based heavy fermion compounds, however, exhibit rather sharp features in the photoemission spectra [26] at E_F (see Fig. 3) while the overall spectra tend to be broad but apparently lacking the peak associated with an f hole as in 4f spectra. Depending on how one models the data, the FWHM of the sharp peak at E_F is in the range 150 - 200 meV which makes it

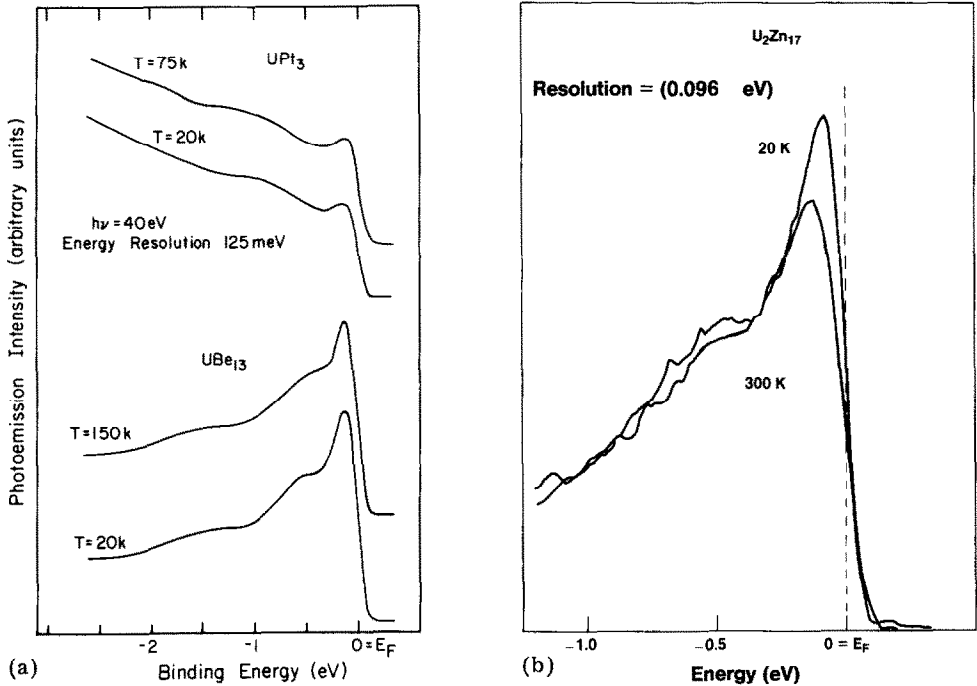


Fig. 3. High-resolution data for (a) UPt_3 and UBe_{13} at several temperatures, note the sharp feature precisely at E_F which we identify with a narrow band structure density of states; (b) U_2Zn_{17} . The sharp feature is still seen at 300 K, thus confirming its band structure origin.

significantly broader than the instrument resolution. The apparent temperature dependence of the intensity is completely accounted for as being due to Fermi function smearing so that in essence there is no temperature dependence associated with this peak. This is obvious in Fig. 3(b) where we see data for U_2Zn_{17} taken at 300 K as well as at 20 K. Only the amplitude within about $3kT$ of E_F is affected in accordance with the shape of the Fermi function.

The obvious question to ask is whether this sharp peak, apparently common to uranium heavy fermion systems, could be a Kondo resonance. In view of the fact that the apparent T_0 for all these heavy fermions is of order 10 K (*i.e.* that $kT_0 \approx 1\text{ meV}$) it is obvious that the observed peak cannot be a Kondo resonance. Moreover, by 20 K one should have only about 20% of the amplitude of the Kondo resonance remaining, based on the Gunnarson-Schönhammer model, and zero amplitude at 300 K. The location of the observed peak almost precisely at E_F also precludes its being a Kondo sideband, since the spin-orbit splitting for uranium is expected to be about 0.8 eV. Additional structure is of course seen at -0.5 eV and at -1.8 eV in UBe_{13} . However, if one interprets this lower structure as reflecting either the f^{n-1} state or the Kondo sideband, then one is still left with the problem of explaining in terms of the Kondo model the peak at E_F which dominates the

spectrum. Moreover, the -1.8 eV peak corresponds to structure obtained in band calculations (see below).

The best explanation for the spectra of Fig. 3 is that the narrow peak at E_F reflects the band structure ground state. Band structure calculations have previously been very successful in representing the electronic structure of uranium compounds. In UIr_3 , for example, the calculated and experimentally derived 5f density of states (from resonant photoemission) agree very well if the calculated spectra are appropriately convoluted with an instrument broadening function as well as a lifetime broadening function whose FWHM increases as $\alpha(E - E_F)^2$.

Thus, at a first glance there appears to be a substantial difference between 4f and 5f photoemission intensities in heavy fermion compounds. The former appear to agree with the Kondo model while the latter point toward a band structure interpretation albeit with far too much intensity in the -1 eV to -2 eV range. We show below that on closer inspection there are in fact substantial similarities between 4f and 5f heavy fermions.

4.2. Resonant photoemission data

The 5f intensity can be separated from the total photoemission spectrum at resonance ($h\nu = 99$ eV) as described above but now with a much poorer resolution which washes out sharp features. This is not a problem since we are now trying to understand the very large width. By doing a systematic set of measurements on a series of intermetallic narrow-band compounds (not necessarily heavy fermions) a clear pattern begins to develop; namely that, as the 5f band becomes narrower, based on bulk property measurements, one obtains more 5f intensity in the -1 to -2 eV range than would be expected from a band structure calculation. The cause is a final state screening satellite, as explained below.

Figure 4(a) shows data obtained on the series URh_3B_x where x ranges from 0.4 to 1.0. The data for $x = 0$ were previously obtained [28] on an instrument with much poorer resolution and are not shown. Instead we show data for UIr_3 , likewise obtained previously, but at resolution comparable to present data. All properties including the low-resolution photoemission data, confirm that this isoelectronic compound is nearly identical to URh_3 . Boron added to URh_3 has the twofold effect of expanding the lattice while also competing with uranium for bonding with the rhodium 4d electrons [28]. Both effects tend to decrease the f-d hybridization which is manifested in Fig. 1(a) by the peak at -2.4 eV. This peak (which we will call the bonding band) clearly decreases in intensity as a function of boron concentration. Extensive band calculations [28] are also consistent with a decrease in the bonding band. At the same time, they show a dramatic increase in the density of states at E_F . Bulk property measurements [29] are consistent with behavior expected from a narrowing within the band regime without heavy-fermion-type renormalizations. The narrowing of the band with increasing boron concentration is also evidenced by the increase in Curie-Weiss-like susceptibility, χ [17]. However, while the bonding peak in Fig. 4(a) is

decreasing in intensity in accord with expectations, the structure at E_F appears to be broadening with boron addition rather than narrowing. A clear shoulder is also developing at about -1 eV. This shoulder appears to be common to nearly all narrow band 5f spectra, including heavy fermions. It is this shoulder, as well as the broadening, which is indicative of a final state 5f satellite (reflecting a localized 5f level).

The case is made substantially stronger in Fig. 4(b) where we show data for UPd_xRh_{3-x} . UPd_3 is the only uranium intermetallic compound where the 5f electrons are known to be localized, as evidenced by neutron diffraction [30] and BIS [31] measurements. This localized behavior is reflected in the photoemission spectrum by the 5f intensity appearing as a broad lorentzian-shaped peak [25] centered at about -1.2 eV and the absence of 5f intensity at E_F . So, in a photoemission spectrum the signature for a localized 5f excitation is a broad peak centered below E_F . Fig. 4(b) also shows the 5f spectra for $UPdRh_2$ and UPd_2Rh . Bulk property measurements [32] show that substitution of palladium for rhodium again results in a dramatic increase in the γ -value. UPd_2Rh meets all the criteria for a heavy fermion system, including the upward curvature in C_p/T vs. T at low T . The 5f spectrum on the other hand is much broader for $x = 2$ than it is for $x = 1$, again in contrast to expectation. We claim that the broadening is again due to the increased intensity of the 5f satellite which can be roughly displayed by simply subtracting the curve for $x = 1$ from that of $x = 2$. The spectrum for $x = 1$ already has a clear shoulder at -1 eV while the bonding peak at -2.5 eV is still clearly in evidence.

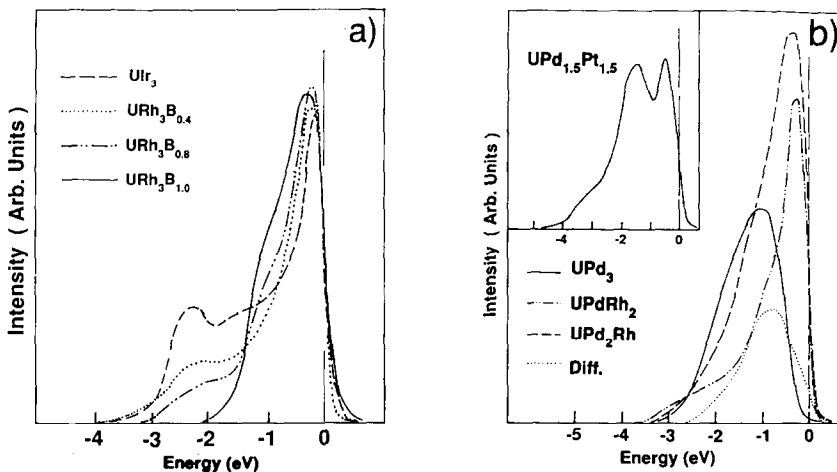


Fig. 4. (a) Resonant 5f photoemission intensity for URh_3B_x (normalized to peak intensity at E_F); data for isoelectronic U_{Ir}_3 has been substituted for that of URh_3 because it is available at the resolution comparable to materials containing boron. (b) 5f intensity for UPd_xRh_{3-x} alloys, UPd_3 has no 5f intensity at E_F ; the difference curve is the subtraction of the $UPdRh_2$ data from the UPd_2Rh data. The inset shows 5f intensity for $UP_{1.5}Pt_{1.5}$; note the very clear "two-peaked" structure. (Normalized to mesh current.)

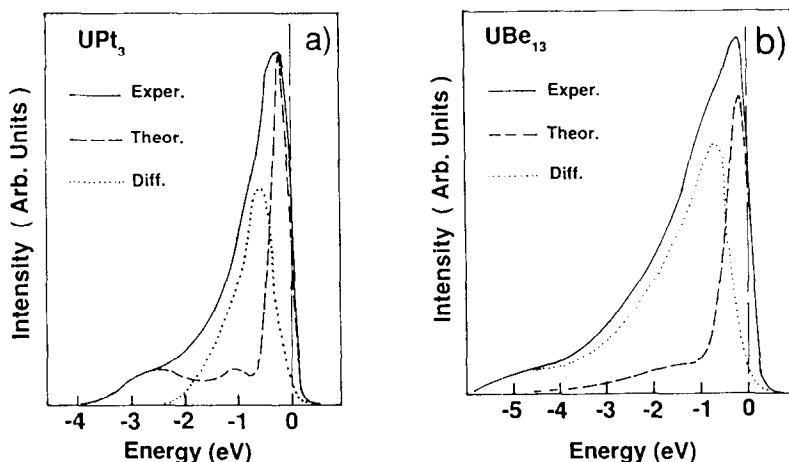


Fig. 5. Experimental 5f intensity compared to a broadened partial 5f density of states (a) for UPt_3 [34], (b) for UBe_{13} [33]. Here the difference curve does not have a lorentzian shape; the extra intensity up to -5 eV is probably due to uranium 6d.

The inset in Fig. 4(b) shows the 5f spectrum obtained for $UPd_{1.5}Pt_{1.5}$. In this material one can clearly distinguish the localized satellite peak since it is of equal intensity with the band structure peak and shifted to slightly higher binding energy (-1.5 eV). No bulk property data exist for this material, but on the basis of the above observations it should have an extremely narrow band at E_F , or perhaps even be completely localized.

These data clearly indicate that the 5f photoemission spectra consist of a superposition of a spectrum expected from a band-like 5f system plus a broad peak (approximately 1 eV FWHM) whose intensity appears to be a function of the inverse of the bandwidth. To test whether this holds for well-known heavy fermion systems as well, we re-examine the spectra for UBe_{13} [3] and UPt_3 [2], two systems for which band calculations exist. The comparison of experiment with the calculated density of states is shown in Fig. 5 where the 5f density of states for UBe_{13} [33] and UPt_3 [34] was broadened by 0.3 eV wide gaussian and then by an energy-dependent lorentzian to simulate lifetime broadening. The difference curves obtained by subtracting the broadened calculated density of states from experiment are strongly suggestive of the localized 5f satellite discussed above. In UBe_{13} there appears to be more structure on the high binding energy side (approximately 2 - 5 eV) than one would expect just from a 5f density of states. The extra structure is in fact quite similar to what is predicted [33] for uranium 6d density. Since there is no *a priori* reason to exclude uranium 6d from the resonance curve, we tentatively assign the extra intensity below -2 eV to uranium 6d emission. Thus we again have a superposition of band structure and a 5f satellite.

If we equate the 5f satellite with the so-called "main peak" observed in cerium compounds (at -2 eV) we obtain the desired systematics between 4f

and 5f heavy fermions and indeed most 4f and 5f narrow-band compounds. The obvious question one must ask then is whether the interpretations of 4f photoemission spectra might not also be done along the lines of final state screening as opposed to the Gunnarson-Schönhammer model, in order to make the comparison complete. After all, a similar interpretation [35] is used to explain the -6 eV satellite in nickel and has been used by Hüffner [15] to explain the “two-peaked” structure not only in cerium but also heavier lanthanides. The problem arises from the fact that in Ce-based systems the peak associated with f-screening near E_F is actually observed at -0.3 eV rather than at E_F . This cannot be a band structure ground state since in the latter case the 4f intensity would be fixed at E_F . The problem can be resolved if we realize [36] that the localized screening f level will actually be spin-orbit split. In an excitation there is a finite probability of occupying either the $f^{5/2}$ (ground state) or $f^{7/2}$ level. If the screening state is $f^{7/2}$, then one does not recover the initial ground state and the photoemission peak will be found below E_F . With this added complication, then, there is complete agreement between 4f and 5f heavy fermion spectra.

We wish to point out that these data are significant toward a proper interpretation of photoemission spectra in heavy fermion systems as well as all narrow-band systems exhibiting substantial correlation effects. They do not, however, conclusively select between the various models proposed for the heavy fermion state (band structure and Kondo resonance). Indeed, it may be yet possible to transform from one model to the other. Rather we should note that the existence of the 5f satellite strengthens the systematic comparison of the rare earths and actinides. Further, the slow build-up of the satellite intensity with band narrowing supports the assertion that the heavy fermion state constitutes perhaps only the extreme edge of a smooth transition from itinerant to localized electrons.

We thus conclude that: (1) The “two-peaked” structure commonly observed in cerium-based compounds is manifested in 5f systems as well; (ii) The failure to recognize it previously is due to the broad nature of the satellite and its proximity to E_F ; (iii) The spectra from narrow-band 5f systems are representative of those expected from a band structure calculation with a localized 5f satellite superimposed. We further suggest that: (i) The photoemission spectra for cerium-based heavy fermions be examined more seriously in terms of screened states. (ii) Heavy fermion behavior arises from a ground state yet describable by band structure techniques but with strong low lying excitations of a local character (*e.g.* spin fluctuations).

Acknowledgments

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