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Journal

Journal of Magnetism and Magnetic Materials, 108(1-3)

ISSN

0304-8853

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Publication Date

1992-02-01

DOI

10.1016/0304-8853(92)91417-r

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Peer reviewed

Photoemission in YbCu$_2$Si$_2$: problems with the Kondo impurity model

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We report valence band photoemission results for YbCu$_2$Si$_2$. The 4f^{13}($J = 7/2$) final state peak, centered 60 meV below the Fermi level ϵ_F, lacks the temperature dependence and is broader than predicted for a Kondo resonance. Together with recent photoemission results for cerium compounds, these results raise serious doubts about the Kondo impurity explanation of heavy fermion photoemission.

The prevailing theory [1,2] of photoemission in heavy fermion compounds is the Kondo impurity theory in the $1/N$ approximation. This approach predicts that a resonance should appear in the spectrum, located (for cerium) a distance T_K above the Fermi level ϵ_F (where T_K is the Kondo temperature) with a spectral weight proportional T_K; the resonance should vanish as the temperature increase above T_K. We have recently shown [3] for a series of cerium compounds that there is no correlation between the spectral weights observed in the 4f peaks near ϵ_F and the known Kondo temperatures; furthermore the temperature dependence is negligible. This suggests that the spectra are not related to a Kondo resonance.

A problem of cerium compounds is that the resonance is expected to lie *above* ϵ_F; hence, only the tail below ϵ_F can be observed in photoemission. For ytterbium compounds the resonance should be *below* ϵ_F, and hence is directly observable [2]. In this paper we report results for YbCu$_2$Si$_2$, which has a Kondo temperature of order 50 K as deduced from specific heat [4] or quasielastic neutron linewidth [5]. While a peak which might be identified as a Kondo resonance is observed, our main results are that we find *no* temperature dependence over a temperature interval [10,300 K], and that the peak is substantially broader than predicted by the Kondo theory.

Experiments were carried out at the Los Alamos U3C beamline at NSLS with supporting work at U. Wisconsin's SRC using an HA-50 hemispherical analyzer and ERG monochromators. The total resolution varied from 60 to 180 meV depending on photon energy. Flux grown single crystals were cleaved in UHV (7 × 10^{-11} Torr). Further details are described elsewhere [3].

The spectrum at $h\nu = 182$ eV is shown in fig. 1. The large peak near 4.5 eV is due to Cu emission. The peaks in the interval 5–12 eV arise from bulk 4f$^{13} \rightarrow$

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4f^{12} transitions while the two peaks within 3 eV of the Fermi energy arise from 4f$^{14} \rightarrow$ 4f^{13} transitions, with the 4f$_{7/2}$ final state closest to ϵ_F and the 4f$_{5/2}$ transition at approximately 1.5 eV binding energy [6]. Also included in fig. 1 is a high resolution spectrum taken at $h\nu = 50$ eV. This shows that the 4f^{14} spectrum consisits of bulk and surface doublets, with the latter at higher binding energy. From the ratio of the 4f^{14} and 4f^{13} bulk transitions we estimate [6] that the Yb valence is 2.8–2.9 (the estimate varies somewhat with photon energy).

In the language of Kondo theory [2] the bulk 4f$^{14} \rightarrow$ 4f^{13} transitions are the Kondo resonance (4f$_{7/2}$, nearest ϵ_F) and a spin–orbit sideband (4f$_{5/2}$, at 1.4 eV). These should be highly temperature dependent on the scale of the Kondo temperature, 50 K. To test for this we show in fig. 2 high resolution spectra taken at $h\nu = 60$ eV for $T = 10$ and 300 K. (The spectra were
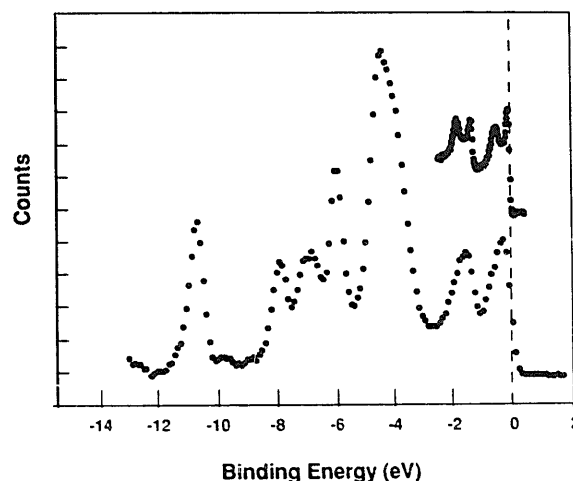

Fig. 1. Photoemission spectra of YbCu$_2$Si$_2$ taken at $h\nu = 182$ eV with 180 meV resolution for $T = 10$ K. The inset, taken at $h\nu = 50$ eV with 60 meV resolution, resolves the 4f^{13} final state into surface and bulk spin–orbit doublets. The peaks from 5 to 12 eV represent the 4f^{12} final state. The valence estimated from this plot is 2.9.
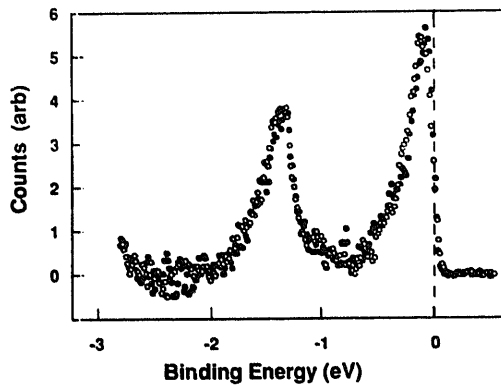
Fig. 2. The 4f¹³ final state doublet at $T = 10$ K (●) and $T = 300$ k (○). The photon energy is 60 eV; the resolution is 90 meV. The surface doublet has been fit to two Gaussians and subtracted out. No temperature dependence is observed.

Fig. 3. The 4f¹³ $(J = 7/2)$ f⁻ tate peak, measured at $hv = 60$ eV with 60 meV reso. .tion and at $T = 20$ K. The dashed curve in both the bottom and top plots is a convolution of the reported crystal field sidebands with instrumental resolution. The data are substantially broader than this fit.

normalized to the Cu emission peak, which should be temperature independent; the surface doublet was fitted to a pair of Gaussians and subtracted off.) It is clear that the temperature dependence is negligible.

A complication [2] arises from the known existence [7] of crystal fields in YbCu₂Si₂. The level scheme as deduced from neutron scattering [7] is shown in fig. 3. The ground state doublet has a width 4 meV. Excited state doublets at 12, 30 and 80 meV have neutron linewidths of order 8 meV; this width is believed to arise from Kondo scattering, and it gives the scale on which the crystal field Kondo sidebands should renormalize to zero [2]. The observed 4f₇/₂ peak near ϵ_F should be the sum of these sidebands, convoluted with instrumental resolution, as shown in fig. 3. Direct comparison to our data for figs. 2 and 3 demonstrates that there is no temperature dependence, neither on the scale of the ground state Kondo temperature, nor on the scale of the sideband Kondo temperature (100 K); secondly the observed emission peak is substantially broader than the peak predicted by convoluting the sideband peaks with instrumental resolution.

The prediction of Kondo impurity theory that the Kondo resonance should renormalize to zero implies that the valence should approach the value 3 at high temperature [2]. Our results seem to imply that the valence remains constant. Perhaps in the Kondo lattice the valence is stabilized. Alternatively, photoemission may be incorrectly measuring valence, perhaps due to final state screening effects. In either case it is clear that the Kondo impurity theory alone cannot explain the valence band photoemission results. Together with our results [3] for cerium compounds, this suggests serious problems for the Kondo impurity theory of heavy fermion photoemission.
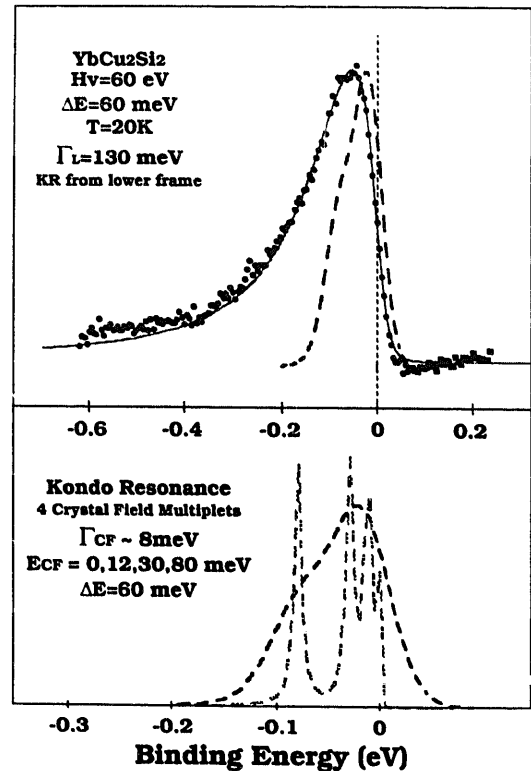
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