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Authors

Andrews, AB
Blyth, RIR
Arko, AJ
et al.

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Temperature dependence of 4f PES features and hole occupancy in ytterbium heavy fermions

A.B. Andrews^{a,*}, R.I.R. Blyth^a, A.J. Arko^a, J.J. Joyce^a, Z. Fisk^a, J.D. Thompson^a,
R.J. Bartlett^a, P.C. Canfield^b, C.G. Olson^b, P. Benning^b, P.S. Riseborough^c

^a Los Alamos National Laboratory, Los Alamos, NM 87545, USA

^b Iowa State University, Ames, Iowa, USA

^c Polytechnic University, Brooklyn, New York, USA

Abstract

We have measured the temperature dependence between 80 and 300 K and spectral weight of the 4f features in clean single crystal Yb heavy fermions, cleaved in a vacuum of 5×10^{-11} Torr. A small ($\sim 15\%$) decrease in spectral weight differs substantially from the reported $\sim 50\%$ decrease over this range. All divalent features are much larger than expected, yielding n_f values far too small. The intensity of the divalent features in YbAuCu₄ is about two orders of magnitude larger than expected.

Although it is widely accepted that the 4f photoelectron spectra (PES) of heavy fermions are well described by the Gunnarsson and Schonhammer [1] (GS) and NCA [2] methodology, we have found previously [3] that there are numerous inconsistencies. In particular, the width of the near Σ_F feature (termed the Kondo resonance, KR) is much too broad and nearly constant with T_K (the Kondo temperature). Since it is this width which sets the low-temperature scale, this constancy is disturbing. In addition the spectral weights, positions in energy, temperature dependencies, etc., all appear to be independent of T_K . In this paper we report a careful temperature dependence study as well as spectral weights of the bulk divalent features in Yb compounds, and their relation to the hole occupancy, n_f .

Determining the temperature dependence of features in a PES spectrum can be difficult, owing to the numerous possible temperature effects. The only convincing result is a null effect which would indicate that all other

effects such as surface reconstructions, lattice parameter changes, phonons, oxidation, photon flux drift, sample position changes, problems associated with fitting, etc., are not active. Toward this end we have previously shown [4] that between 20 and 80 K no temperature effect whatsoever is seen in YbAl₃, YbCuSi₂, and YbAgCu₄, materials with T_K 's of 400, 35 and 100 K, respectively. These results differ sharply from recent data on scraped polycrystalline samples [5, 6] in which a large temperature dependence of the KR is reported below 80 K. In addition we found no temperature dependence to 300 K in the first two materials, but these had slightly contaminated surfaces during the measurement.

A minor variation in our technique has allowed us to measure clean single crystal surfaces between 300 and 80 K. We report on these in the present paper. The measurements were made at the NSLS Los Alamos U3C beamline. Samples were cleaved in situ in a vacuum of 5×10^{-11} Torr, while the temperature was maintained either at 300 K or at 80 K with liquid-nitrogen cooling a cold finger. The spectra reported here were normalized

*Corresponding author.

to photon flux only, taking into account the decaying beam during measurement. The correctness of the normalization is evident in the fact that the Al-2p or the Si-2p core levels in YbAl₃ or YbCu₂Si₂, respectively, show no amplitude variation with temperature. A photon energy of 120 eV was used for YbAl₃ since the 4f cross-section is here at a maximum and the Al-2p line from third-order light fortuitously falls in the spectrum. To obtain a similar effect with the Si-2p levels in YbCu₂Si₂ it was necessary to use 104 eV photons.

In Fig. 1(a) we show the mesh-normalized raw spectra of YbCu₂Si₂ taken at 80 and 300 K. There is a small but clearly evident temperature dependence of the so-called KR. To obtain a better determination of the spectral weight change, it was necessary to (i) subtract the secondary-electron spectrum via an iterative procedure, (ii) subtract off the non-4f features in the spectra, (iii) remove the surface-related 4f features at -1 and -2.3 eV. The non-4f portion of the spectrum was obtained by measuring the isostructural LuCu₂Si₂ and normalizing to the Cu-3d bands. The remaining near- E_F divalent portion of the spectrum was then fit with Gaussians for the surface states and asymmetric Lorentzians for the bulk 4f peaks using a least-squares minimization procedure. The Gaussian surface states were then subtracted off. The resulting spectrum is representative of bulk 4f features only (both divalent and trivalent) and is shown in Fig. 1(b) for YbCu₂Si₂, and, after a similar procedure, in Fig. 1(c) for YbAl₃. (In this latter spectrum we have retained in the spectrum the Al-2p features to emphasize the excellent normalization.)

Several things can be noted in the reduced spectra. In the first place, this most careful measurement of the temperature dependence (but a cursory fit) of the KR yields a spectral weight change for the KR which is no more than $\approx 15\%$ for both materials. NCA theory would predict [6] an $\approx 50\%$ decrease between 80 and 300 K. The recent claims [6] of exact agreement with NCA in YbAl₃ appear to be an artifact of an incorrect background subtraction as well as incomplete fitting. The FWHM of the KR is of the order of 160 meV in both materials, nearly 1.5 times greater than the expectation at our resolution of 90 meV, and nearly 3 times the unbroadened NCA prediction, even after accounting for the crystal field states in YbCu₂Si₂. Moreover, direct integration of the spectral weights of the divalent and trivalent features yields a valence of 2.60 ± 0.05 for YbAl₃ and 2.55 ± 0.05 for YbCu₂Si₂ (thus n_f equals 0.60 and 0.55, respectively). The n_f value for YbAl₃ is in agreement with Cho et al. [7], but in strong disagreement with Tjeng et al. [6], and with X-ray absorption. Although the temperature dependence of the trivalent portion of the spectrum is difficult to measure (it is no more than $\sim 5\%$ to 10%), we nonetheless performed a consistency check via

Fig. 1. (a) YbCu₂Si₂ PES spectra at 80 and 300 K, normalized to photon flux. The Si-2p line about E_F is from second order. (b) Only bulk 4f PES features of YbCu₂Si₂ spectra (see text for subtraction procedure) ratio of divalent to trivalent intensities yields $n_f = 0.55$. (c) Bulk 4f PES features in YbAl₃ with non-bulk-f features subtracted. The Al-2p line is from third order. Here $n_f = 0.67$.

the method of Tjeng et al. [6], using the combination of divalent and trivalent temperature dependencies. The values of n_f obtained in this way are consistent with the direct integration of the divalent and trivalent intensities.

Thus, while a small but definite temperature dependence of the so-called KR is observed, it is by no means clear that it confirms the correctness or even applicability of NCA, particularly since no predicted parameter is experimentally reproduced. It is especially telling that the n_f for YbCu₂Si₂ ($T_K = 35$ K) is smaller than that of YbAl₃ ($T_K = 400$ K). Moreover, both materials have n_f values more correctly associated with the mixed valence regime rather than the Kondo regime. It would thus

Fig. 2. (a) Wide scan PES spectra for YbAgCu₄ ($T_K = 100$ K) and YbAuCu₄ ($T_K \approx 0.5$ K) normalized to each other at the Cu-3d peak. (b) Blow-up of the near- E_F region. The intensity of the YbAuCu₄ 4f features is about 2 orders of magnitude larger than NCA predictions.

appear that the spectral weight of the KR in Yb compounds is again independent of T_K . We suggest that the small temperature dependence could well be accounted for by a change in valence due to lattice parameter changes. In addition, the comparable Lutetium compounds display a 5% decrease in intensity as the temperature increases from 80 to 300 K, thus suggesting that phonons cannot be ruled out.

In case there remains doubt regarding the inconsistencies with GS and NCA we show in Fig. 2 the spectra for YbAgCu₄ and YbAuCu₄ ($T_K \approx 100$ and ≈ 0.5 K, respectively [8]). Recently, Weibel et al. [5] have reported a 50% decrease of the KR between 20 and 80 K on scraped samples of YbAgCu₄, while we consistently

obtain a null result in this temperature range on cleaved polycrystalline samples. All data shown in Fig. 2 are taken at 80 K. We now call attention to the data for YbAuCu₄, whose crystal field levels are presumed [5] identical to YbAgCu₄. A GS calculation at $T = 0$ indicates that the spectral weights of the KRs in the two materials should be in the ratio of 1:40. A glance at Fig. 2(b) shows them to be about 1:2, a discrepancy of a factor 20. When one factors in the measuring temperature of 80 K, which represents approximately T_K for the Ag material but as much as $\sim 160 T_K$ for the Au material (the KR should have been renormalized to near-zero based on NCA), the discrepancy between predicted and measured spectral weights of the KR in YbAuCu₄ exceeds two orders of magnitude. The natural linewidth is likewise of the order 100 meV, or 2 orders of magnitude too large.

We conclude that in Yb compounds the small temperature dependence does not reproduce the NCA predictions, while the spectral weights and widths are independent of T_K , reminiscent of Ce compounds. The hole occupancy, n_f , is small and does not scale with T_K .

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