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Spectroscopic Evidence for the $5f$ Coulomb Interaction in UAl_2 and UPt_3

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The $5f$ spectral weight in the heavy-fermion materials UAl_2 and UPt_3 has been measured by electron spectroscopy, and compared to that in density-functional calculations. The one-electron $5f$ widths are both too *large* to account for the enhanced values of the specific-heat coefficient γ , and too *small* to account for the measured widths. In the Anderson Hamiltonian, both discrepancies arise from the $5f$ Coulomb interaction. It is pointed out that an enhanced γ value does not imply enhanced magnitude of the $5f$ spectral weight at E_F .

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A number of materials containing Ce and U have very large low-temperature specific heats, leading to the descriptive term "heavy fermion."¹ The ground states vary from magnetic to superconducting, and it is generally agreed that the Ce and U f electrons give rise to these novel and variable properties. A central assumption of current many-body approaches^{2,3} to these materials is the existence of an f Coulomb repulsion U_{ff} which is larger than the one-electron f width. For cerium materials this is well established^{4,5} by the observation of separate peaks split by U_{ff} in combined photoemission (PES) and bremsstrahlung isochromat (BIS) spectra, which measure the f spectral weight of the single-particle Green's function. For metallic uranium materials the situation is much different. Typically, separated peaks are not observed and in uranium metal,⁶ for example, the total observed f width is comparable to the one-electron width obtained in band calculations. For only one metallic uranium material has spectroscopic evidence for U_{ff} been reported, and this material is not of the heavy-fermion type.⁷ Thus far UBe_{13} is the only heavy-fermion uranium material for which a complete BIS-PES $5f$ spectrum has been published.⁸ The total $5f$ width was found to be about the same as in uranium metal, i.e., much too large to relate to the large specific heat. No conclusion about the relative roles of one-electron and Coulomb interactions was reached.

In this paper we report combined BIS-PES spectra for the heavy-fermion uranium materials UAl_2 and

UPt_3 . The specific heats of both materials^{9,10} have a large T -linear coefficient γ , and a $T^3 \ln T$ term often taken⁹⁻¹³ to imply spin fluctuations. Additionally, UPt_3 is a superconductor for which triplet Cooper pairing has been speculated.¹⁰ We find BIS spectra which are more structured than found for UBe_{13} , and can compare our $5f$ spectral weights to density-functional calculations^{14,15} very recently available for these two materials. These calculations provide a one-electron fiduciary for the interpreting of the observed $5f$ widths, leading to an interesting conclusion not given previously, that the one-electron $5f$ bandwidths are, on the one hand, too *large* to account for the large γ values, but are, on the other hand, too *small* to account for the measured $5f$ widths. Appealing to existing theoretical results for the Anderson Hamiltonian, we argue below that both these apparently contradictory effects arise simultaneously from the Coulomb interaction U_{ff} .

BIS and PES spectra were obtained at a photon energy of 1486.6 eV with use of a Vacuum Generators ESCALAB, operated under conditions to yield resolutions of 0.5 and 0.8 eV, respectively. Clean sample surfaces were obtained by the fracturing of polycrystalline ingots in a vacuum of 5×10^{-11} Torr, after which the sample was transferred under the same vacuum to the measurement position and cooled to ~ 100 K. Valence-band and core-level PES were then measured, followed by BIS. During data taking the chamber pressure was always below 7×10^{-11} Torr for x-ray pho-

toelectron spectroscopy and 1×10^{-10} Torr for BIS. Subsequently the PES and BIS measurements were repeated at room temperature, with no significant differences in the data.

Room-temperature PES measurements with variable photon energy were performed at the Stanford Synchrotron Radiation Laboratory, using beam line I-1 for UPt_3 and beam line III-1 for UAl_2 . The UPt_3 sample was the same as that used for the PES-BIS measurements, while the UAl_2 sample was a small unoriented single crystal. Both samples were fractured and measured in a vacuum of 8×10^{-11} Torr, in a vacuum chamber equipped with a commercial cylindrical-mirror analyzer to measure electron energies. The resolution, which is determined almost entirely by the electron analyzer for the photon energies used, was 0.4 eV.

Figures 1 and 2 show resonant photoemission and BIS spectra for UAl_2 and UPt_3 , respectively, with their inelastic backgrounds removed by a standard method.¹⁶ The PES spectra for photon energy $h\nu = 92$ eV have the U $5f$ emission resonantly suppressed¹⁷ and thereby reveal Al $3s-3p$ or Pt $5d$ emission, plus some U $5d$ emission which is only partly suppressed by the resonance. The PES spectra labeled U $5f$ result from subtraction of the $h\nu = 92$ -eV spectrum from that for $h\nu = 98$ eV, a photon energy for which the U $5f$ emission is resonantly enhanced.¹⁷ For UPt_3 the two spectra were scaled to match below -5 eV before subtracting. The results are generally consistent with previous resonant¹⁸ and x-ray¹⁹ PES on UPt_3 and uv PES on UAl_2 .²⁰ The portions above E_F are BIS data taken with $h\nu = 1486.6$ eV, for which the U $5f$ spectral weight is known⁸ to dominate. The BIS and U $5f$ PES spectra have been scaled to reproduce the n_f values, ~ 2.5 for both materials, obtained from the density-

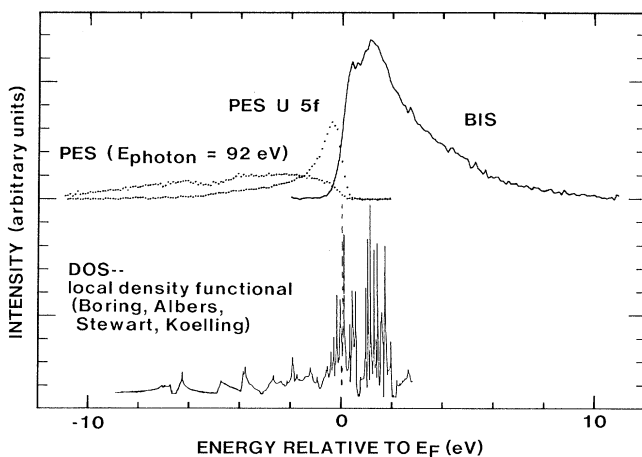


FIG. 1. Top: PES and BIS spectra for UAl_2 , as described in the text. Bottom: local density-functional density of states from Ref. 14.

functional calculations,^{14,15} which are expected to be accurate for this average ground-state property. The PES spectra are similar to one another and to those of UIr_3B_2 ¹⁶ and UBe_{13} ,^{8,17} while the two BIS spectra differ from one another in how many peaks occur above E_F . They also differ from that reported⁸ for UBe_{13} in that both show very clearly, in addition to the slope change marking the onset of E_F , a second slope change which occurs after a distinct plateau at ~ 0.5 eV, and is followed by a very sharp rise to the peak at ~ 1 eV.

For comparison with our data, Figs. 1 and 2 also show the results of recent density-functional calculations of the electronic structure of UAl_2 ¹⁴ and UPt_3 .¹⁵ From partial densities of states, one finds for UAl_2 that the U f weight is almost entirely confined between -1 and 2 eV, the U d weight occurs in this region and extends further below E_F to -3 eV, and the weight between -3 and -10 eV is of Al $s-p$ origin. For UPt_3 , the U-site weight from -0.5 to 1.5 eV is dominated by f states, the weight from -0.5 to -4.5 is

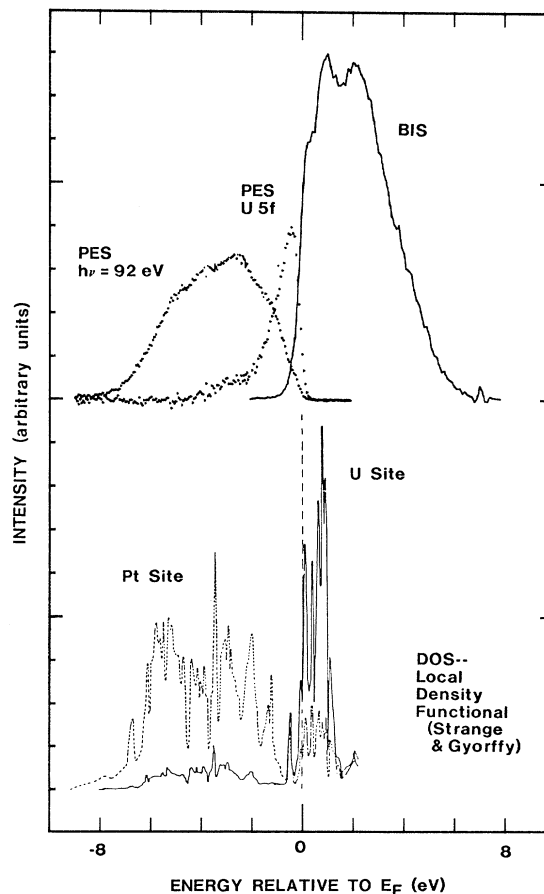


FIG. 2. Top: PES and BIS spectra for UPt_3 , as described in the text. Bottom: local density-functional density of states from Ref. 15.

about equally d and f states, and the weight between -4.5 and -7 eV is dominantly d states. The Pt-site weight is dominantly d everywhere, and relative to Pt metal, the empty and filled Pt $5d$ states are split off above and below E_F to reduce greatly the Pt weight at E_F . There is substantial U-Pt and U-Al hybridization. A sharp contrast with the calculated f bands of uranium metal is provided by the sharpness of the various subbands, the clear spin-orbit separation into spin- $\frac{5}{2}$ and $-\frac{7}{2}$ groups, and the sharp cutoffs above E_F at 1.5 and 2 eV for the two materials. It is an important point that the theoretical curves extend above E_F far enough to show all the $5f$ structure.

Many features of the density-functional results are seen in the experimental spectra. For UPt_3 the $h\nu = 92$ -eV spectrum shows a small Pt weight at E_F , and the calculated width, if not the detailed shape, of the Pt states. U $5f$ weight appears at the top of the Pt $5d$ band, a predicted consequence of U-Pt hybridization. For UAl_2 the $h\nu = 92$ -eV spectrum shows the calculated Al s - p states, including the dip at -5 eV, and the U d states nearer E_F . The spin-orbit splitting of the U $5f$ bands suggests a spin-orbit origin of the 0.5-eV shoulder and 1-eV peak found in the BIS spectra of both materials.

The most striking and informative feature of Figs. 1 and 2 is that the measured widths of the $5f$ spectral weights greatly exceed the one-electron ones. Above E_F the BIS spectra show very much weight over an energy range as great as 6 eV above the $5f$ cutoffs, and below the E_F the PES spectra show much more weight in the -1 to -2 -eV region than is predicted or can be explained by the experimental resolution. Indeed, recent high-resolution PES spectra²¹ of UBe_{13} and UPt_3 , which show much sharpening of the $5f$ structure, nonetheless have much $5f$ weight between -1 and -2 eV.

Although direct comparison to our spectra implies that the one-electron $5f$ widths are too *small*, comparison of calculated and experimental γ values suggests^{14,15} that the one-electron $5f$ widths are instead too *large*, in that the experimental γ values and the relation $\gamma = \pi^2 k_B^2 N(E_F)/3$ imply values of $N(E_F)$, the density of states at E_F , which are much larger than calculated, in spite of the sharpness of the subband structure. As described below, this situation occurs for the Anderson Hamiltonian because of U_{ff} , which provides the basic mechanism for enhancing the γ values so dramatically and at the same time increases the total spreading of the $5f$ weight by displacing energetically unfavorable $5f$ valence states away from E_F . From Figs. 1 and 2 a conservative lower limit on U_{ff} is ~ 1.5 eV, consistent with previous theoretical²² estimates, $U_{ff} \sim 2$ eV, for metallic uranium.

Given the large U-U separations (3.38 and 4.1 Å for UAl_2 and UPt_3 , respectively) the N_f -fold degenerate

Anderson Hamiltonian is most widely used to model the interplay of one-electron and Coulomb interactions in these materials. Theories for the lattice case are still in progress,^{3,23} but for an impurity^{24,25} with the local orbital occupation $n_f < 1$, and in the limit of large degeneracy and Coulomb interaction, the situation described qualitatively above is well understood quantitatively, as follows. For $U_{ff} = 0$ the Green's-function spectral weight $\zeta(E)$ is a single Lorentzian of degeneracy N_f and one-electron hybridization width Δ , centered above the Fermi level such that $\zeta(E_F) = \pi n_f^2 / N_f \Delta$. This is the impurity analog, with neglect of spin-orbit coupling, of the density-functional results. For the same value of n_f , but with U_{ff} large, $\zeta(E)$ spreads over a range $\sim U_{ff}$, leaving around E_F only the Kondo resonance, having a reduced width approximating the Kondo temperature $\delta \ll \Delta$.²⁴ A remarkable consequence²⁶ of the Friedel sum rule is that $\zeta(E_F)$ has the same value, for fixed n_f , as for $U_{ff} = 0$. The usual expression above for γ continues to hold, if $N(E_F)$ is taken as the quasiparticle density of states, given by $N(E_F) = Z^{-1} \zeta(E_F)$. $Z^{-1} = 1$ for $U_{ff} = 0$, but for infinite U_{ff} , and to order $(1/N_f)^0$, one has^{24,25} $Z^{-1} = N_f \Delta / \pi n_f \delta \gg 1$, leading to an enhanced $\gamma = \pi^2 k_B^2 n_f / 3\delta$. Thus the width Δ is too large to account for the enhanced specific heat, but is too small to account for the total f width. The enhanced γ value is reflected in the PES-BIS spectrum only by the small width δ of features near E_F , but not by a large magnitude of $\zeta(E_F)$,²⁷ contrary to commonly stated^{17,18,28} expectations. Typically the BIS-PES experimental resolution is inadequate to measure δ directly so that $N(E_F)$ can be obtained from BIS-PES spectra only by use of theoretical relations²⁴ between δ and the large-energy-scale features of the spectrum, such as U_{ff} .

For cerium materials, the theoretical results for the impurity Anderson Hamiltonian have led to a unified interpretation^{5,24,29,30} of much of the large-energy-scale spectroscopic and low-energy-scale transport data, apart from lattice-coherence effects. A major barrier, that^{5,18} measured BIS-PES $4f$ spectra often have more weight near E_F than expected from the Kondo resonance, has been removed by very recent theories which find that this weight arises from spin-orbit sidebands^{30,31} of the Kondo resonance, and from f^2 valence states present for finite U_{ff} .³² These results for cerium are encouraging that the substantial weight near E_F in our uranium spectra can also be accounted for with the Anderson Hamiltonian. For example, the expectation of spin-orbit sidebands provides an explanation of the spectral weight occurring above and below E_F at the uranium spin-orbit energy. More generally, the sideband spectrum around E_F , as well as the relation between high and low energy scales, should reflect the details of the quantum struc-

ture of the relevant valence states, and so a final judgment must await explicit calculations in the regime $n_f \sim 2.5$, for which the relevant valence states, $n_f = 2, 3$, and 4 , have a quantum structure richer than just spin-orbit splitting.

To summarize, the one-electron $5f$ bandwidths for UAl_2 and UPt_3 are too large to account for the large γ values and too small to account for the measured BIS-PES widths, precisely the situation that occurs in the Anderson Hamiltonian as a result of U_{ff} . Success with cerium gives hope that the situation can be modeled quantitatively by the Anderson Hamiltonian when the theory is extended to treat larger numbers of f electrons.

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