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**Authors**
Lassailly, Y
Allen, JW
Ellis, W
et al.

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# ELECTRON SPECTROSCOPY STUDY OF THE HEAVY FERMION COMPOUND U$_2$Zn$_{17}$

Y. LASSAILLY†#, J.W. ALLEN†, W. ELLIS+, L. COX+, B. PATE*, Z. FISK+
and I. LINDAU*

†Xerox PARC, 3333 Coyote Hill Road, Palo Alto, CA 94304, USA
+Los Alamos National Laboratory, Los Alamos, NM 87545, USA
*Stanford Synchrotron Radiation Laboratory, Stanford, CA 94305, USA
#CRTBT, C N R S, BP166, 38042 Grenoble, France

We report X-ray photoemission, resonant photoemission and Bremsstrahlung isochromat spectra of the 4f core levels, the valence band and the conduction band, respectively, of the heavy fermion compound U$_2$Zn$_{17}$ and the reference compound Th$_2$Zn$_{17}$, and we assess current efforts to interpret such spectra

The low temperature properties of cerium and uranium materials have much in common, including a diversity of ground states varying from magnetic to superconducting, and the occurrence in some materials of unusually large specific heats, the so-called 'heavy fermion' behavior [1] It is generally agreed that the Ce and U 4f and 5f electrons, respectively, are responsible for these properties The 3d X-ray photoemission (XPS) and 4f photoemission (PES) and Bremsstrahlung isochromat (BIS) spectra of cerium materials have been interpreted with considerable success using the impurity Anderson Hamiltonian, leading to a new unified picture [2–5] of those spectroscopic and low temperature properties that do not involve lattice coherence effects In this picture the low temperature properties are determined by spin fluctuations with a characteristic energy scale set by the Kondo temperature $T_K$, and it is assumed that coherence effects set in at some temperature lower than $T_K$

For uranium materials, no such unified picture exists Hill introduced the idea that the 5f electrons are localized or itinerant depending on the U–U separation, which one might model with the Hubbard Hamiltonian But heavy fermion materials typically have large U–U separations putting them in the localized regime of the Hill plot and yet can display low temperature itinerant properties This and other results, such as the effect [6] of substitutions on Zn sites in U$_2$Zn$_{17}$, suggest that the important factor is hybridization with neighboring atoms, which could be modeled

with the Anderson Hamiltonian However, the 5f spectra do not resemble the cerium 4f spectra as measured or as calculated from the impurity Anderson Hamiltonian, particularly in having much more f-weight around the Fermi energy $E_F$ Also, as we have pointed out, the only manifestation of the Coulomb energy $U$, which must be present for either the Anderson or Hubbard Hamiltonians, is that the total measured 5f width greatly exceeds the one-electron 5f width obtained in density functional calculations [7]

There are two important differences between uranium and cerium The U 5f wavefunction is more extended than the Ce 4f wavefunction so the hybridization may be larger and the screened Coulomb interaction smaller In addition, the valence states for uranium are 5f$^2$, 5f$^3$ and 5f$^4$, so multiplet effects will be much more important than for cerium with valence states 4f$^0$, 4f$^1$ and 4f$^2$ Mechanisms have been suggested whereby these effects may lead to a negative effective Coulomb interaction [8], or to the greatly increased 5f weight around $E_F$ [7] A realistic treatment of the impurity Anderson Hamiltonian accurately including these effects for uranium has yet to be done so it is not known if the measured 5f spectra and low temperature properties can be accounted for as well as can be done for cerium There has been some success in applying the impurity Anderson Hamiltonian treatment used for Ce 3d core level XPS spectra [5] to the 4f core level XPS spectra of Th [9,10] and U [11] materials

In this paper we report combined BIS/PES 5f and XPS 4f spectra for the magnetic heavy fermion material $U_2Zn_{17}$ [12] and for $Th_2Zn_{17}$ where the 5f level is nearly empty Resonant photoemission measurements were made at the Stanford Synchrotron Radiation Laboratory in the photon energy range 80–140 eV to identify the U 5f valence band features by tuning the photon energy through the U 5d threshold at 90 eV Polycrystalline samples were fractured and measured at 300 K in a vacuum of $8 \times 10^{-11}$ Torr The resolution, determined almost entirely by the cylindrical mirror analyzer used to measure electron kinetic energies, was 0 4 eV BIS and XPS spectra were obtained for the same samples at a photon energy of 1486 6 eV using a Vacuum Generators ESCALAB, operated under conditions to yield resolutions of 0 5 and 0 8 eV, respectively The samples were fractured, cooled to $\approx 100$ K and XPS spectra taken under a vacuum of $6 \times 10^{-11}$ Torr During BIS data-taking the chamber pressure was always below $1 \times 10^{-10}$ Torr

Fig 1 shows PES/BIS spectra for $U_2Zn_{17}$ The PES spectrum for photon energy $h\nu = 92$ eV is the Fano minimum where 5f emission is suppressed The spectrum labeled 5f is the result of subtracting the $h\nu = 92$ eV spectrum from the $h\nu = 108$ eV Fano maximum spectrum The intense feature 9 7 eV below $E_F$ is the spin–orbit split Zn 3d emission, which anti-resonates weakly at the U 5d edge, implying hybridization with the U 5f states This conclusion is substantiated by
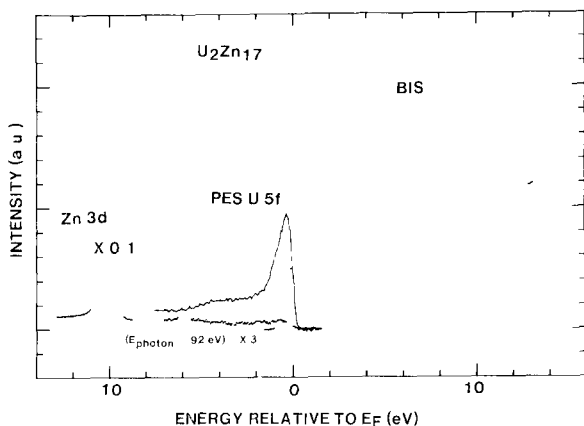
the similar photon energy dependences of the emission near $E_F$ and the Zn 3d emission from $h\nu = 115$ eV to $h\nu = 140$ eV We have no reliable information about the number $n_f$ of f-electrons and have arbitrarily made a rough scaling of the BIS and U 5f PES spectra to give $n_f \approx 2$ 5 as found in density functional calculations for other uranium intermetallics The BIS spectrum somewhat resembles that of $UPt_3$ [7], showing a shoulder at 0 5 eV, two peaks at about 1 5 and 2 1 eV, and a total width extending at least 6 eV above $E_F$ In $UAl_2$ and $UPt_3$ we have previously associated the separation of the shoulder and the lowest-energy peak with the 5f spin–orbit splitting, and the large width with the 5f Coulomb interaction [7] Sarma et al [13] have recently explored the latter possibility in the framework of a Hubbard Hamiltonian for the f-electrons, arguing that if the hybridization is very large, a hybridized band structure is a better starting point than the impurity Anderson Hamiltonian The particular model of Sarma et al neglects the fact that the Coulomb interaction occurs only for the 5f parts of the hybridized band structure, and for the parameters used, it probably cannot achieve sufficient mass enhancement to explain heavy-fermion materials But it does have the merit of placing much 5f weight around $E_F$ and is an interesting effort to address the differences between U and Ce set forth above

Fig 2 shows PES/BIS data for $Th_2Zn_{17}$ As for U, the emission near $E_F$ exhibits Fano resonance behavior at the 5d threshold, and we assume this
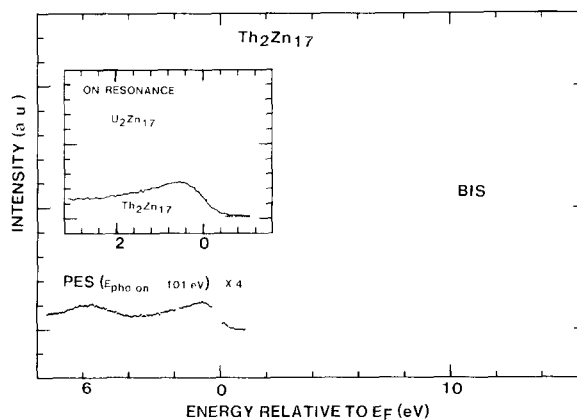


Fig 1 The PES/BIS spectra of $U_2Zn_{17}$ (see text)



Fig 2 The PES/BIS spectra of $Th_2Zn_{17}$ (see text)

indicates resonance of the Th 6d states, but cannot exclude the possibility that it arises from hybridized Th 5f states The spectrum shown is for the Fano maximum at $h\nu = 101$ eV, and the large Zn 3d peaks are not shown The data were taken from the cleanest fracture but still oxygen contamination is detected at 6 eV below $E_F$ The Zn 3d emission has a weak photon energy dependence in this range and hence can be used to place the PES spectra of the two materials on a common intensity scale for values of $h\nu$ not too different In the figure inset we show the emission near $E_F$ at the Th and U Fano maxima, $h\nu = 101$ eV and $h\nu = 108$ eV, respectively, scaled so that the Zn 3d's in each spectrum have the same intensity We believe this is the first direct comparison of the strength of the 6d and 5f resonances in actinides Thorium 5f states cause the BIS structure between 5 and 7 eV above $E_F$ while the 6d and 7s states give the flat and featureless background, showing that the large peak near $E_F$ in the U$_2$Zn$_{17}$ BIS spectrum is indeed due to 5f states

We have also measured the U 4f and Zn 2p core level XPS spectra of U$_2$Zn$_{17}$ The Zn 2p peaks are narrow and rather symmetric while the U 4f spectra reported in ref [17], are asymmetric and show weak and strong satellites at about 3 and 7 eV higher binding energy, respectively Satellites at 7 eV have been observed in several uranium materials, e g , UGa$_2$ [14], UBe$_{13}$ [15], UO$_2$ [16], URu$_2$Si$_2$ [17] and UAl$_2$ [12,17], and 3 eV satellites also occur in other uranium compounds [11] We also observe a satellite in the Th 4f spectra of Th$_2$Zn$_{17}$ at about 7 eV higher binding energy This is surprising and possibly interesting, as 4f satellites in intermetallic Th compounds typically occur at $\approx 3$ to 4 eV higher binding energy [9,10], but we cannot be sure that this is not due to some oxide in our sample, since ThO$_2$ has a 7 eV satellite [18]

To summarize, there remains the hope that the impurity Anderson Hamiltonian treatment will provide a means of deducing Hamiltonian parameters from U 4f spectra, as was done for Ce

3d spectra, and that a realistic impurity theory of the 5f spectrum will account for at least the gross differences between the experimental spectra of U and Ce

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