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High-Resolution, Low-Temperature, Photoemission Studies of Heavy-Fermion Systems: UBe$_{13}$ and UPt$_3$

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High-resolution (0.13 eV) photoemission measurements performed at low temperatures (≈20 K) show that a sharp feature (measured width ≈0.15 eV at full width at half maximum) exists at the Fermi edge in the electronic structures of UBe$_{13}$ and UPt$_3$. In UBe$_{13}$ the feature shows some temperature dependence.

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The unusual properties of the recently discovered heavy-fermion superconductors$^{1,2}$ are presently the subject of considerable investigation,$^3$ primarily because of the prospect of $p$-wave superconductivity. The similarity to the $p$-wave superfluid $^3$He has been pointed out by a number of investigators.$^{4-6}$ While the theoretical models are not yet on firm ground it appears that a common feature of all models$^5-8$ is the existence of a very narrow band at $E_F$ which is primarily $f$-like in nature and only very slightly hybridized with the $s$-$p$ bands of the neighboring atoms. It is this narrow band at $E_F$ which yields the astonishingly large specific-heat $\gamma$ values$^{1,2,9-11}$ and effective masses (two or three orders of magnitude larger than values of typical metals), which are characteristic of heavy-fermion systems. One of the current models$^5$ states that the magnetism in this nearly localized state is suppressed by the Kondo effect so that the materials can be described as “Kondo lattices”$^{8,12}$ characterized by a Fermi liquid of extremely heavy electrons.

The coexistence of superconductivity and spin fluctuations has been demonstrated recently$^{10,11}$ in UPt$_3$. The spin-fluctuation temperature $T^*$ can be deduced experimentally as the degeneracy temperature in the specific-heat vs temperature curve. For both UBe$_{13}$ and UPt$_3$ it is found$^2,10$ that $T^* \approx 10$ K. Since in heavy-fermion compounds $T^* \approx T_F$, i.e., the bandwidth of the narrow feature,$^5$ this translates into a bandwidth of order 0.001 eV. The first photoemission study of heavy-fermion systems$^{13}$ was undertaken in the hope of observing this narrow band. None was observed. Indeed the resonant photoemission data on UBe$_{13}$ pointed to a 5-eV-wide $5f$ band, which is anomalously large for actinide systems. A subsequent study$^{14}$ pointed out that at least part of the problem in the initial work on UBe$_{13}$ was rapid oxygen diffusion from the bulk onto the surface. In addition to suppressing any narrow features this also led to an erroneous interpretation of the oxygen peak at −7 eV as being due to Be $2p$ bands. Parks et al.$^{14}$ pointed out that the large width of the $5f$ band is probably due to strong $p$-$f$ hybridization and not the lack of hybridization as initially assumed.$^{15}$ However, they too failed to see any narrow features at $E_F$ because of insufficient resolution.

The failure of photoemission measurements on heavy-fermion systems thus far to observe sharp peaks at $E_F$ has been disappointing. A narrow band surely must exist at $E_F$ to yield the extremely large $\gamma$ values. It is difficult to believe that a 5-eV-wide band as measured in UBe$_{13}$ condenses into a state only $10^{-3}$ eV wide. A reasonable assumption must be that in these systems we have a narrow, nearly pure, $5f$ subband at $E_F$, while the remaining $5f$ electrons strongly hybridize with the ligand $p$ or $d$ electrons below $E_F$. Recent results by Wuilloud et al.$^{15}$ point out that most of the high density at $E_F$ is on uranium sites only, consistent with this point of view.

In order for photoemission measurements to make a critical contribution to the heavy-fermion investigation, it would be necessary to obtain resolutions on the order of the bandwidth of the sharp
feature and work at $T < T^*$. Clearly, this is not possible with present day technology if the condensed bandwidth is indeed $10^{-13}$ eV. On the other hand the temperature dependence of the Kondo effect often goes as $\log T$ so that we would expect some remnant of this state at temperatures presently obtainable ($\approx 20$ K) in photoemission experiments. At the very least we can look for the narrow band at $E_F$ which possibly condenses into this state, assuming of course that such condensation actually occurs and the Kondo model applies to heavy-fermion systems.

In this paper we report measurements on $\text{UBe}_3$ and $\text{UPt}_3$ at temperatures down to $20$ K and with a resolution of $0.13$ eV. Low temperatures were used in the hope that some remnant of a temperature-dependent narrow resonance could be seen at $20$ K. The low temperature had the added benefit in $\text{UBe}_3$ in that it prevented oxygen diffusion from the bulk, thus enabling us to maintain a contamination-free surface for several hours. A sharp feature having a measured width $\approx 0.15$ eV was observed in both materials at $E_F$.

The measurements were done at the Tantalus storage ring of the Synchrotron Radiation Center in Stoughton, Wisconsin. Polycrystalline samples of $\text{UPt}_3$ and $\text{UBe}_3$ were mounted onto the cold head of a closed-cycle helium refrigerator. Temperatures were monitored via a AuFe-Chromel thermocouple embedded in the copper holder in close proximity to the sample. The typical operating vacuum was $\approx 5 \times 10^{-11}$ Torr. Samples were fractured at temperature in situ. It was found that any change in temperature (up or down) resulted in some surface contamination, as evidence by a deterioration of the spectrum quality. The analyzer was a double-pass cylindrical-mirror analyzer operated at $0.095$-eV resolution (6-eV pass energy). The resolution of the monochromator ranged from $0.05$ eV at $40$-eV photon energy to $0.3$ eV at $100$-eV photon energy. The sharp feature thus could not be properly studied at resonance, but rather only at $40$ eV (i.e., at maximum resolution). Fortunately the $5f$ cross section at $40$ eV is sufficient to yield all necessary information.

In Fig. 1, we show data on both $\text{UBe}_3$ and $\text{UPt}_3$ obtained at $40$-eV photon energy. The long (10-eV) scans are at $0.3$-eV resolution (15-eV pass energy) and their primary aim is to demonstrate the oxygen-free surface as evidenced by a lack of structure in the 6–7-eV binding-energy region. Additional broad features are observed in the long-scan spectra of both compounds which were not reported in earlier investigations. As a result of space limitations we will discuss the broad features in a later publication. Resonance work, however, shows that in $\text{UBe}_3$ these broad features are $f$-like, but probably Pt derived in $\text{UPt}_3$.

We wish to focus attention on the short (3 eV wide) scans in Fig. 1, taken at $\approx 0.13$-eV resolution. In both $\text{UBe}_3$ and $\text{UPt}_3$ a sharp peak is observed precisely at the Fermi energy, with a measured half-width of $0.15$ eV. At $0.3$-eV resolution this peak is drastically diminished so that failure to observe it previously is understandable. Moreover, even a slight oxygen contamination substantially distorts all valence-band features. Thus it is nearly impossible to observe this sharp peak at room temperature.

This sharp feature at $E_F$ almost certainly must be associated with the band resulting in the heavy-mass electrons. Since the measured peak is only slightly broader than our resolution we can in principle determine its true width. We have done a modeling study using a Lorentzian peak centered at $E_F$, multiplied by a temperature-gradient Fermi
function and broadened by a Gaussian instrument function. A best comparison to the data was obtained with a full width at half maximum of 0.15 eV for the Lorentzian. We associate the filled part of this Lorentzian (0.075 eV) with the true width of the narrow feature, which of course still makes it nearly 2 orders of magnitude broader than the $10^{-3}$-eV-wide band inferred from specific-heat data. One possible explanation for the large width is that at 20 K the "Kondo lattice" no longer exists and we are observing only the "broad" band which condenses into the Kondo state and which is stabilized by slight hybridization with the Be-derived $s$-$p$ bands. Another possibility, however, is that of hole-lifetime broadening of a much narrower peak. Such broadening is of course expected to be small at $E_F$, but certainly is not negligible. Such a feature would show a temperature dependence for the amplitude, but not the width of the photoemission peak. Indeed the amplitude of the sharp feature in UBe$_{13}$ in Fig. 1 decreases by 10%-15% between 20 and 150 K. While this is less than anticipated from a Kondo-type system, modeling studies show that this decrease is considerably larger than one would expect from thermal broadening of a 0.075-eV-wide Lorentzian-shaped peak ($\approx 3\%$). The width of the narrow feature, however, does not change up to 150 K and neither do the remaining features in UBe$_{13}$. This would seem to rule out of favor the lifetime-broadening picture with the assumption that no experimental artifact contributes to the amplitude of this sensitive feature. Possible strains induced during sample fracture may also broaden the spectra. Obviously these data need to be taken with better than 0.075-eV resolution and $T = 10$ K.

The amount of hybridization can be inferred from the constant-initial-state (CIS) scans of Fig. 2. Curve I is for UPt$_3$ while curves II and III are for UBe$_{13}$. For curves I and II in Fig. 2 the initial state, $E_i$ at which the CIS scan is obtained, is at the point of the maximum in the EDC near $E_F$. Curve III is taken at $E_i = -5.0$ eV where the Be $s$-$p$ bands are prominent. We note that at $E_F$ we obtain the standard two-peaked structure in the CIS spectra (features B and D in Fig. 2) characteristic of 5f-electron emission at resonance. In addition, there are features A and C for both UPt$_3$ and UBe$_{13}$, and feature E for UBe$_{13}$ only. Features A and C are most likely associated with uranium 6$d$ resonant emission on the basis of the observation that in the rare-earth elements it is found$^{16}$ that valence electrons of different orbital character have cross-section maxima at different energies. Moreover, Iwan, Koch, and Himpsel$^{17}$ reported the existence of a "satellite" in uranium metal which resonates at the antiresonance of the uranium 5f's (i.e., $\approx 94$ eV). The energy position of the satellite matches the position of the 6$d$ states in uranium.$^{18}$ A difference spectrum between an EDC at 94-eV photon energy (maximum in 6$d$ emission) and an EDC at 92-eV photon energy in UBe$_{13}$ shows that the 6$d$ intensity is peaked at $E_F$ and is already of zero intensity at $-1.5$ eV. Clearly, uranium 6$d$ character exists at $E_F$ in UBe$_{13}$, and to a lesser extent in UPt$_3$. Band-structure calculations for UBe$_{13}$ bear this out this observation.$^{19}$ EDC's were not measured at the $d$ resonance in UPt$_3$. A substantial amount of Pt-derived $d$ density exists at $E_F$, however, on the basis of the 92-eV scans.

Perhaps a more interesting peak in Fig. 2 is feature E at 112-eV photon energy in UBe$_{13}$, which is just barely discernible near $E_F$ but is quite prominent in the $-5$-eV scan. This is the beryllium 1$s$-$p$ resonance which enhances the Be-derived part of the spectrum. Clearly the amount of Be-derived density at $E_F$ is very small, of the order of 1% of the density at $-5$ eV. This is consistent with the results of Wuilloud $et$ $al.$$^{15}$ who find that most of the high density is confined to the U atom with only weak hybridization with the $s$-$p$ bands from Be. Thus, it appears likely from our data that the sharp peak at $E_F$ is primarily $f$-like, with the $f$ band stabilized by a very small amount ($\approx 1\%$) of $p$ admixing from Be atoms. Such weak hybridization has been found sufficient to stabilize the $f$ band$^{20}$ in CeSn$_3$ where the Ce atoms are also too far apart for direct $f$-$f$ overlap. The metallic behavior of UBe$_{13}$ at high
temperatures probably comes from the broader 6d band at $E_F$.

While the narrow band at $E_F$ is almost purely f-like, the remaining f electrons are apparently strongly hybridized with the $s$-$p$ bands of Be and most likely do not condense into the Fermi liquid. Indeed both f- and p-derived features have substantial amplitude at $-2$ eV, which indicates maximum hybridization at this energy.

In summary, we have observed a narrow feature at the Fermi energy in the photoemission spectra of UBe$_3$ and UPt$_3$ which we believe can be associated with the narrow f band responsible for the heavy-fermion superconductivity. This feature is only slightly hybridized with the Be s-$p$ bands in UBe$_3$ and it shows a temperature dependence larger than expected from thermal broadening. A uranium-derived 6d band likewise exists at $E_F$ in UBe$_3$ and in UPt$_3$.

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3For a review, see G. R. Stewart, Rev. Mod. Phys. (to be published).
7H. Razafimandiby, P. Fulde, and J. Keller, to be published.
18B. Reihl, private communication.
19D. D. Koelling, private communication.