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Fermi-energy 5f spectral weight growth in $Y_{1-x}U_xPd_3$

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Abstract

High-resolution small-spot resonant photoemission measurements of the $Y_{1-x}U_xPd_3$ alloy system have revealed the variation in the U 5f lineshape down to 1% uranium dilution. Qualitatively, the spectrum consists of a peak below E_F and a second peak impinging on E_F. As x decreases from 1 to 0, the first peak moves toward E_F due to a process known as Fermi-level tuning. In addition, the relative intensity of the E_F peak is observed to grow in accordance with the onset of Kondo-like transport properties. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Uranium; 5f; Resonant photoemission; Anderson impurity model

Photoemission and transport studies show that the $Y_{1-x}U_xPd_3$ alloy system exhibits Fermi-level tuning [1,2] and develops Kondo and non-Fermi liquid properties [3] as x decreases from 1. In this alloy system, when U^{4+} ions are replaced by Y^{3+} ions, the number of conduction electrons decreases and the chemical potential is lowered. The consequence is that the main weight of the f-spectrum (and core levels) moves toward the Fermi energy E_F. Consistent with the Anderson impurity model, this decrease of the 5f binding energy ε_f leads for $x < 0.3$ to Kondo effects which were found to show non-Fermi liquid departures from usual behaviors [3]. The Kondo temperature scaling with x is quantitatively consistent with this picture for x at least as small as 0.05 [4].

Resonant photoelectron spectroscopy (PES) measurements were performed on cleaved polycrystalline uranium-alloy samples at the Advanced Light Source Beamline 7.0. A large order-of-magnitude U 5f cross-section enhancement at the U 5d edge allows the U 5f spectral weight to be isolated from alloy contributions by subtraction of on- and off-resonance valence spectra. High flux delivered by the undulator beamline allowed rapid characterization of the alloy surface with high resolution (60 meV) before appreciable grain boundary diffusion of oxygen or carbon to the surface occurred. Equally important as resolution and surface cleanliness for enhancing the sharpness of the PES spectra obtained is the use of a 100 μm focused beam spot to characterize surface inhomogeneity and minimize line broadening effects. With higher resolution and stronger signal intensities than in previous synchrotron studies of this alloy system [1,2], new spectral features have been observed, and measurement of weak uranium intensities down to an unprecedented 1% dilution was achieved with good statistics.

Movement toward E_F of the central weight of the U 5f spectra shown in Fig. 1 is consistent with previous lower resolution PES measurements. In addition, finer structures in the U 5f spectral shape are revealed, most importantly, a previously unobserved E_F peak which increases in relative spectral weight as x decreases. Such E_F weight

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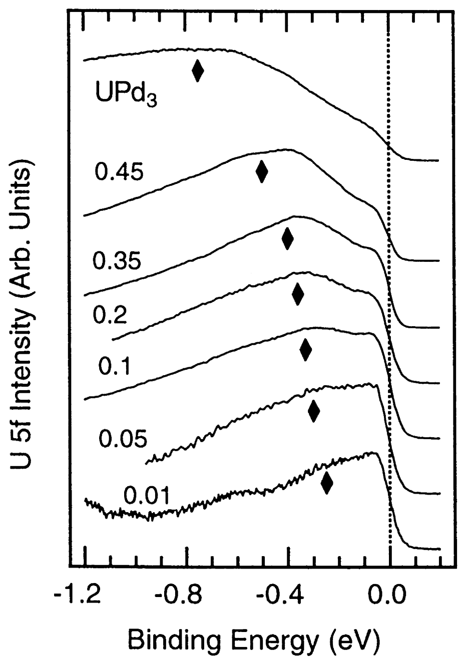

Fig. 1. Resonant photoemission U 5f spectral weight for the $Y_{1-x}U_xPd_3$ system obtained from the difference spectra of $h\nu = 108$ and 102 eV. Spectra were not measured for $0.5 < x < 0.9$, which is a mixed phase region.

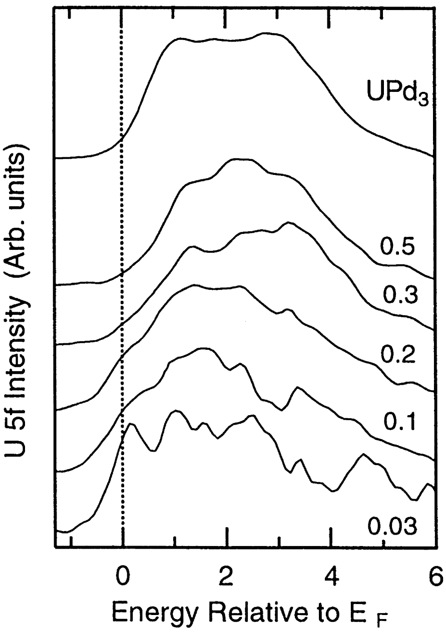

Fig. 2. X-ray inverse photoemission U 5f spectral weight for the $Y_{1-x}U_xPd_3$ system obtained by subtraction of a YPd_3 BIS spectrum as detailed in Ref. [5].

growth correlated with Kondo behavior is expected in the Anderson impurity model and has not been observed before in photoemission on uranium materials. This U 5f weight growth in PES is the counterpart of the weight growth just above E_F, observed previously in Bremsstrahlung isochromate spectroscopy (BIS) [5] and shown in Fig. 2. Note that the integrated weights of the BIS spectra need to be scaled by the ratio of the number of f-holes to f-electrons ($\approx 12:2$) for a direct comparison of E_F weight between the two spectroscopies. The PES spectra, with their much better spectral resolution and signal-to-noise, confirm the BIS results of Fig. 2 and complete the total picture. For this system, where uranium is progressively and severely diluted, the use of the impurity model framework to describe E_F weight variation is automatically credible. But the findings also are encouraging that the impurity model may be useful to capture some of the essential physics of concentrated systems as well.

The systematic variation of the U 5f spectral weight in $Y_{1-x}U_xPd_3$ has been measured with resonant photoemission. The growth of a narrow 5f peak at the Fermi level in $Y_{1-x}U_xPd_3$ correlated with movement of the main 5f spectral weight towards E_F and the onset of Kondo transport behavior is generic to the impurity Anderson model and gives promise to its application to the understanding of actinide electronic structure.

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