Temperature Dependent $5f$-states in URu_2Si_2

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Abstract

A dramatic temperature dependent enhancement of U $5f$ spectral weight at E_F is observed in angle-resolved photoemission measurements of URu_2Si_2 at the center of an X-point hole-pocket. Comparison of this temperature dependent behavior for excitation both at and below the U $5d \rightarrow 5f$ resonant threshold is presented.

Key words:
URu_2Si_2, $5f$ states, temperature dependence

A fundamental property of Kondo systems is a temperature scale below which local magnetic moments are gradually quenched as the system settles to a singlet ground state. An important aspect of the Anderson single impurity and lattice models for photoemission spectroscopy of f-electron systems is the prediction of an observable temperature dependence of the associated Kondo resonance near E_F as the system is cooled through the Kondo temperature T_K. For impurity models, either a sharpening or an increase of the k-integrated photoemission f-spectral weight is expected for Ce or Yb, respectively, with the difference arising from the former having one f-electron and the latter one f-hole [1]. Such temperature dependence has been reported for Ce [2] and Yb [3] compounds, but also disputed [4]. Predictions for lattice models exist [5], but thus far the models are not realistic as to the local orbital degeneracy or conduction electron number for Ce, Yb or U. Experimentally no significant temperature variation in photoemission has yet been reported for any U compound [4].

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URu_2Si_2, with an intermediate Kondo temperature of ≈70 K [6], is favorable for such a temperature-dependent study of the f-weight at different locations in **k**-space. Previous ARPES experiments on URu_2Si_2 [7,8] have (a) determined the crystal inner potential ($V_0 \approx 12$ eV), (b) mapped the basic d-band structure along high symmetry directions and (c) established the existence of hole pockets at the Γ, Z and X-points of the Brillouin zone, and (d) provided k-dependent $5f$ spectral signatures of the Anderson lattice using resonant ARPES. In this paper, a dramatic temperature-dependence of the U $5f$ spectral weight at the center of an X-point hole-pocket is presented and compared at on- and off-resonance photon energies. Previously no significant temperature variation of angle-integrated valence spectra from scraped single crystals of URu_2Si_2 [9] was observed.

URu_2Si_2 has the $ThCr_2Si_2$ crystal structure with a body-centered tetragonal Brillouin zone. Single crystal URu_2Si_2 samples were cleaved in ultra-high vacuum ($\leq 5\times10^{-11}$ torr) at 100 K exposing the [001] surface for ARPES measurements at ALS Beamline 10.0.1. Temperature measurements and regulation of a flowing He cryostat was performed with Si diode sensors attached close to the base of transferable sample stubs. Photon energies above and below the U $5d \rightarrow 5f$ absorption thresholds (108 eV) were used to compare spectra dominated by d-band spectral weight to spectra with U $5f$ weight resonantly enhanced. A total instrumental resolution of ≈40 meV and full angular acceptance of ≈0.36° was employed.

Fig. 1 shows such angular-resolved spectra measured at the X-point with both on- and off-resonance photon energies for temperatures in the experimental sequence of 100 K, 50 K and 25 K. Insets in Fig. 1 illustrate the existence of a distinct hole-pocket centered on the X-point (102 eV) and the photon energy dependence of the spectral weight. Namely, the on-resonance spectra ($h\nu$=108 eV) with enhanced $5f$ cross section, shows a clear confinement of the U $5f$ spectra weight to the interior of the d-band hole-pocket. This behavior represents a basic signature in the Anderson lattice model of f-d mixing near an idealized d-band crossing [5,8]. Important for this temperature dependent study is the relative absence of d-states below E_F at the center of this hole-pocket, thus providing a nearly ideal location at which f-states alone can be probed with minimal interference from other spectral weight.

In the spectra of Fig. 1, a striking additional enhancement of the E_F peak is observed for lower temperatures. Using 100K spectra taken just outside the X-point hole pocket at $k_{\|}$=1.5 Å^{-1}, as a model for the temperature-invariant background weight (short dashed spectrum in Fig. 1), the $5f$ spectral weight amplitude (area) enhancement at 108 eV, after subtraction of the background spectrum, is approximately 2.2 (1.5). For the off-resonance photon energy the $5f$ amplitude (area) enhancement of 2.6 (2.1) is even more dramatic.

A noticeable difference between the on- and off-resonance spectra is the existence of some additional temperature-invariant on-resonance spectral weight between 0.1 and 0.3 eV below E_F. This $5f$ spectral weight is approximately isolated by taking the on- minus off-resonance difference of the spectra outside the X-point pocket (see long dashed spectrum in Fig. 1(a)). Probing the spatial homogeneity of the sample surface reveals that this spectral weight is enhanced at "bad" regions of the sample surface where the observation of d-band dispersions is also weakened. These locations are approximately correlated to macroscopically rough topography regions with enhanced surface area and possibly faster oxidization.

It is significant that angle-integrated spectra from this cleaved surface do not reveal a discernable temperature dependence, due to the small ratio of temperature-dependent to temperature-invariant spectral weight. Hence, three factors of (*i*) reduced surface component, (*ii*) angle-resolution, and (*iii*) special **k**-space location are found to be necessary ingredients for this first ever observation of temperature dependence in photoemission of a U compound, providing an explanation for the previous null result for angle-integrated photoemission of scraped single crystals of URu_2Si_2 [9].

The above interpretation assumes that the observed temperature dependent spectral weight impinging on E_F arises from the bulk [10]. The temperature range of these variations is consistent with the bulk-sensitive observation by point-contact spectroscopy of a resonance at E_F appearing below 60 K [11]. Hence we are encouraged to interpret this large temperature variation of U $5f$ spectral weight in ARPES of URu_2Si_2 as possible evidence for Kondo singlet condensation. More detailed temperature dependent measurements are in progress.

Supported by the U.S. NSF at U. Mich. (DMR-9971611) and by the U.S. DoE at U. Mich. (DE-FG02-90ER45416) and at the Advanced Light Source (DE-AC03-76SF00098).

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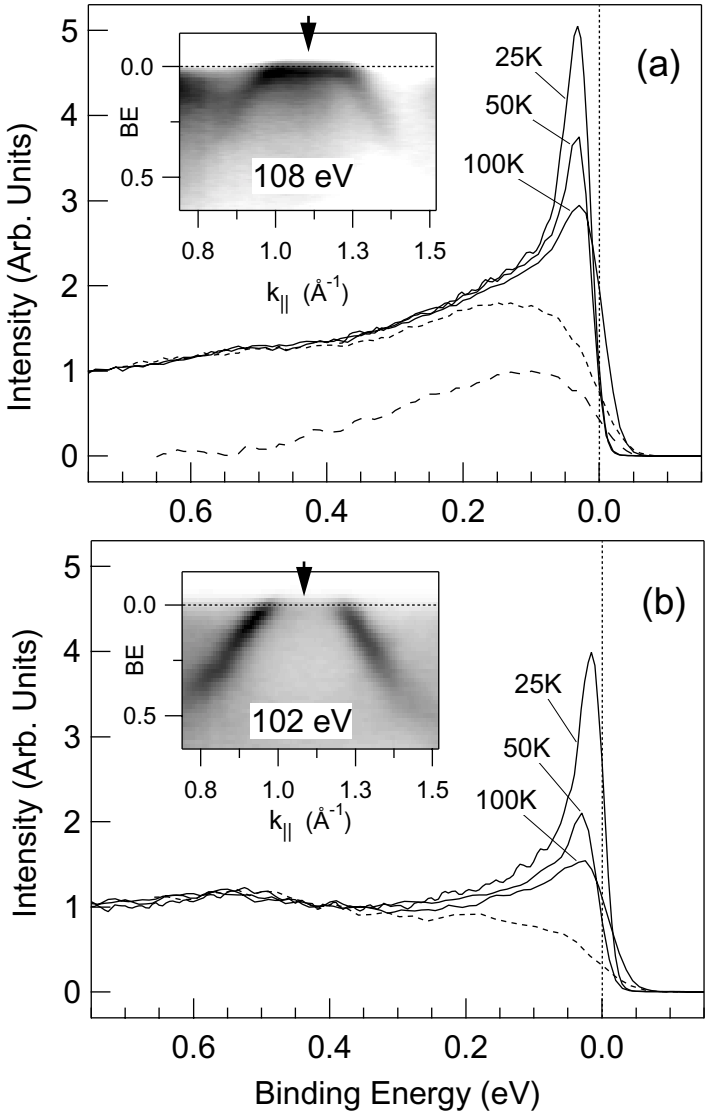

Fig. 1. Temperature dependence of angle-resolved valence spectra at (a) 108 eV and (b) 102 eV for URu_2Si_2 at the center of the X-point hole-pocket. Insets show the corresponding X-point valence band structure at 100 K. See text for explanation of dashed-line spectra.