

Temperature dependence of $5f$ states in URu_2Si_2

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INTRODUCTION

A fundamental property of so-called Kondo systems is the temperature scale below which spin excitations freeze out and the spin-singlet ground state is realized where local spin magnetic moments are fully screened by conduction electrons. An important aspect of the Anderson single impurity and lattice models for photoemission spectroscopy of f -electron systems is the prediction of a significant temperature dependence of the high density-of-states Kondo resonance near E_F as the system is cooled below the Kondo temperature T_K . Predictions for lattice models exist [1], but thus far the models are not realistic as to degeneracy or conduction electron number for Ce, Yb or U. For the impurity model below T_K a sharpening or increase of the \mathbf{k} -integrated photoemission f -spectral weight is expected for Ce or Yb, respectively, with differences arising from the former having one f -electron and the latter one f -hole [2]. Such temperature dependence has been reported for Ce [3] and Yb [4] compounds, but also disputed [5]. Experimentally no significant temperature variation in photoemission has yet been reported for any U compound [5].

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 \mathbf{k} -space. Previous ARPES experiments on URu_2Si_2 have (a) determined the crystal inner potential ($V_0 \sim 12$ 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 [7, 8], and (d) provided \mathbf{k} -dependent $5f$ spectral signatures of the Anderson lattice using resonant excitation. In this abstract, a Kondo-like temperature-dependence of the U $5f$ spectral weight at the center of an X -point hole-pocket is presented. Previously no significant temperature variation of angle-integrated valence spectra from scraped single crystals of URu_2Si_2 [9] was observed

EXPERIMENT

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 ($< 4 \times 10^{-11}$ torr) at 100K 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 $\approx 0.2^\circ$ was employed.

RESULTS

Figure 1 presents off- and on-resonance valence spectra at the “X”-point of URu_2Si_2 at a sample temperature of 100K. The off-resonance spectra ($h\nu = 102$ eV) show a distinct hole-pocket centered at $k_{\parallel} = 1.1 \text{ \AA}^{-1}$ while 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 of f - d mixing in the Anderson lattice model near an idealized d -band crossing [ref]. 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.

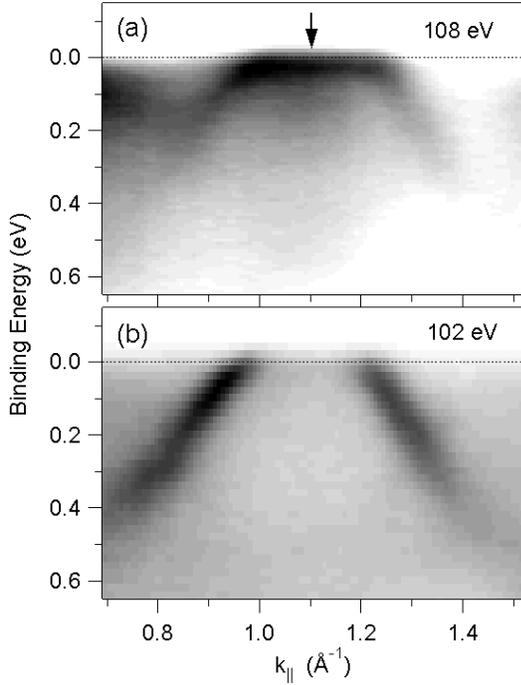


Figure 1. Off- and on-resonance valence band structure at the “X”-point of URu_2Si_2 at (a) 102 eV and (b) 108 eV at 100K.

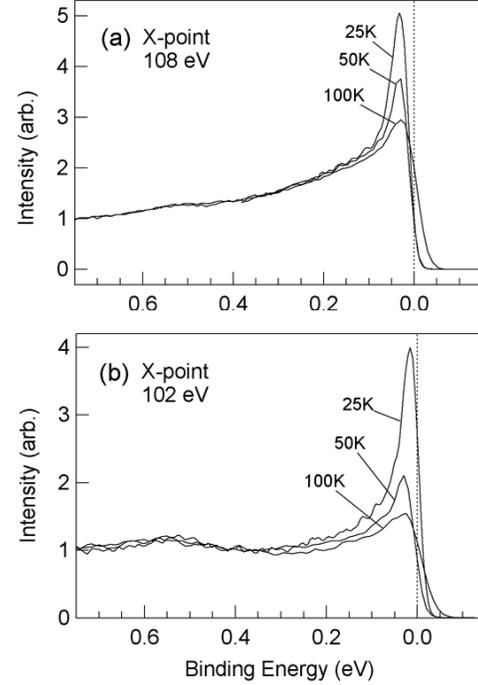


Figure 2. Temperature dependence of angle-resolved valence spectra at (a) 108 eV and (b) 102 eV for URu_2Si_2 at the center of the “X”-point hole-pocket.

Fig. 2 shows such angular-resolved spectra from the center of the X-point measured at both on- and off-resonance photon energies for temperatures in the experimental sequence of 100 K, 50 K and 25 K. A striking enhancement of the E_F peak is observed for lower temperatures. The amplitude increase from 100K to 25K is almost a factor of two in the raw data at 108 eV without consideration of a temperature-invariant inelastic background and greater than 400% with subtraction of this background. The relative enhancement in the off-resonance spectra in Fig. 2(b) at low temperature is even more dramatic (>800%).

A noticeable difference between the on- and off-resonance spectra is the additional existence of some temperature-invariant on-resonance spectral weight between 0.1 and 0.3 eV below E_F . 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 enhance surface area and possibly with slight oxidization (either inherently prior to cleavage or with rapid adsorption of residual gases). An experimental inability to fully suppress this weight results from relative spatial inhomogeneity of the cleaved surface and an excitation spot size ($\sim 0.1 \times 1$ mm) that is wide in one-dimension due to a 7° grazing-incidence geometry of the ARPES end-station.

ANALYSIS

Angle-integrated “transmission mode” spectra from this cleaved surface also does not reveal a discernible 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 viewed as necessary ingredients for this first ever observation of temperature dependence in photoemission of a U compound and provides an explanation for the previous null result for angle-integrated photoemission of scraped single crystals of URu₂Si₂ [9].

The above interpretation implies that the observed temperature dependent spectral weight impinging on E_F arises from the bulk. The temperature range of these variation is consistent with the bulk-sensitive observation by point-contact spectroscopy of a resonance at E_F appearing below 60 K [10]. Hence we are encouraged to interpret this large temperature variation of U $5f$ spectral weight in ARPES of URu₂Si₂ as possible evidence for the Kondo singlet condensation. A more complete analysis of this Kondo-like temperature dependence is in progress.

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