High resolution PES investigations on the prototype heavy fermion compound $CeCu_6$

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We present high resolution photoemission investigations on the heavy fermion compound $CeCu_6$. By application of a normalization procedure we can resolve the Kondo resonance and its crystal field structures in both the He II_α and He I_α data. A comparison with NCA calculations allows a quantitative determination of the Kondo temperature and the crystal field energies.

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1. Introduction

Since more than two decades heavy fermion (HF) compounds are under intensive investigation by photoemission spectroscopy (PES). This method allows to study directly core-level spectra, the valence states and the occupied part of the $4f$ spectral function close to the Fermi level (E_F). In contrast, inverse PES has access to the spectral function above E_F, but – due to its finite energy resolution – is not able to resolve in detail the $4f$ features, *i.e.* the Kondo resonance (KR) and its crystal field (CF) and spin-orbit (SO) partners close to the Fermi level. Recently it has been shown [1] that high resolution PES can resolve the KR in γ-like $CeCu_2Si_2$. This direct observation of the KR was possible by application of a well known normalization procedure based on the division by the Fermi-Dirac distribution (FDD) [3], that allows to recover the thermally occupied spectrum up to $\sim 5k_BT$ above E_F. At higher temperatures this energy range even includes the CF structures above E_F. To determine T_K and the CF splittings of the $J = 5/2$ multiplet (Δ_{CF}) one can directly compare the PES $4f$ spectrum with the spectral function extracted from calculations in the *non crossing approximation* (NCA) based on the *Single Impurity Anderson Model* (SIAM) [2]. By an iterativ fitting of the data one is able to determine all relevant model parameters, from which T_K and Δ_{CF} can be extracted and systematically compared to values *e.g.* determined by inelastic neutron scattering (INS). Surprisingly a reasonable coincidence can be found between PES and INS results, although PES investigates the surface region of the sample, whereas INS reflects bulk properties.

Here we turn our attention to the prototype heavy fermion compound $CeCu_6$, which is characterized by a huge value of the Sommerfeld coefficient ($\gamma_0 = 1.6$ JK^{-2}mole^{-1}) and a very low T_K of about 5 K. There exist only few PES studies in the literature on this compound [4, 5, 6] because of its low T_K value and the corresponding weak $4f$ spectral features in the PES data. We present temperature dependent high resolution ($\Delta E = 5.4$ meV) PES spectra, which clearly show the $4f$ spectral features by irradiation with He II$_\alpha$ and He I$_\alpha$. Against previous claims [7], the $4f$ PES cross-sections at He I$_\alpha$ are still strong enough to observe the KR and its SO structure.

2. Experimental setup

The PES experiments presented here have been performed using a SCIENTA SES 200 analyzer in combination with a monochromatized GAMMADATA VUV lamp at photon energies of $h\nu = 21.2$ eV (He I$_\alpha$) and $h\nu = 40.8$ eV (He II$_\alpha$) [8]. The energy resolution of the system was set to 5.4 meV to have sufficient photoemission intensity during the He II$_\alpha$ measurements. The single-crystalline $CeCu_6$ samples were grown by Czochralski technique in a high-purity argon atmosphere using a tungsten crucible.

3. Results

Typical high resolution near-E_F spectra on $CeCu_6$ are displayed in Fig. 1, measured at different temperatures with He I$_\alpha$ (right panel) and He II$_\alpha$ (left panel) radiation. It should be mentioned that although the photoionization cross-sections of the Ce $4f$ states increase about a factor 3.2 from irradiation with He I$_\alpha$ and He II$_\alpha$ [9] the PES spectra over an extended energy range (not shown here) clearly show the $4f$ derived spectral features, namely the SO structure at around $E_B = 250$ meV and the tail of the KR directly at E_F. The latter can be observed with He II$_\alpha$ and even with He I$_\alpha$ radiation in the insets of Fig. 1, but with different intensities. The spectral intensity of the raw He I$_\alpha$ and He II$_\alpha$ spectra at energies of a few meV above E_F and higher is suppressed by the FDD (see insets of Fig. 1). If we now apply the normalization method to the raw data, we can reconstruct the spectral information up to an energy of $\sim 5k_BT$ (see Fig. 1). Obviously there appears a narrow peak with a full width at half maximum (FWHM) of ~ 6 meV and a maximum at about 3 meV above

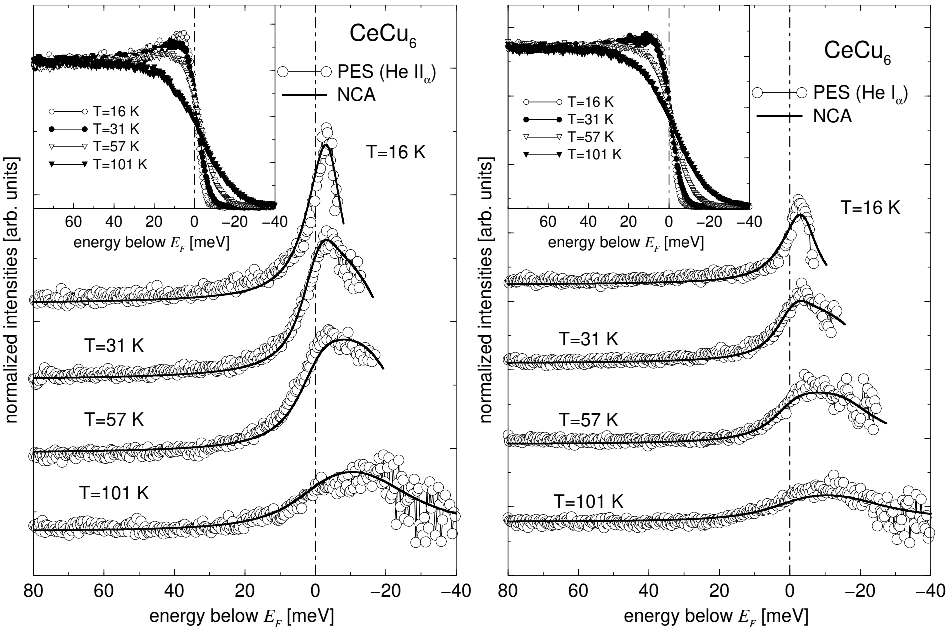

Fig. 1. Theoretical and experimental spectra of $CeCu_6$ after application of the mentioned normalization procedure. The PES data are taken with He II_α (left) and He I_α radiation (right) at different temperatures. Model parameters of the NCA: $\epsilon_f = -1.05$ eV, $D = 2.8$ eV, $\Delta_{CF} = 7.2/13.9$ meV, $\Delta_{SO} = 250$ meV, $V = 116$ meV. The insets show the data on the same energy scale prior to the normalization. All spectra are normalized to the same intensity at ≈ 100 meV for clarity.

E_F both in the low temperature He I_α and He II_α data. This is sufficiently below $5k_BT \approx 7$ meV, which defines the limit of the usable energy range.

In order to identify this narrow peak we have performed NCA calculations with a set of parameters that is given in the figure caption of Fig. 1. The striking coincidence between theory and experiment is obvious at the first glance. The NCA spectra are able to describe the lineshape and the energy position of this narrow peak just above E_F – the Kondo resonance – both in the spectra taken with He I_α and He II_α radiation. In addition, the temperature dependence of the spectra is described consistently. Towards higher T, the linewidth of the Kondo resonance increases, while the maximum intensity becomes smaller and the KR and the CF structures are successively smeared out. Above $T = 101$ K the thermal broadening of the Fermi-Dirac distribution is large enough to have access to the CF excitations at ~ 15 meV.

Compared to the He I_α irradiation, a considerable increase of the relative intensity of the KR can be observed in the spectra taken with He II_α radiation. If we compare both normalized low temperature spectra we find an intensity gain by a factor of 2. This is identical with the observed intensity loss in the SO feature by changing from He II_α to He I_α. The difference between this value and the theoretical one can be explained by the influence of the Cu $3d$, Cu $4s$ and Cu $4p$, as well to the Ce $5d$ states, that partly give a considerable contribution to the spectra in this energy range.

In order to estimate T_K from our data, we analysed the linewidth of the KR in the NCA spectra calculated at $T = 0.1 \times T_K \approx 1$ K with the parameter set extracted from the fit described above [1]. By this procedure we could determine $T_K = 4.6$ K for $CeCu_6$. We are now able to compare this Kondo temperature and the CF energies with the values determined by INS measurements. From INS we find $T_K = 5.0 \pm 0.5$ K [10] and $\Delta_{CF} = 7.0/13.8$ meV [11]. Surprisingly our results are in reasonable agreement with the INS bulk values, considering especially that in the surface sensitive PES the spectral function, and hence T_K, might be modified compared to the bulk values. The reason for this can be found in the fact that $CeCu_6$ belongs to the class of γ-like Ce compounds, where no real difference exists between surface and bulk electronic properties [12]. Thus we have shown that PES studies can contribute important information to the discussion of CF excitations in Ce compounds.

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