$CeRu_2Si_2$: temperature dependence of the Kondo resonance and its satellites

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Abstract

By use of high-resolution photoelectron spectroscopy we investigate the $4f$ spectral function of the heavy fermion compound $CeRu_2Si_2$ in the energy range close to the Fermi level. A normalization method proposed by Greber *et al.* [1] allows to resolve the Kondo resonance and (with some restrictions) its crystal field satellites. We discuss the applicability of this method with respect to a quantitative analysis of the data, and compare our experimental results to non-crossing approximation (NCA) calculations in the frame of the single impurity Andersson model (SIAM).

Key words: Cerium compounds, Kondo resonance, Photoemission, high resolution

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The most prominent spectral feature of intermetallic heavy fermion compounds is the so called "Kondo resonance"(KR). In cerium systems it arises as a narrow peak structure just above the Fermi level E_F. Up to now it is still a challenging task to investigate this KR experimentally. There exist three established, spectroscopic methods to investigate the spectral function close to the Fermi energy, namely photoemission spectroscopy (PES), inverse photoemission spectroscopy (IPES) and scanning tunneling spectroscopy (STS). The difficulty in observing the KR lies by IPES in its limited energy resolution ($\sim$ 100 meV), whereas PES with a much better energy resolution has merely access to the tail of the KR below E_F and STS probes the local $4f$ spectrum only indirectly by tunneling into the conduction states. In a recent publication [2] it has been shown that a careful analysis based on the procedure suggested

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by Greber *et al.* [1] allows to observe the KR and its crystal field (CF) satellites below and *above* E_{F} in $\mathrm{CeCu_2Si_2}$.
Here we present temperature dependent PES results on the isostructural heavy fermion compound $\mathrm{CeRu_2Si_2}$, where in the data analysis we applied the same normalization procedure. We also show in which temperature range this analysis yields reasonable qualitative and quantiatative results. In order to compare the experimental results with theoretical predictions we have calculated the $4f$ spectral function using the non-crossing approximation (NCA) [3] based on the single impurity Anderson model (SIAM) including all CF and spin-orbit (SO) excitations (see also Ref. [2]). The SIAM is the most simple approach for the description of thermodynamic and spectral properties of strongly correlated Ce compounds.

Greber *et al.* [1] have demonstrated that a careful analysis of PES data based on a normalization with the Fermi-Dirac-distribution (FDD) at the respective temperature allows to investigate the spectral function up to energies of approximately $5k_{\mathrm{B}}T$ *above* E_{F}. Two conditions for this method are required: (i) A satellite free and highly intense photon source must provide low noise data, and (ii) the experimental energy resolution ΔE must be at least comparable to the temperature broadening. We fulfilled both conditions by performing the PES experiments with a net energy resolution of $\Delta E = 5.4$ meV and the use of a monochromized microwave driven discharge lamp. Condition (ii) can be traced back to the following problem:

$$\frac{[S(E,T)\cdot f(E,T)]\otimes g(E,\Delta E)}{f(E,T)\otimes g(E,\Delta E)} \neq S(E,T)\otimes g(E,\Delta E), \tag{1}$$

where $S(E,T)$ and $f(E,T)$ describe the spectral function and the FDD at given energy E and temperature T, respectively, and $g(E,\Delta E)$ is a Gaussian with full width at half maximum (FWHM) of ΔE. The left hand side of Eqn.1 exactly express the normalization suggested by Greber *et al.* [1]. Because $[S\cdot f]\otimes g$ is not associative Eqn.1 stated that this procedure in general is not an exact one and only can be held for T of the order of ΔE. Therefore, in our case normalized spectra at $T < 100$ K can be artificially modified. As also stated in Ref. [1] the denominator of the left side of Eqn.1 can well be accounted for by introducing an effective temperature of $T_{eff} = \sqrt{T^2 + (\Delta E/4k_{\mathrm{B}})^2}$ in the FDD ($\tilde{f}(E,T_{eff})$). The effect of the normalization method described by the left hand side of Eqn.1 can clearly be shown by its application to spectral functions resulting from NCA calculations (S^{NCA}) at different T. Thus we first execute the prescription $[S^{NCA}\cdot f]\otimes g$ of the numerator at the left side in Eqn.1. Then we perform the normalization by using $f\otimes g$ on the one hand and $\tilde{f}$ on the other hand. For a more realistic comparison with these results we also carry out $S^{NCA}\otimes g$ (right side of Eqn.1) to prove in which temperature range Eqn.1 is still valid for a fixed ΔE.

At low temperature a clear difference between the results of the normalization procedure and the Gaussian broadened spectrum can be observed. The error in the position and width of the KR is of the order of $\simeq \Delta E$. This shows that the normalization to an FDD with T_{eff} ($\tilde{f}$) can be used without a loss of information in comparison to a Gaussian broadened FDD ($f \otimes g$). Increasing the temperature now leads to a more and more vanishing difference in width and position between the results of two normalizations and $S^{NCA} \otimes g$. Above 100 K no difference will be observable.
Although the normalization to an effective temperature broadened FDD ($\tilde{f}$) cannot exactly reproduce $S \otimes g$, we can use this method for the quantitative comparison between experiment and theory. Herefore it is necessary to apply the normalization to $\tilde{f}$ to both experimental and theoretical spectral functions. The difference of both can be used as a χ^2 measure for the quality of the fit.
In Fig. 1 we apply the latter method for the analysis of the temperature dependence of $CeRu_2Si_2$ and $CeCu_2Si_2$. In contrast to $CeCu_2Si_2$ an additional peak can be observed in the $CeRu_2Si_2$ spectra below 300 meV binding energy (see insets). This feature is due to the Ru $4d$ states which overlap with the spin-orbit (J=7/2) satellite at $\Delta_{SO} = 295$ meV. The separation of these two peaks clearly can be seen in the difference spectrum. The model parameters for the NCA calculations (see caption of Fig. 1) have been taken from independent experimental measurements where possible [4] and where slighly adjusted to reproduce the measured PES data and therefore represent surface rather than bulk properties.
After application of the normalization procedure it is obvious from Fig.1 that the experimentally observed spectra are accurately described both quantitatively and qualitatively by the NCA simulations. At low T both the PES and NCA data show the KR with FWHM of ~ 6 meV and maxima at about 5 meV above E_{F} for $CeRu_2Si_2$ as well as $CeCu_2Si_2$. By determining the intrinsic line width, i.e. the line width of the KR in the original low T NCA spectral functions, it is now possible to get access to the Kondo temperature, which for both systems coincidence with the values extracted from bulk measurements. It should be mentioned that no CF-splitting could be observed at energies below E_{F} for $CeRu_2Si_2$ in contrats to the case of $CeCu_2Si_2$.
At higher temperatures a comparison now allows to identify the origin of the other spectral features above E_{F} (well marked in the spectra of $CeCu_2Si_2$): With increasing T, the line width of the KR increases, while the maximum intensity becomes smaller and the distinction of KR and CF satellites is successively smeared out. At $T \geq 100$ K the thermal broadening of the FDD is large enough for PES to have access to the CF excitations above ~ 20 meV. At these temperatures the KR has disappeared as a separate peak, and the enhanced $4f$ DOS near E_{F} is rather due to the CF excitations which are broadened and positioned somewhat above E_{F}. Thus, the persistence of an enhanced DOS at the Fermi level even at room temperature, despite a substantially lower T_K, is naturally explained within the SIAM in combination

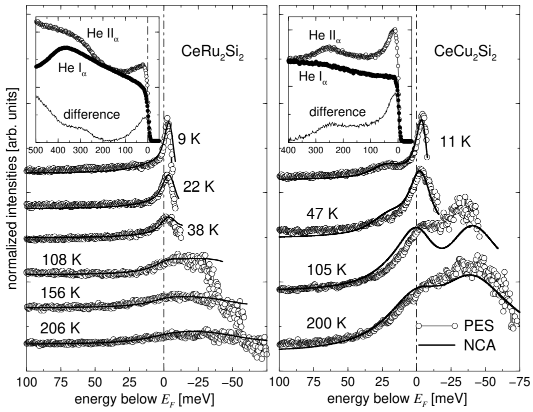

Fig. 1. Theoretical (NCA) and experimental (PES) T dependence of the $4f$ spectral function of $CeRu_2Si_2$ (left) and $CeCu_2Si_2$ (right) after application of the described normalization procedure (see text). Used model parameters for the NCA calculation for $CeRu_2Si_2/CeCu_2Si_2$: $\epsilon_f = -2.4$ eV, $D = 4.2/4.0$ eV, CF splittings of the $J = 5/2$ sextet $\Delta_{CF} = 19/30$ meV and $34/36$ meV, SO splitting $\Delta_{SO} = 295/270$ meV, hybridization $V = 170/200$ meV. The photoemission spectra for different T are measured with He II$_\alpha$ radiation on poly-crystalline samples. The insets show an extended energy range (measured with $\Delta E \sim 13$ meV, $T \sim 20$ K) including the spin-orbit (J=7/2) excitation at $\approx 295/270$ meV below E_{F} and the non-4f background as determined using He I$_\alpha$ radiation.

with CF excited states.

In conclusion we have demonstrated that the application of the described normalization procedure on high resolution PES data provides access to spectral features above E_{F} in heavy fermion cerium compounds. A comparison with NCA calculations allows to identify the experimentally observed structures as the KR and its CF satellites. In addition we have demonstrated that even a quantitative analysis is possible if the method is applied to both experimental and theoretical spectra.

This work was supported by Sonderforschungsbereich SFB 277 and Bundesministerium für Bildung und Forschung.

References

[1] T. Greber, T. J. Kreutz, and J. Osterwalder, Phys. Rev. Lett. **79**(22), 4465 (1997).

[2] F. Reinert et al., submitted to Phys. Rev. Lett. (cond-mat/0104037).

[3] N. E. Bickers, D. L. Cox, and J. W. Wilkins, Phys. Rev. B. **36**(4), 2036 (1987).

[4] G. Zwicknagl, Adv. Phys.**41**, 203–302 (1992).