Kondo satellites in photoemission spectra of heavy fermion compounds

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Outline

- Introduction: Kondo resonances in the photoemission spectra of Ce systems
- Single ion Anderson model of 4f multiplets:
  slave boson mean field theory
- Spectral function from perturbative RG method
- Spectral function in non-crossing approximation
- Comparison with experiment and conclusion

Collaborators:

Theory

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Experiment

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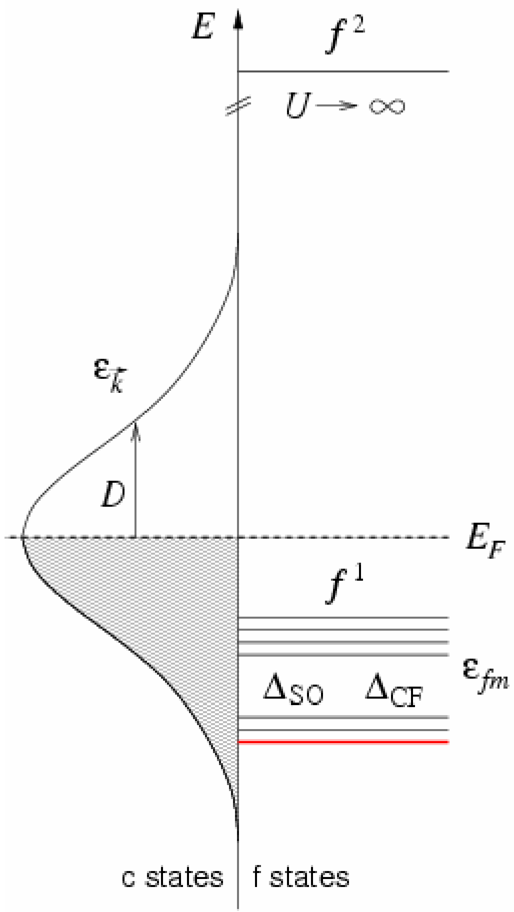
Modelling Ce compounds: Periodic Anderson model

Localized 4f electrons hybridizing with conduction electron band

Single ion physics expected to dominate local properties observed in photoemission spectra

Lattice effects leading to coherent transport less important for PES



Multiorbital single impurity Anderson model

4f-states |m, σ > : 7 spin-degenerate levels split by

spin-orbit interaction	→ octet	→	four doublets
		crystal field	
	→ sextet	→	three doublets

$$H = H_0 + \sum_{m\sigma} \varepsilon_{fm} f^\dagger_{m\sigma} f_{m\sigma} + \sum_{\vec{k}m\sigma} \left(V_{\vec{k}m} c^\dagger_{\vec{k}\sigma} f_{m\sigma} + \text{h.c.} \right)$$

$$+ \frac{U}{2} \sum_{(m\sigma)\neq(m'\sigma')} f^\dagger_{m\sigma} f_{m\sigma} f^\dagger_{m'\sigma'} f_{m'\sigma'} \ , \qquad (1)$$

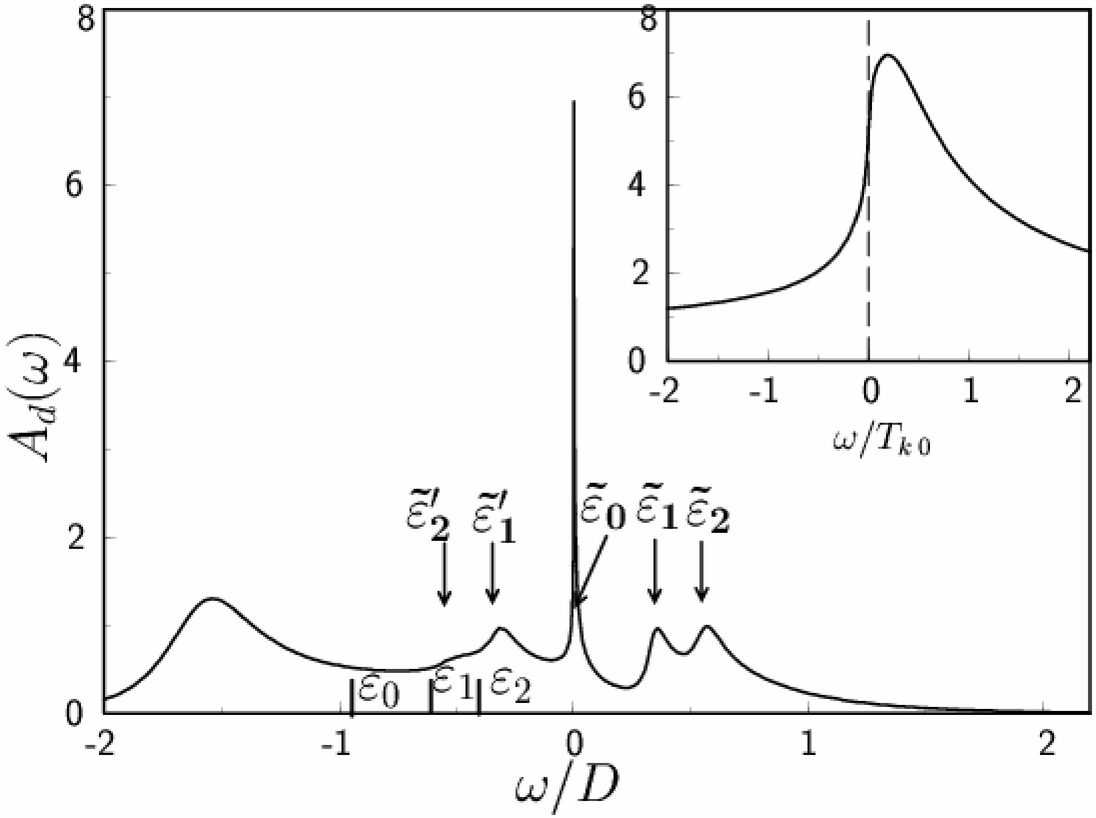
$H_0 = \sum_{\vec{k}\sigma} \varepsilon_{\vec{k}} c^\dagger_{\vec{k}\sigma} c_{\vec{k}\sigma}$ conduction band

Kondo resonances in the single particle spectra

Model system: triplet of localized states, occupation n=1

Kondo resonances

- main resonance at Fermi level
- above Fermi level : rigid shift of multiplet
- below Fermi level: mirror image of excited levels
- width of excited levels of order a few T_K



Methods: Pseudoparticle representation

Limit of infinite Coulomb interaction U: multiple occupancy of f-states excluded

Pseudoparticle operators: b^+ (empty level), f_m^+ (singly occupied level)

f-electron operator: $f_{el,m}^+ = f_m^+ b$

Constraint: $$Q = b^+ b + \sum_m f_m^+ f_m = 1$$

Implementation of constraint $H \to H + \lambda Q \;, \quad \lambda \to \infty$

Methods: Slave boson mean field theory

Replace Bose operators by mean field plus fluctuations:

$$\hat{b} = r + \hat{a}, \quad r = \langle b \rangle, \qquad \hat{\lambda} = \lambda_0 + \hat{\tilde{\lambda}}$$

Mean field Hamiltonian

$$H = H_{\text{kin}} + \sum_{m\sigma} \tilde{\varepsilon}_{dm} f^\dagger_{m\sigma} f_{m\sigma} + \sum_{pm\sigma} [\tilde{V}_{mp} f^\dagger_{m\sigma} c_{p\sigma} + h.c.]$$
$$+ (\tilde{\varepsilon}_{d0} - \varepsilon_{d0})[r^2 + a^\dagger a - 1] + H_{\text{int}} \qquad (3)$$

Mean field equations

$$r^2 + \langle a^\dagger a \rangle + \sum_{m\sigma} \langle f^\dagger_{m\sigma} f_{m\sigma} \rangle - 1 = 0$$
$$(\tilde{\varepsilon}_{d0} - \varepsilon_{d0}) r + \sum_{pm\sigma} V_{mp} \langle f^\dagger_{m\sigma} c_{p\sigma} \rangle = 0 \; .$$

Level shifts:

$$\tilde{\varepsilon}_0 = \varepsilon_0 + \lambda_0 = \alpha T_K, \qquad \alpha \ll 1$$
$$\tilde{\varepsilon}_m = \tilde{\varepsilon}_0 + (\varepsilon_m - \varepsilon_0)$$

Kondo temperature:

$$T_K = \left\{ \prod_{m=1}^{M-1} \left(\frac{D}{\varepsilon_m - \varepsilon_0} \right)^{\frac{\Gamma_{mm}}{\Gamma_{00}}} \right\} D e^{-\frac{\pi|\varepsilon_0|}{2\Gamma_{00}}}$$

Electron spectral function: sb MFT + fluctuations

$$A_{d\sigma}(\omega) \simeq \sum_m \frac{T_{K\,m}}{\Gamma_{mm}} \mathrm{Im} G_{f\,mm\sigma}(\omega - i0)$$

$$-\left[\Theta(\tilde{\varepsilon}_{d0} - \omega) - \frac{1}{2}\right] \mathrm{Im}\Big[-\omega + \tilde{\varepsilon}_{d0} + \varepsilon_{d0}$$

$$-\sum_m \frac{\Gamma_{mm}}{\pi} \ln \frac{(\omega + \varepsilon_{dm} - \varepsilon_{d0})^2 + T_{K\,m}^2}{\tilde{\varepsilon}_{dm}^2 + T_{K\,m}^2}$$

$$-i\mathrm{Im}\Sigma_a(\omega - i0) - i\,\mathrm{sgn}\omega\, T_{K\,0}\Big]^{-1}.$$

where $G_{f,\sigma,m,m'}(\omega) = [(\omega - \tilde{\varepsilon}_{d,m})\delta_{m,m'} - i\tilde{\Gamma}_{m,m'}(\omega)]^{-1}$

Resonance peaks at $\tilde{\varepsilon}_{d,m} = \tilde{\varepsilon}_{d,0} + (\varepsilon_{d,m} - \varepsilon_{d,0})$ and at $\tilde{\varepsilon}'_{d,m} = -(\varepsilon_{d,m} - \varepsilon_{d,0})$

Resonance widths: $\tilde{\Gamma}_{m,m} = r^2 \Gamma_{m,m} = O(T_K)$

bare level spacings

weak dependence on $\Delta\varepsilon$

Methods: Mapping on to Kondo model

Elimination of empty and multiply occupied states yields Kondo model :

$$H_K = H_0 + \sum_{\sigma,\sigma',m,m'} J_{m,m'} f^{+}_{m',\sigma'} f_{m,\sigma} \sum_{k,k'} c^{+}_{k,\sigma} c_{k',\sigma'} + \sum_{m} (\varepsilon_m - \varepsilon_0) f^{+}_{m,\sigma} f_{m,\sigma}$$

where $J_{m,m'} = 2V_m V^*_{m'} / (\varepsilon_m + \varepsilon_{m'})$ Constraint: $\sum_{m,\sigma} f^{+}_{m,\sigma} f_{m,\sigma} = 1$

Perturbative second order correction to exchange constants ($\Delta\varepsilon_m = \varepsilon_m - \varepsilon_0$):

$$J^{(2)}_{mm'}(\omega) = -N(0) \sum_{l} J_{ml} J_{m'l} \{ \int_{-D}^{D} d\varepsilon \frac{f(\varepsilon)}{\varepsilon + \omega + \Delta\varepsilon_m - \Delta\varepsilon_l} + (\omega \to -\omega) \}$$

$$\approx -N(0) \sum_{l} J_{ml} J_{m'l} \ln \{ ([\Delta\varepsilon_m - \Delta\varepsilon_l]^2 - \omega^2) / D^2 \}$$

→ Logarithmically divergent terms at $\omega = \pm[\Delta\varepsilon_m - \Delta\varepsilon_l]$

Methods: Renormalization group equations

- **Poor man‘s scaling** **(Anderson, 1970)**
  remove high energy states at ±D and absorb change into coupling constant g(D), depending on running cutoff D, take D $\to$ 0 .

- **Extend renormalization group meth. to energy dependent coupling functions**

$$\frac{dg_{\|m,m'}(\omega)}{d\ln D} = -2\sum_l g_{\perp m,l}\, g_{\perp l,m'}\,\Theta(D-\Delta\omega) \qquad \Delta\omega = |\,\omega - \Delta\varepsilon_l + \Delta\varepsilon_m\,|$$

$$\frac{dg_{\perp m,m'}(\omega)}{d\ln D} = -2\sum_l g_{\perp m,l}\, g_{\|l,m'}\,\Theta(D-\Delta\omega)$$

Θ-(step)-functions account for absence of renormalization if energy is outside the bandwidth D

Two levels, splitting Δε:

$$g_{\|01}(\omega) = \Theta(|\,\omega-\Delta\varepsilon\,| - \Delta\varepsilon)\frac{1}{2\ln[|\,\omega-\Delta\varepsilon\,|/T_K]}$$

$$+\,\Theta(\Delta\varepsilon - |\,\omega-\Delta\varepsilon\,|)\frac{1}{2\ln(\Delta\varepsilon/T_K)}\left|\frac{\Delta\varepsilon}{\omega-\Delta\varepsilon}\right|^{1/\ln(\Delta\varepsilon/T_K)}$$

Methods: Decoherence stops RG flow

Logarithmic divergencies involving excited states are cut off by the finite spin relaxation rate (even at T=0):

$$\gamma \approx \frac{\Delta\varepsilon}{[\ln(\Delta\varepsilon / T_K)]^2}$$

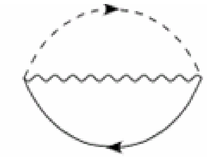
To account for this effect, we replace the conduction electron energy in the RG equations by $\sqrt{\omega^2 + \gamma^2}$

→ Width of Kondo satellite peaks $\approx \gamma$, increasing with Δε

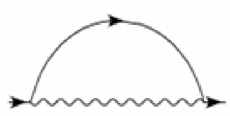
Systematic and controlled method, provided

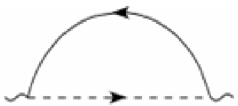
$$\gamma \gg T_K, \quad \text{or} \quad \Delta\varepsilon / T_K \gg \frac{1}{\ln^2(\Delta\varepsilon / T_K)}$$

Methods: Non-crossing approximation

Conserving approximation derived from generating functional $\Phi =$ 

Pseudoparticle self energies:

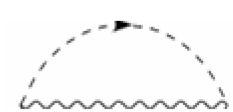
$\Sigma_{f,\sigma,m,m'}(\omega) =$  $= \Gamma_{m,m'} \int d\varepsilon\, f(\varepsilon) A_{c,\sigma}(\varepsilon) G_b(-\varepsilon + \omega)$

$\Sigma_b =$  $= \sum_{m,m',\sigma} \Gamma_{m,m'} \int d\varepsilon\, f(\varepsilon) A_{c,\sigma}(\varepsilon) G_{f,\sigma,m,m'}(\varepsilon + \omega)$

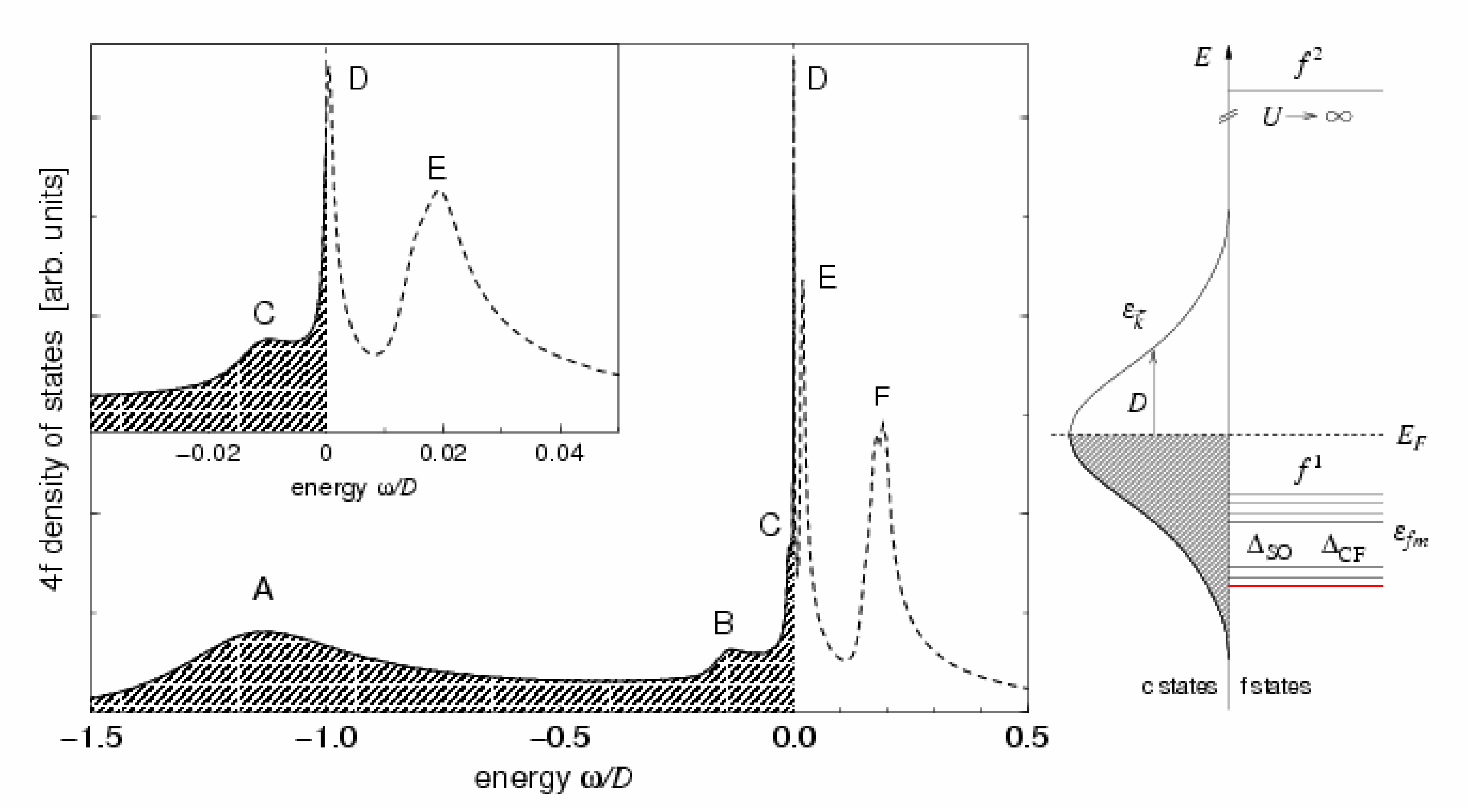
Conduction electron DOS $A_{c,\sigma}(\varepsilon)$ Bare level broadening $\Gamma_{m,m'}$

$$G_{f,\sigma,m,m'}(\omega) = [(\omega - \lambda_0 - \varepsilon_{f,m})\delta_{m,m'} - \Sigma_{f,\sigma,m,m'}(\omega)]^{-1}$$

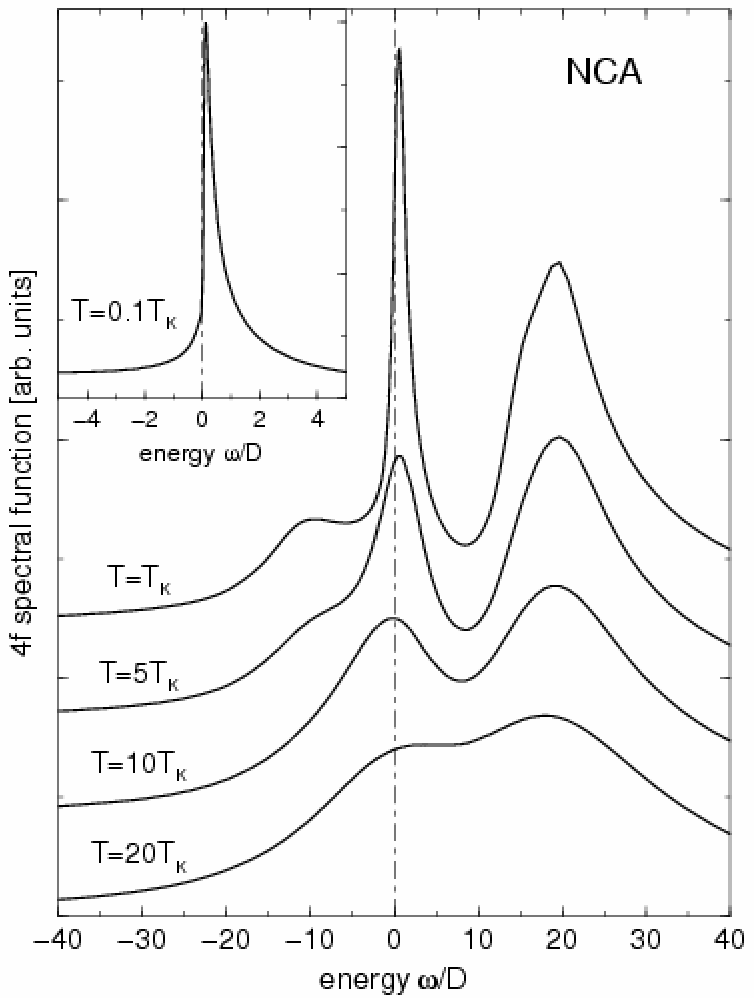
$$G_b(\omega) = [\omega - \lambda_0 - \Sigma_b(\omega)]^{-1}$$

$G^{el}_{f,\sigma,m,m'}(\omega) =$ 

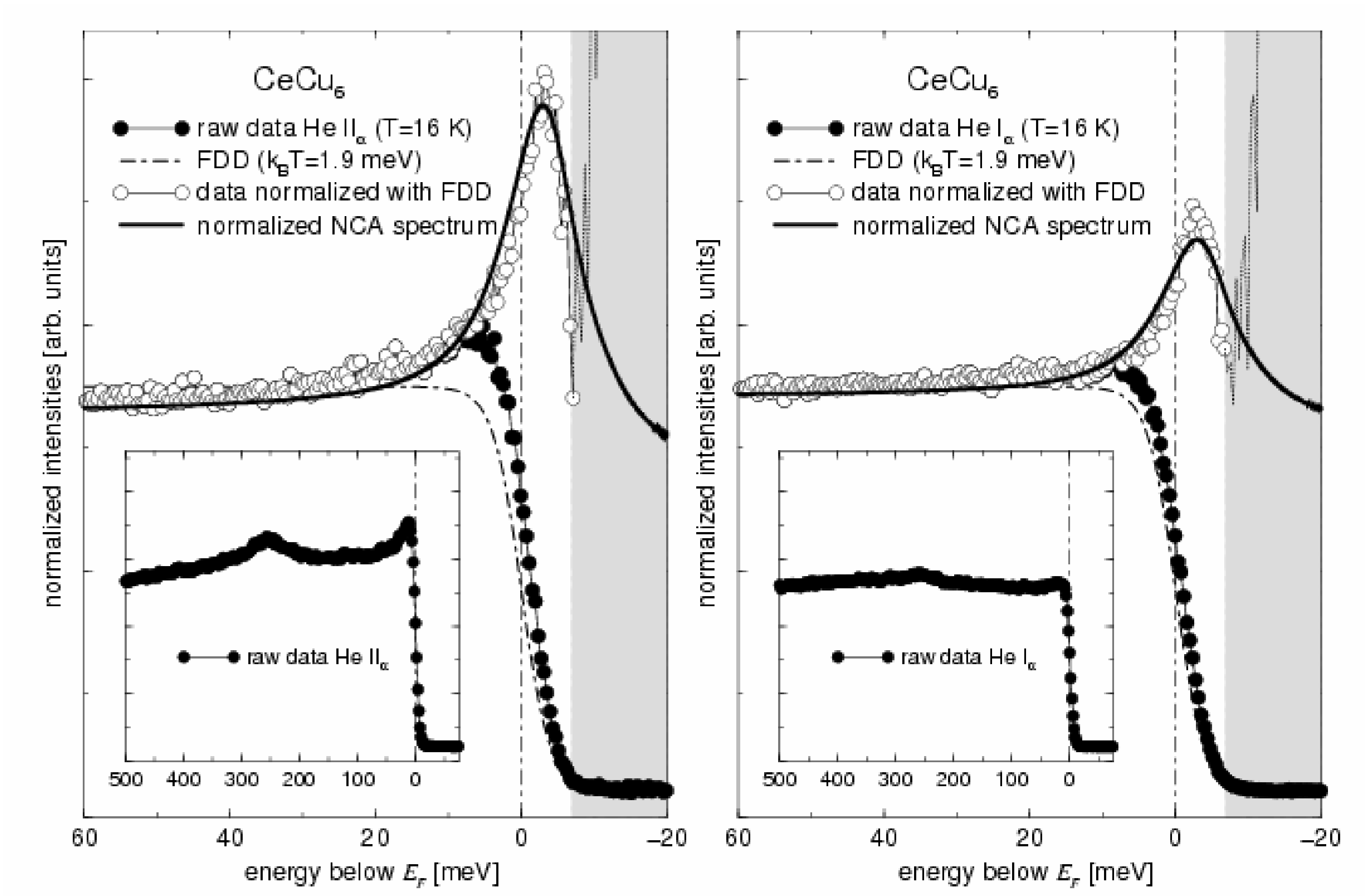
Results: Theoretical 4f spectrum in NCA



Results: Temperature dependence of 4f spectrum

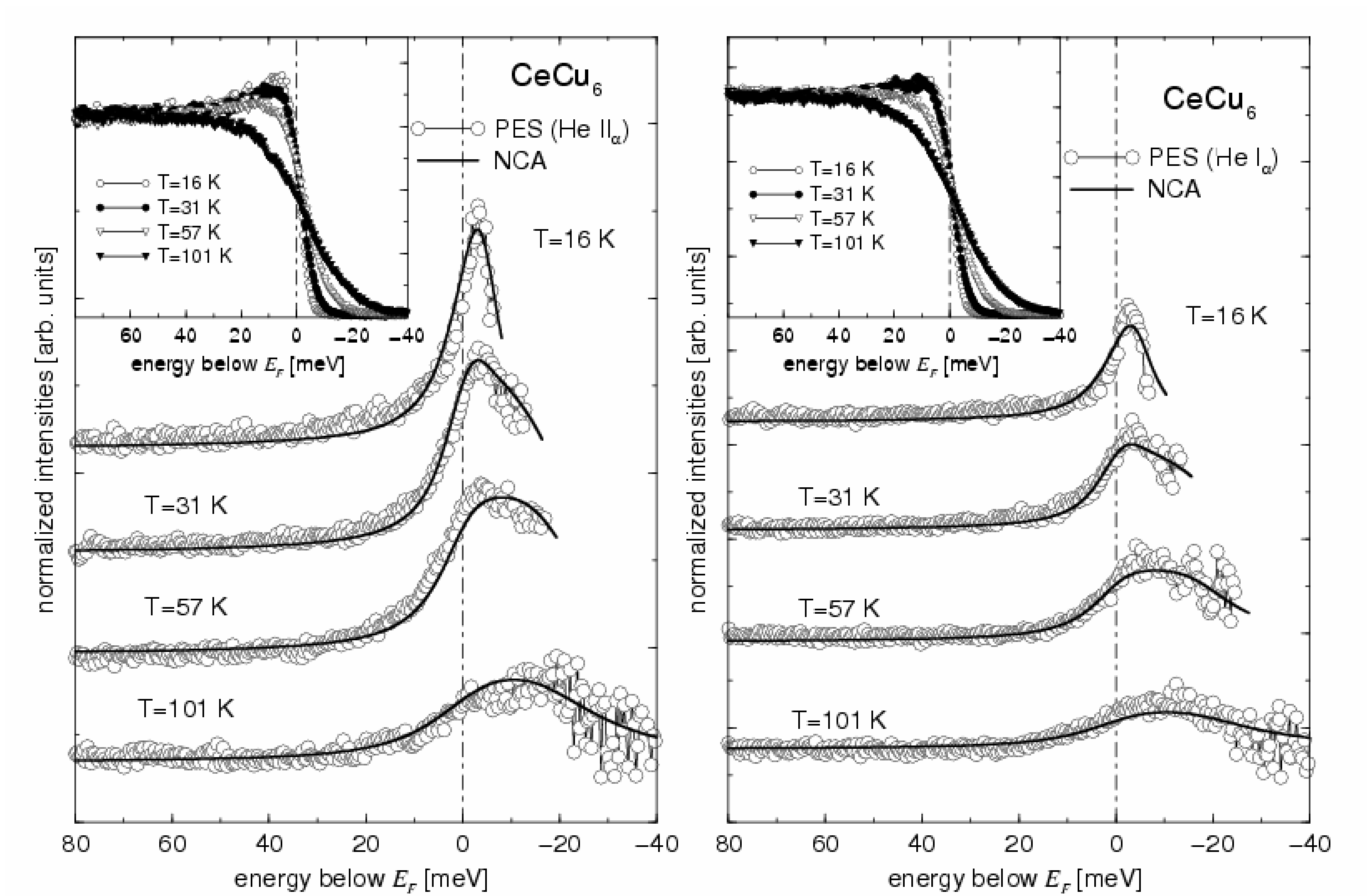


Results: PES of $CeCu_6$

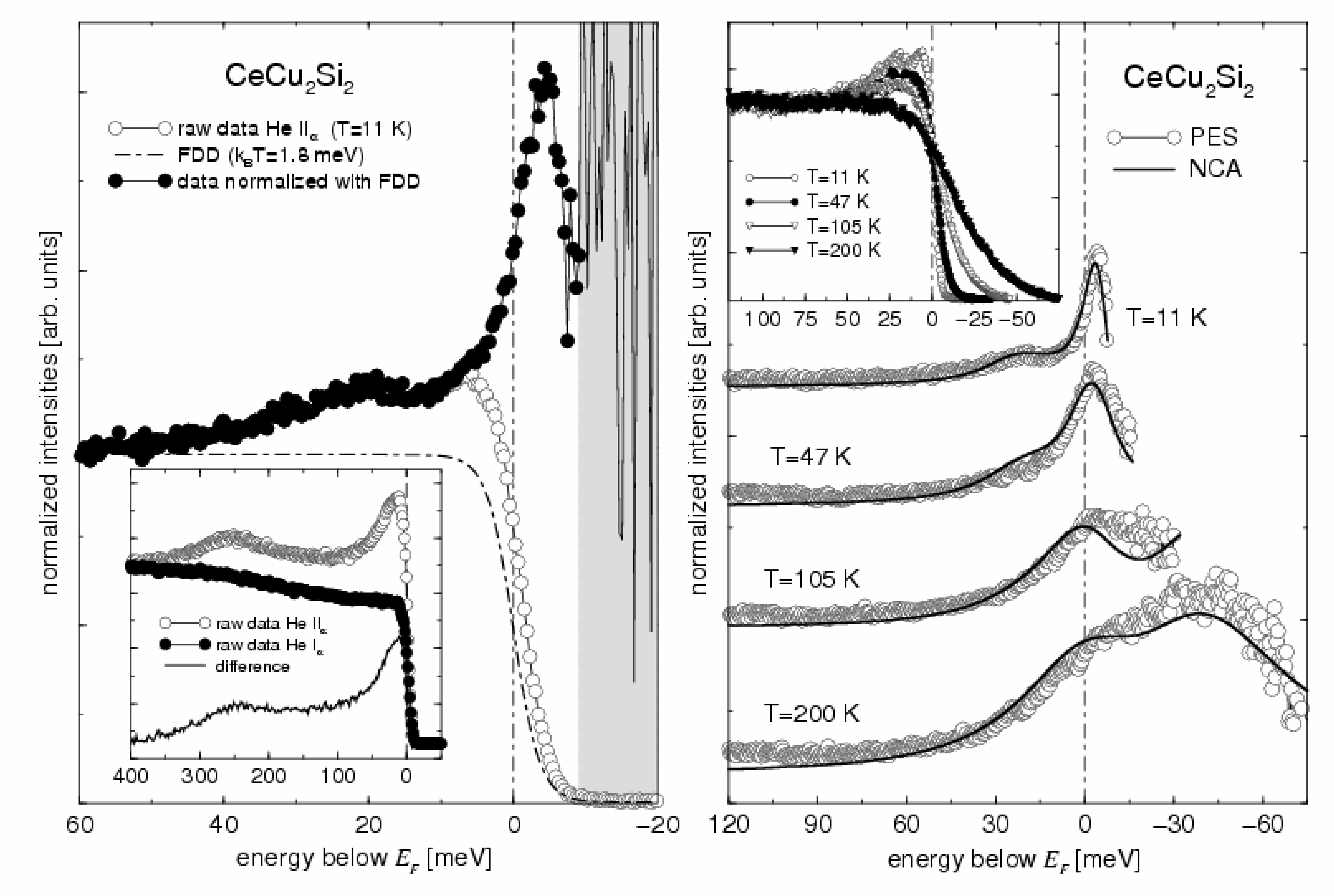


D. Ehm et al. (2002)

Results: Temperature dependence of PES of $CeCu_6$

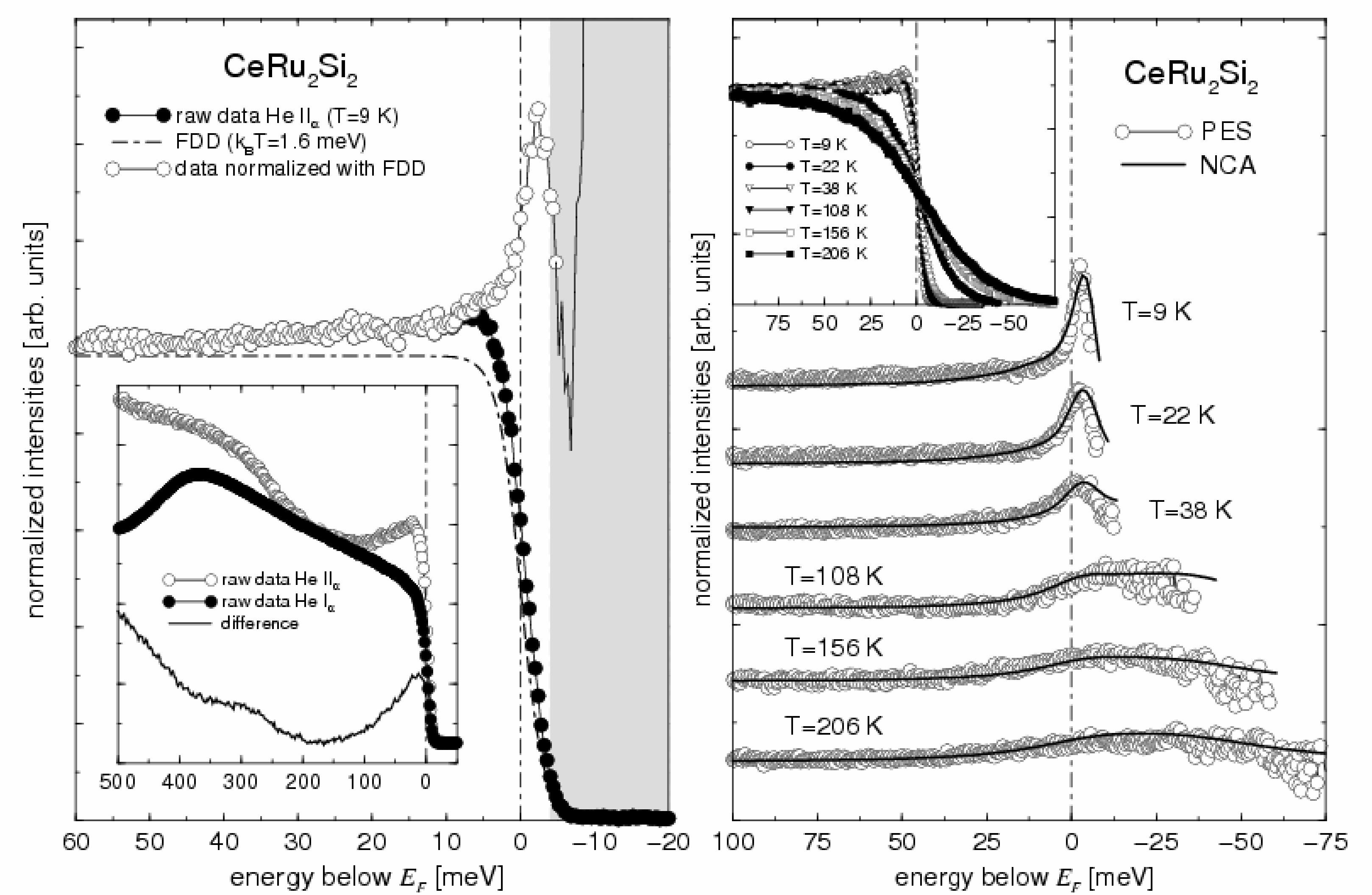


Results: PES of $CeCu_2Si_2$

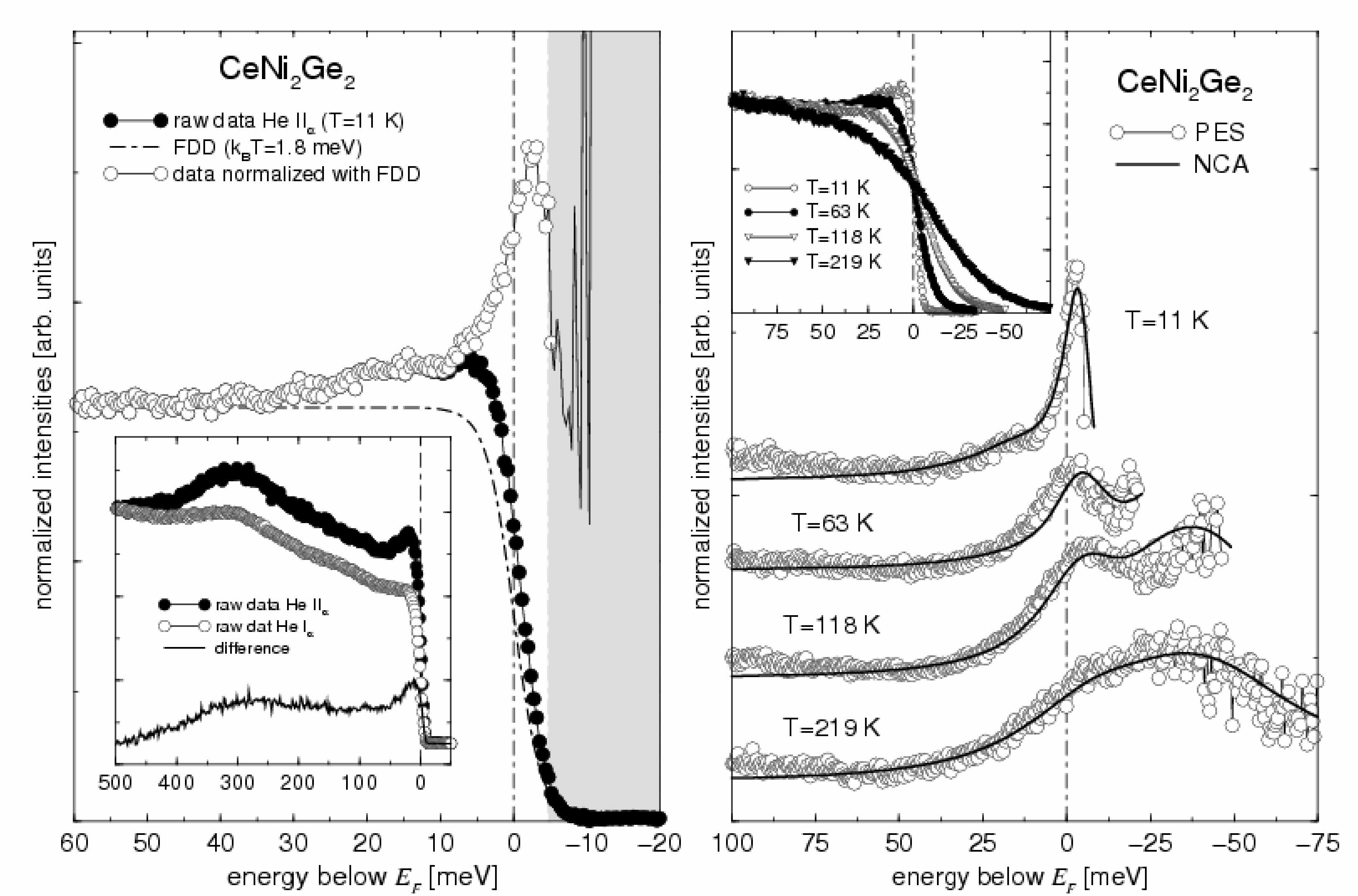


F. Reinert et al.(2001)

Results: PES of $CeRu_2Si_2$



Results: PES of $CeNi_2Ge_2$



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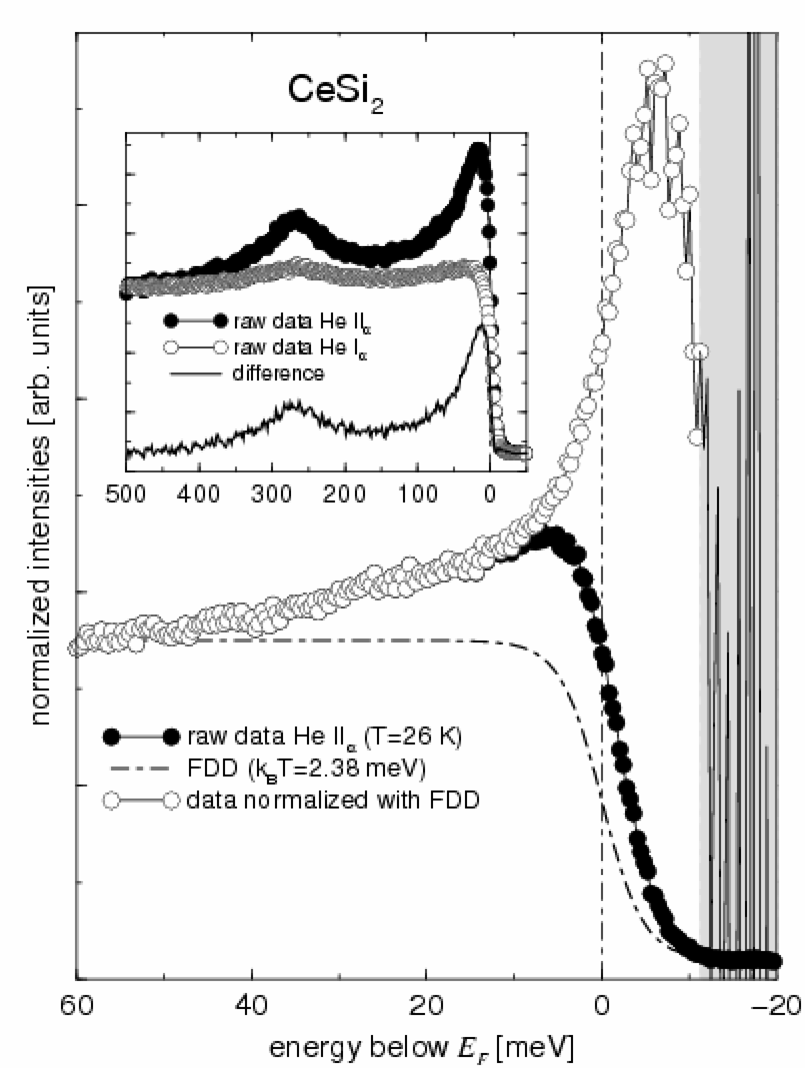
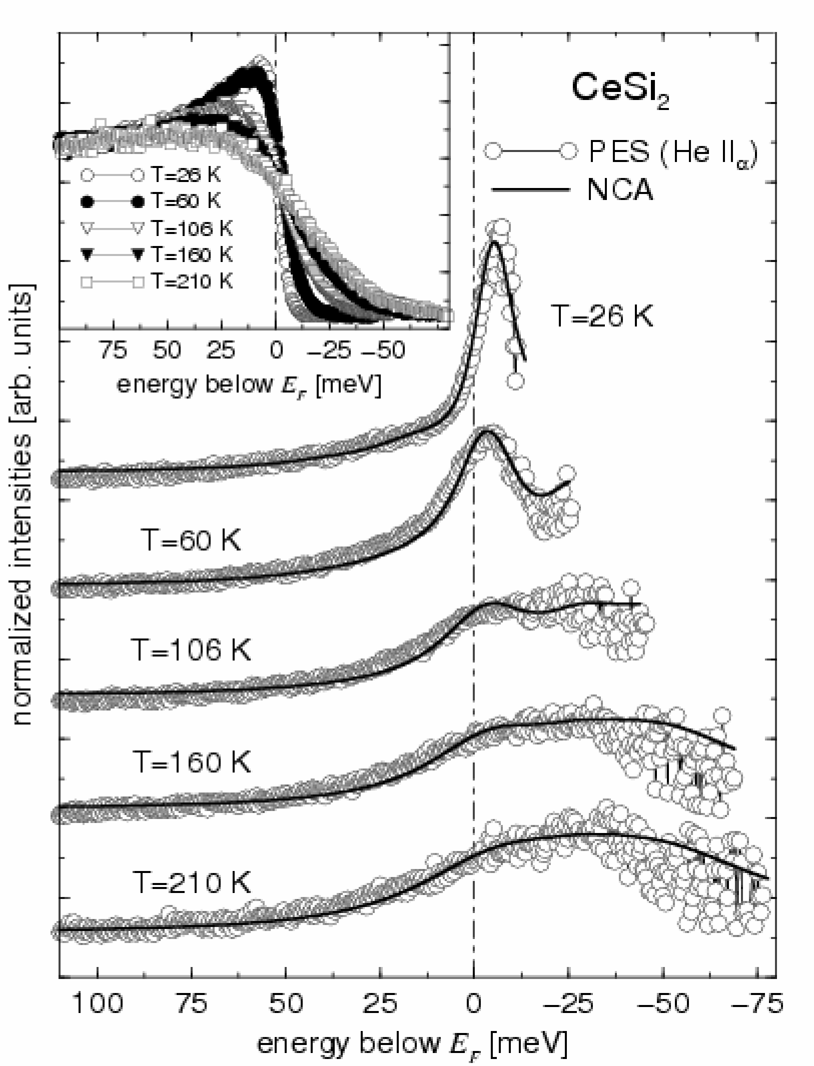
D. Ehm et al.(2005)

Results: PES of $CeSi_2$





D. Ehm et al. (2005)

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Results: Kondo temperature and crystal field splittings

	from other experiments			from PES		
	T_K [K]	CEF [meV]		T_K [K]	CEF [meV]	
		$\Delta_{1\to 2}$	$\Delta_{1\to 3}$		$\Delta_{1\to 2}$	$\Delta_{1\to 3}$
$CeCu_6$	$5.0 \pm 0.5^{(a)}$[39]	$7.0^{(a)}$[40]	$13.8^{(a)}$[40]	4.6	7.2	13.9
$CeCu_2Si_2$	$4.5^{(b)}$[41] – $10^{(a)}$[42]	$30^{(a)}$[40] – $36^{(c)}$[43]	—	6	32	37
$CeRu_2Si_2$	$16^{(a)}$[44]	$19^{(d)}$[45]	$34^{(d)}$[45]	16.5	18	33
$CeNi_2Ge_2$	$29^{(a)}$[23]	$(4)^{(a)}$[46]	$34^{(a)}$[46]	29.5	26	39
$CeSi_2$	$22/41^{(a)}$[47]	$25^{(a)}$[47]	$48^{(a)}$[47]	35	25	48

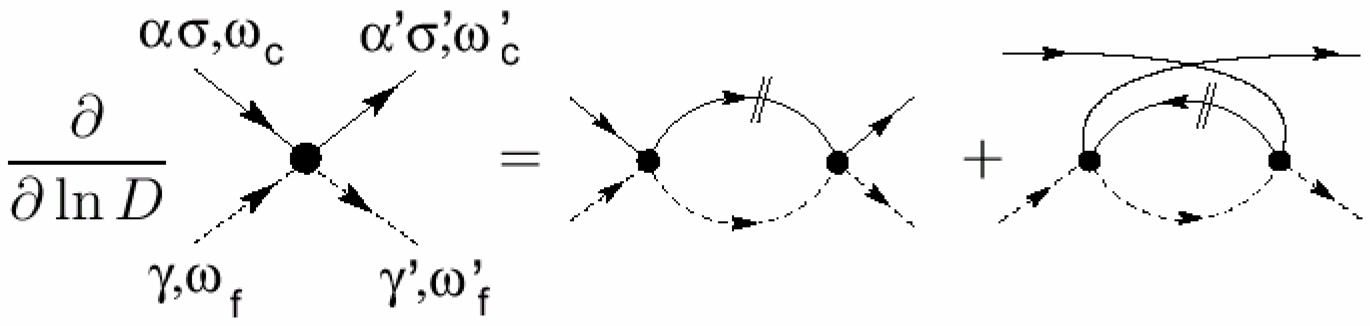
TABLE I: Comparison of the Kondo temperatures T_K and the CF energies Δ_{CF} determined from PES and from other experimental methods, namely from (a): INS studies, (b): specific heat measurements, (c): Raman scattering experiments, and (d): theoretical considerations based on specific heat measurements.

D. Ehm et al.(2005)

Conclusion

- Photoemission spectra of Ce compounds may be modelled within single ion Anderson model of spin-orbit and crystal field split ionic states
- Excited crystal field split 4f-states lead to Kondo-type satellite resonance peaks in the single particle spectral function
- The peak positions are given by a rigid shift of the multiplet up to the Fermi level, as obtained by slave boson mean field theory
- The Kondo character of the peaks is apparent from logarithmically divergent terms in perturbation theory
- Summing the leading logarithms by renormalization group methods, observing the effect of phase decoherence for excited states yields resonance peaks of width increasing with level splitting
- Quantitative results were obtained within NCA, in excellent agreement with experiment

Methods: Diagrams of one loop RG equation



Diagrammatic form of the RG equation . The strokes symbolize derivatives with respect to $\ln D$ and $\alpha, \alpha' = L/R$, $\sigma, \sigma' = \uparrow/\downarrow$ and $\gamma, \gamma' = \uparrow/\downarrow$ denote the quantum numbers of the incoming and outgoing conduction electrons and pseudo fermions, respectively. Their frequencies are given by $\omega_c, \omega'_c, \omega_f, \omega'_f$ with $\omega_c + \omega_f = \omega'_c + \omega'_f$.

Main contribution from Keldysh comp. of conduction electron G and real part of pseudofermion G:

$$\frac{\partial}{\partial \ln D} \int_{-D}^{D} d\omega \frac{\text{sign}\ \omega}{\omega - \Delta\omega} \approx 2\Theta(D - |\Delta\omega|)$$